



# Hydrogen production, storage, utilisation and environmental impacts: a review

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## Abstract

Dihydrogen (H<sub>2</sub>), commonly named ‘hydrogen’, is increasingly recognised as a clean and reliable energy vector for decarbonisation and defossilisation by various sectors. The global hydrogen demand is projected to increase from 70 million tonnes in 2019 to 120 million tonnes by 2024. Hydrogen development should also meet the seventh goal of ‘affordable and clean energy’ of the United Nations. Here we review hydrogen production and life cycle analysis, hydrogen geological storage and hydrogen utilisation. Hydrogen is produced by water electrolysis, steam methane reforming, methane pyrolysis and coal gasification. We compare the environmental impact of hydrogen production routes by life cycle analysis. Hydrogen is used in power systems, transportation, hydrocarbon and ammonia production, and metallurgical industries. Overall, combining electrolysis-generated hydrogen with hydrogen storage in underground porous media such as geological reservoirs and salt caverns is well suited for shifting excess off-peak energy to meet dispatchable on-peak demand.

**Keywords** Hydrogen production · Life cycle assessment · Hydrogen utilisation · Hydrogen storage · Climate change

## Abbreviations

CCSU	Carbon capture storage and utilisation
Eq	Equivalent
LCA	Life cycle assessment
LCOH	Levelised cost of hydrogen
WGSR	Water–gas shift reaction
USHS	Underground Seasonal Hydrogen Storage
PEMFCs	Proton exchange membrane fuel cells
PAFCs	Phosphoric acid fuel cells
SOFCs	Solid oxide fuel cells
MCFCs	Molten carbonate fuel cells
DMFCs	Direct methanol fuel cells

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## Introduction

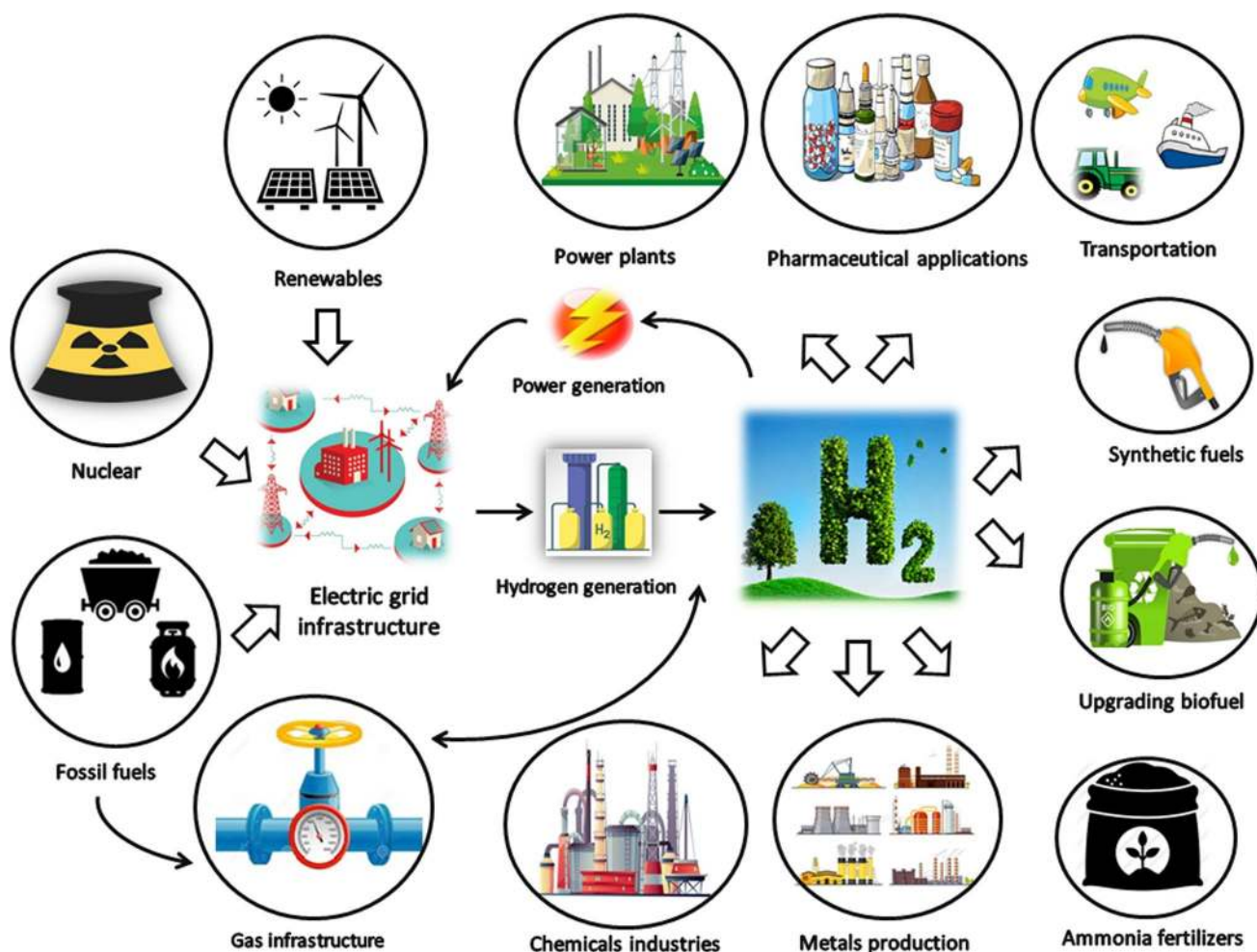
The continual growth and rapid urbanisation of the world population and economy have resulted in an enormous increase in energy need, urging the switch from fossil-based fuels into alternative clean renewables (Dawood et al. 2020). Consequently, global decarbonisation in the transportation, industry and electricity generation sectors is crucially needed to mitigate anthropogenic climate change (Fawzy et al. 2020; Osman et al. 2021a). In this context, there has been a growing interest from scholars

and industries with versatile production routes. There is abundant availability of renewable sources used in hydrogen production; however, the variable and intermittent nature of these resources is the major challenge in the transition towards a hydrogen economy. Hence, this calls for technical accommodation, especially for balancing variable renewable supply, i.e. solar, wind and others, and varying energy demand. Furthermore, cost-effective production methods, policies, research and development and hydrogen infrastructure development are areas that need more investigation when transitioning towards the hydrogen economy.

More than 100 current and planned hydrogen production technologies are reported to date, with over 80% of those technologies are focused on the steam conversion of fossil fuels and 70% of them are based on natural gas steam reforming. However, in order to minimise carbon footprint emissions, a wider range of hydrogen extraction processes, such as methane pyrolysis and seawater electrolysis using

alternative energy sources, must be addressed. All hydrogen production routes are highlighted in Fig. 1.

Hydrogen is the most abundant element in the universe, and due to its reactivity, it only exists on earth in compounds such as water and organic materials. It is an odourless, flammable and colourless gas, which is leading to its safety concern, especially if a leak is not detected and gas collects in a confined area; it can ultimately ignite and causes explosions. Furthermore, metal hydrogen embrittlement is an issue as it could damage pipelines and containers due to its small molecular size; thus, it escapes through materials. The higher heating value (HHV) of hydrogen is 141.8 MJ/kg at 298 K, and the lower heating value is 120 MJ/kg at the same temperature. This is significantly higher than that of most fuels such as gasoline with a value of 44 MJ/kg at 298 K. However, liquid hydrogen has a lower energy density by volume than hydrocarbon fuels such as gasoline by a factor of four with a density of 8 MJ/l versus density of 32 MJ/l. While hydrogen gas has a high energy density by weight but



**Fig. 1** Hydrogen production routes, including renewables, fossil fuels and nuclear, with hydrogen being produced in power plants, pharmaceutical applications, synthetic fuels or their upgrades in transportation, ammonia synthesis, metal production or chemical industry applications

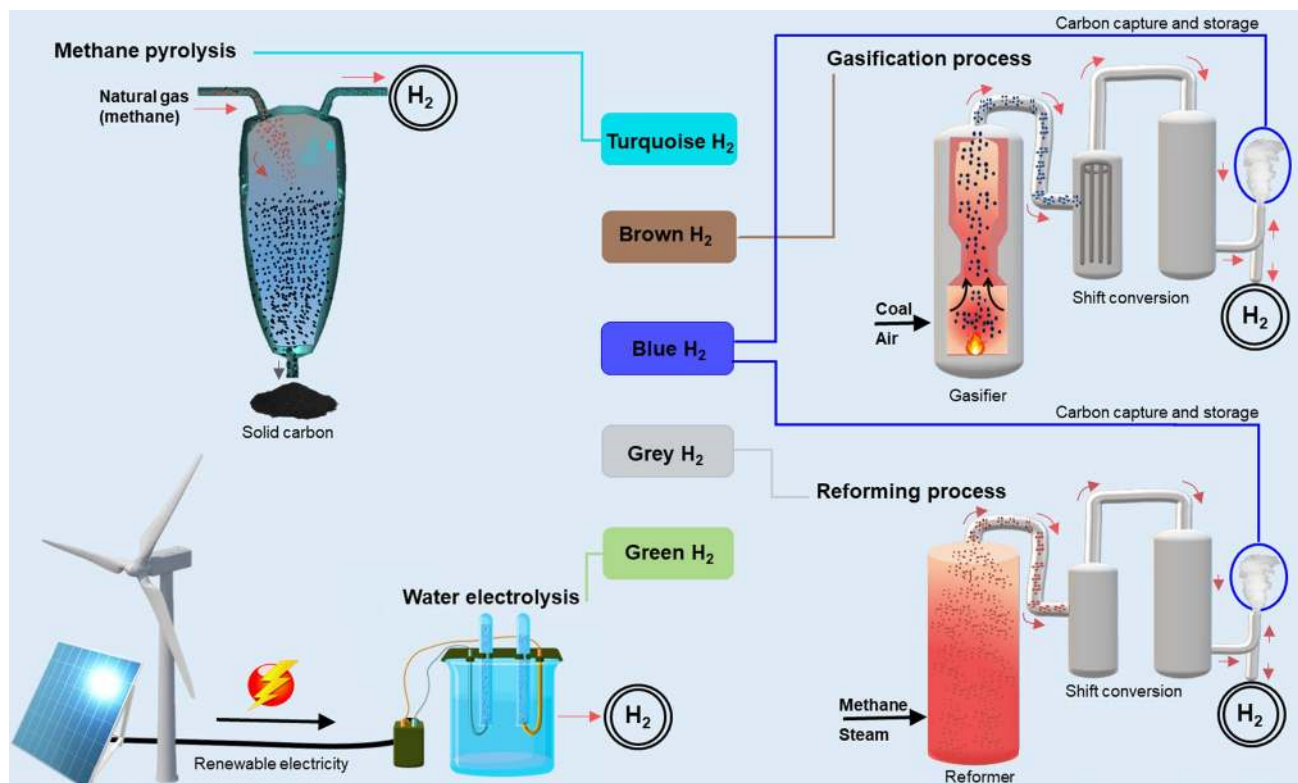
a low energy density by volume compared to hydrocarbons, it requires a larger tank to store. For example, as opposed to liquified natural gas, liquified hydrogen contains 2.4 times the energy but takes 2.8 times the volume to store. At the same time, the low temperature for liquified hydrogen storage at ambient pressure and a temperature of  $-253\text{ }^{\circ}\text{C}$  raises quite a few risks. When exposed, it can cause cold burns; furthermore, leakage can result in a combination of liquefied air and hydrogen, resulting in an explosive mixture or the formation of flammable or explosive conduits (Atilhan et al. 2021; El-Halwagi et al. 2020).

Like electricity, hydrogen is an energy carrier and not an energy source; using it to store renewable energies instead of being wasted when not in use is crucial since it is storable, utilisable and transportable (Parra et al. 2019; Abe et al. 2019).

## Hydrogen cleanness and colour coding

Dawood et al. (Dawood et al. 2020) reported the four main stages in hydrogen economy: production, storage, safety and utilisation, where hydrogen purification and compression

(subsystems) need to be considered along with the life cycle assessment (LCA) when selecting the production method for hydrogen. Hydrogen cleanness level is described in the literature with many colour coding: mainly green, blue and grey, which relies only on the production route, i.e. hydrogen origin, and fails to assess the deep cleanness of the produced hydrogen (Merzian and Bridges 2019), for instance: (1) Grey hydrogen is produced using fossil fuels such as natural gas, one tonne of hydrogen produced in this way is responsible for 10 tonnes of carbon dioxide (Dvoynikov et al. 2021), as shown in Fig. 2; (2) blue hydrogen is produced from fossil fuels like grey hydrogen but with combination of carbon capture and storage to mitigate emissions; (3) green hydrogen is typically produced from 100% renewable sources such as wind or solar energies with lower carbon footprint; (4) brown hydrogen is produced from gasification of coal-based fuel; and (5) turquoise hydrogen is produced from the thermal decomposition of natural gas, i.e. methane pyrolysis or cracking by spitting methane into hydrogen and carbon at a temperature range from 600 to 1200–1400  $^{\circ}\text{C}$  (Dvoynikov et al. 2021). This process produces black carbon (soot) as a by-product instead of carbon oxide emissions in the grey hydrogen, allowing for the sequestration of carbon emissions



**Fig. 2** Hydrogen colour coding for various manufacturing processes. Green hydrogen is produced using renewable energy sources such as solar or wind energy, followed by water electrolysis. Grey and brown hydrogen are produced by methane steam reforming and coal gasi-

fication, respectively, and when combined with carbon capture and storage, blue hydrogen is produced. Turquoise hydrogen is produced through the pyrolysis of methane, with solid carbon as a by-product

in the form of solid carbon. However, carbon stability in this black soot is critical for long-term carbon sequestration, along with the utilisation of renewable energy sources in the high-temperature process to achieve carbon neutrality. Interestingly, hydrogen could be produced with a negative carbon footprint via biogas pyrolysis.

However, this colour coding is not precise as it assumes that green hydrogen always has low-carbon emission than blue or grey hydrogen, which is not applicable in all cases. Blue hydrogen, for example, is regarded as less safe than green hydrogen, even though it releases no carbon at the point of use or during the entire process, while green hydrogen may do. For instance, bioenergy feedstocks such as biomass emit greenhouse gas emissions such as CH<sub>4</sub>, SO<sub>x</sub>, NO<sub>x</sub> and CO<sub>2</sub> during their growth or thermochemical conversions. Furthermore, the carbon capture and storage technique used in the blue hydrogen reduces toxic emissions significantly. The manufacture of photovoltaic panels as renewable energy technology also has a significant carbon footprint and generates various types of waste, liquid and gaseous by-products that are hazardous to the environment. Starting from the extraction of quartz and other materials used to manufacture solar panels, this is coupled with the carbon and sulphur emission in the energy-intensive process when producing metallurgical silicon. Moreover, the solar panel has a 30-year lifespan, and then, it must be handled as a particular waste at its end of life.

A recent LCA study compared environmental impacts for steam methane reforming with water electrolysis using wind, solar photovoltaic, hydropower, solar thermal and biomass gasification as energy sources (Al-Qahtani et al. 2021). It was concluded that among all the technologies evaluated, solar photovoltaic electrolysis had the most damaging environmental implications because of the significant acidification potential in the photovoltaic panel production phase and the relatively poor efficiency of photovoltaic systems.

Thus, measuring the emitted greenhouse gas emissions accurately in the entire production process along with the life cycle of the equipment used is crucial. This is required to determine how green is the green hydrogen and how blue is the blue hydrogen. A recently proposed model for improved hydrogen colour coding consisted of a hydrogen cleanness index followed by the number of depth levels (Han et al. 2021). For instance, 80 green-4 means hydrogen is produced via renewable resources; however, it is not a zero-emission process, only 80% green, due to emissions related to the process. The number after the colour, which in this case is 4, indicates that greenhouse gas emissions (CO<sub>2-e</sub>) linked with the purification during the production route have been considered. This model still requires much more analysis to decide the start and end of the continuum thresholds for each colour, as well as the evaluation depth levels and related weight for each level.

## Hydrogen production routes

According to the International Energy Agency (IEA), green hydrogen could help reduce our carbon footprint if major challenges such as infrastructure, logistics, cost-effective manufacturing methods and safety are overcome. Globally, hydrogen is responsible for about 843 metric tonnes of CO<sub>2</sub> emissions per annum, equal to the combined total emissions of the UK and Indonesia (IEA 2019). The global hydrogen demand is projected to increase from 70 million tonnes in 2019 to 120 million tonnes by 2024 (Global hydrogen market insights 2020; Atilhan et al. 2021; Safari and Dincer 2020). In 2025, the largest global green hydrogen plant will be built, with a capacity of 237,250 tonnes per annum, i.e. 650 tonnes/day hydrogen output through electrolysis and 4 gigawatts of renewable energy from wind, solar and storage.

A wide range of resources is available for hydrogen production, mainly fossil-based and renewable fuels (Dawood et al. 2020; Saithong et al. 2019; Osman et al. 2020a). The former is the more mature and most common used industrially as it is a cost-effective method that deploys cracking or reforming fossil-based fuels. In 2016, hydrogen production globally was about 85 million tonnes used in petroleum, metal industry, fertiliser, food processing, semiconductor production, power plants and generations (Chen and Hsu 2019; El-Emam and Özcan 2019; Acar and Dincer 2019).

There are many ways to extract hydrogen from hydrogen-containing materials, either hydrocarbon or non-hydrocarbon, such as photonic, electric, chemical, bioenergy, heat and a combination of those methods together (Abe et al. 2019; El-Emam and Özcan, 2019; Osman et al. 2020b). Table 1 shows different hydrogen production routes with different energy sources, technology readiness level (TRL) and their % energy efficiency.

## Advances and challenges in water electrolysis

Water is typically purified and then sent to an electrolyser, which produces hydrogen and oxygen. The hydrogen is then dried, purified and compressed from a 10.3 to 413.7 bar pressure, and then stored in a tank. Although the electrolysis pathway offers a 100% renewable route for hydrogen production, it represents less than 5% of worldwide hydrogen production (Han et al. 2021). Despite this low percentage contribution, water electrolysis is gaining momentum for various reasons such as zero-carbon emissions, the absence of unwanted by-products such as sulphates, carbon oxides and nitrogen oxides, and high hydrogen purity. The cost of producing hydrogen through

**Table 1** Hydrogen production routes with various energy feedstock sources, technology readiness level (TRL) and their % efficiency

Feedstocks	Production method	% Energy efficiency	TRL*	References
Water	Electrolysis (alkaline electrolysis)	61–82	9–10	Parra et al., 2019
	Electrolysis (photo-electrochemical)	0.5–12	1–2	El-Emam and Özcan, 2019
	Photolysis (photosynthesis)	< 5	1–3	Alanne and Cao, 2019
	Thermolysis (water thermolysis)	20–55	1–3	El-Shafie et al., 2019
	Chemical (redox reaction)	3–5 wt	4–6	Alanne and Cao, 2019
Water + biomass	Biolyis (aqueous phase reforming)	35–55	5–7	El-Shafie et al., 2019
Brine	Proton exchange membrane	67–84	7–9	Dawood et al., 2020
Biomass	Bioelectrolysis (microbial electrolysis)	70–80	1–3	El-Shafie et al., 2019
	Biothermolysis (co-fermentation hydrolysis)	35–45	1–3	Cheng et al., 2019
	Thermolysis (pyrolysis)	35–50	8–10	Dawood et al., 2020
	Thermolysis (gasification)	35–50	10	El-Emam and Özcan, 2019
	Thermolysis (partial oxidation)	60–75	7–9	El-Shafie et al., 2019
Coal	Biophotolysis (photofermentation)	< 1%	1–3	Dawood et al., 2020
	Thermolysis (gasification)	74–85	10	Mah et al., 2019
Microorganism	Biolyis (dark fermentation)	60–80	3–5	Mah et al., 2019
Microalgae	Biophotolysis (photofermentation)	< 1%	1–3	El-Shafie et al., 2019
Hydrocarbons	Partial oxidation process of fossil fuels	60–75	9	Pinsky et al., 2020
Natural gas	Steam methane reforming	74–85	9	Pinsky et al., 2020
Hydrocarbons	Partial oxidation of fossil fuels	60–75	9	Pinsky et al., 2020

\*TRL is referring to the technology readiness level

electrolysis would be reduced by approximately 70% over the next decade, allowing for the widespread adoption of a green hydrogen production approach.

By 2040, the worldwide market for hydrogen electrolyzers is expected to have grown by 1000-fold. Aurora Energy Research predicted that about 213.5 gigawatts of projects will be completed over the next 19 years; this compares to an estimated 200 megawatt that is currently in service. They reported that 85 per cent of anticipated projects are in Europe, with Germany accounting for 23 per cent of expected global electrolyser capacity. The European Union has already set a goal of 40 gigawatts of electrolyser capability by 2030 (Research, 2021). If all this power is available, it will supply up to 32 million tons of hydrogen per year, which is already half of the currently demanded hydrogen. In a 1.5-degree climate change mitigation scenario, meeting 24% of energy demand with hydrogen will necessitate massive amounts of additional renewable electricity generation. To power electrolyzers in this scenario, approximately 31,320 terawatt-hours of electricity would be required, i.e. more than is currently produced globally from all sources combined (BNEF 2020). Besides, an investment of more than \$11 trillion in manufacturing, storage and transportation infrastructure would be required.

Proton exchange membrane (PEM) along with alkaline anion exchange membrane (AEM) and concentrated potassium hydroxide solution KOH are the most common

techniques used in low-temperature water electrolysis. The key benefit of alkaline anion exchange membrane electrolysis over other methods is lower cost since no platinum group metals are used as catalysts herein. The main challenge, however, is the low rate of hydrogen production and the instability of the alkaline method owing to its susceptibility to pressure drop (Dvoynikov et al. 2021; Yu et al. 2019). A typical electrolysis system consists of two metal electrodes, an anode and a cathode, separated by a membrane and immersed in an electrolyte solution (Zhu et al. 2019). As an electric current flows through the solution, oxygen and hydrogen bubbles rise above the anode and cathode, respectively. Both electrodes are typically coated with a catalyst to reduce the amount of energy needed to liberate hydrogen from water.

However, large amounts of freshwater would be needed to generate hydrogen, and these supplies are already depleted worldwide; thus, the utilisation of seawater will be an option to overcome this issue. However, seawater utilisation in hydrogen production is associated with challenges such as the corrosion of chloride ions in seawater to the anode metal. Hung et al. reported a solution to this issue by designing the anode material as a porous nickel foam pan collector coated with an active and inexpensive nickel and iron catalyst, which showed strong conductivity and corrosion resistance. It is worth noting that, while using freshwater is more expensive than using seawater, the cost of water usually accounts

for less than 2% of the total cost of hydrogen production via electrolysis (Milani et al. 2020). The affordability and accessibility of freshwater is one side of the coin, while inexpensive and sustainable green energy alternatives are the other, and the proximity of these two supplies, i.e. renewable energy and freshwater, does not always coincide. The main areas that need further investigation in water electrolysis are reducing the capital cost of electrolysis technology, finding water resources and increasing efficiency.

According to the recent literature summarised in Table 1, membrane reactor technology is increasingly being recognised as an encouraging route to expand clean hydrogen production paths from hydrocarbons and hydrogen purification. At least 99.8% can be achieved without any gas purification using a proton exchange membrane analyser (Jorschick et al. 2021).

Recently, it was reported for Australia that the levelised cost of hydrogen (LCOH) for steam methane reforming could reach a cost of \$(1.88–2.30)/kg H<sub>2</sub> and \$(2.02–2.47)/kg H<sub>2</sub> for coal gasification production routes. In comparison, the LCOH via electrolysis technologies costs between \$4.78 and \$5.84/kg H<sub>2</sub> for alkaline electrolysis and \$6.08–7.43/kg H<sub>2</sub> for proton exchange membrane technologies (Milani et al. 2020).

When using partial methane oxidation for hydrogen production via synthesis gas, the average cost is 1.33 euros/kg H<sub>2</sub>, while the cost of large-scale H<sub>2</sub> processing ranges between 1 and 1.5 euro/kg H<sub>2</sub> (Dvoynikov et al. 2021). It is important to note that the economic viability of using natural gas or related petroleum gas for hydrogen production should be seen in the light of transportation systems or the direct use of hydrogen on-site of the gas or oil plant.

In terms of blue hydrogen, carbon capture and utilisation lower greenhouse gas emissions but raise the overall production cost. Chemical looping reforming, for instance, has a comparatively short life cycle, global warming potential and low fossil fuel intake. Nevertheless, adding carbon capture and liquefaction process units raises the expense of the steam methane reforming by 18% and autothermal reforming processes by 2% (Atilhan et al. 2021). The process of liquefying hydrogen absorbs approximately 30% of the energy content of hydrogen. Additionally, keeping liquified hydrogen under one atmospheric pressure and at a low temperature of –253 °C is difficult. Furthermore, evaporation and leakage can occur even with robust insulation, losing typically 1 per cent of the stored volume per day (Atilhan et al. 2021).

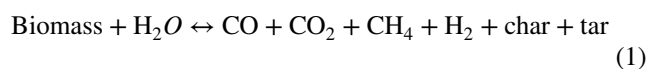
### Biomass gasification

Biomass gasification is seen as one of the most feasible, sustainable and potentially carbon-neutral alternatives to generate hydrogen (Saidi et al. 2020). Since biomass is a

renewable feedstock that absorbs atmospheric carbon dioxide during growth, it has a much lower net CO<sub>2</sub> footprint than fossil-based fuels. However, the economic feasibility of hydrogen output from biomass must be closely related to the availability and affordability of raw materials in the local area. The biomass physicochemical properties, distribution and hydrogen rate are the main attributes of the supply materials. Since biomass feedstocks vary widely in structural composition and shape, all of these characteristics must be taken into account when combining the feedstock with the appropriate conversion technology (Srivastava et al. 2020).

Consequently, moisture, energy and ash contents are the core criteria for evaluating biomass utilisation in this route. The hydrogen yield from biomass is comparatively poor since the hydrogen content of biomass is roughly 5.9 wt% compared to 25 wt% for methane (natural gas), and the energy content is also low due to high oxygen content within the biomass of 40%. Thus, techno-economic studies backed by adequate life cycle assessment evaluation are crucial in this matter. Since biomass has a lower density, transportation and storage costs for either biomass feedstock or the produced hydrogen should be well justified in terms of economies of scale. In certain ways, these characteristics would make it impossible for biomass-based hydrogen production to compete with common natural gas such as steam methane reforming method unless new regulatory frameworks such as carbon tax favour competitively sustainable hydrogen production routes.

Biomass gasification, like coal, is the most practical process for biomass feedstocks because it produces the best yield at high temperatures, generally, 500–1400 °C, where the overall reaction is presented in Eq. 1. Interestingly, the integration between biomass gasification and carbon capture and storage can potentially lead to an overall negative carbon footprint.



### Advances and challenges in fossil-based hydrogen production route

The breakdown of the long-chain hydrocarbon via gasification, reforming or pyrolysis reaction routes is required for hydrogen production from fossil-based feedstocks. The primary product in the reforming reaction is the synthesis gas (a mixture of H<sub>2</sub> and CO), followed by H<sub>2</sub> separation via autothermal reforming, steam methane reforming, partial oxidation or membrane reforming. Another well-known method that is commonly used in hydrogen production is the gasification of fossil fuels, such as coal gasification (Milani et al. 2020).

Al-Qahtani et al. evaluated and compared the most common hydrogen generation routes on a monetary basis, such as steam methane reforming, coal or biomass gasification, methane pyrolysis with or without carbon capture and storage technology. Besides, the hydrogen production from the water via electrolysis derived from solar or nuclear energy were also assessed. They reported that, at the moment, steam methane reforming with carbon capture and storage appeared to be the most viable alternative (Al-Qahtani et al. 2021).

### Steam methane reforming and methane pyrolysis

The primary feedstock for steam methane reforming is natural gas, predominantly methane mixed with other hydrocarbons and carbon dioxide (Osman 2020). Natural gas and steam reaction occur in a two-step reaction, as shown in Eq. 2 at high temperatures, followed by an interaction between the carbon monoxide and the produced hydrogen along with the unreacted natural gas. Following that, more steam is supplied to react with carbon monoxide in a water–gas shift reaction (WGSR), as shown in Eq. 3, to recover further hydrogen and convert carbon monoxide into carbon dioxide. The entire process efficiency is around 76% (Al-Qahtani et al. 2021). The entire process releases a significant amount of carbon dioxide emissions, which may be decreased by installing carbon capture and storage technology, removing and separating the flue gases from the product stream. Following that, an amine solvent such as monoethanolamine absorbs about 90% of the carbon dioxide emission, and then, the processed flue gas stream is released into the environment. Afterwards, carbon dioxide is thermally desorbed and compressed to 110 bars for storage. The integration between steam methane reforming and carbon capture and storage (SMR + CCS) technologies has an energy efficiency of 68 per cent, owing mostly to the energy necessary to regenerate the monoethanolamine and the power required for compression. After the WGSR, hydrogen is further purified to 99.99 per cent in both situations, with or without carbon capture and storage, in a pressure swing adsorption unit, which is also utilised in the gasification technology such as coal or biomass gasification routes.



Regarding methane pyrolysis at high temperatures, thermally or catalytically, the processes degrade hydrocarbons into hydrogen and solid carbon, as shown in Eq. 4. Because there is no oxygen in the process, no carbon oxides are generated, possibly removing the requirement for subsequent processing stages such as the WGSR and lowering

the capital and operating expenditures compared to steam methane reforming (Al-Qahtani et al. 2021). The greater  $\text{H}_2$  content in the product gas stream has the potential to reduce downstream clean-up operations significantly. The cost of methane pyrolysis is heavily influenced by the natural gas prices, processing method and solid carbon by-product.



### Coal gasification

During the coal gasification process at high temperatures ranging from 800 to 1300 °C and 30–70 bar pressures, coal is partially oxidised in oxygen or air atmosphere into synthesis gas, as shown in Eq. 5. The synthesis gas is typically composed of carbon monoxide and dioxide, hydrogen and unreacted methane, where the WGSR process (Eq. 3) enriches the syngas further to recover additional hydrogen. Thus, combining Eqs. 3 and 5 will lead to the overall reaction as in Eq. 6. Coal gasification is less efficient than steam methane reforming with 55%, although it has a larger single-train capacity.



### Bibliometric analysis

Key research studies were identified to summarise state of the art and discover knowledge gaps in the hydrogen production and LCA research arenas. The advanced search tool for publications from the Web of Science was used for this study, using the terms ‘Hydrogen production’ AND ‘Life cycle assessment’ as inputs. The results were manually scanned, and 24 most complete and relevant studies published from 2019 to 2021 were selected for review in the present study.

### Life cycle assessment

Life cycle assessment (LCA) is recognised as a comprehensive tool to evaluate environmental impacts associated with products and processes. There are many hydrogen production methods, such as steam methane reforming, electrochemical routes through water electrolysis using renewable power sources, thermochemical pathways involving renewable feedstock as the hydrogen carrier and biological processes (Valente et al. 2021; Owgi et al. 2021). However, environmental sustainability based on LCA remains one of the key requirements for selecting these processes for

hydrogen production (Falcone et al. 2021). This is because policymakers need to adopt transformative solutions based on robust data and evidence-based research to identify processes that go beyond a one-fits-all approach.

To this end, we reviewed 24 LCA studies published from 2019 to 2021 on hydrogen production and life cycle assessment (Table 2). The four main stages defined by ISO 14040 and ISO 14,044 for conducting LCA are: (1) goal and scope definition, (2) life cycle inventory analysis, (3) environmental impacts assessment and (4) life cycle interpretation (Al-Muhtaseb et al. 2021).

### Goal and scope of the life cycle assessment

The first stage of LCA consists of defining a goal and the scope of the study. This stage determines whether a study would be attributional or consequential, what functional unit will be considered to evaluate environmental impacts and the extent of the system boundary. This is an important initial step as the questions to be answered determine the results and associated policy implications.

#### Types of life cycle assessment: attributional and consequential

Life cycle assessment studies can be broadly classified into two categories: (1) Attributional LCA incorporates immediate physical flows such as raw materials, energy and emissions involved across the life cycle of a product (Jeswani et al. 2020), and (2) consequential LCA accounts for how physical flows can change as a consequence of an increase or decrease in demand for the product system under study (Earles and Halog 2011). It includes unit processes inside and outside the product's immediate system boundaries; therefore, consequential LCA studies are more suited for policy decisions. However, as LCA for hydrogen production remains at an embryonic stage, attributional studies are more commonly found. Nevertheless, both attributional and consequential approaches were considered for the purpose of this study.

#### Functional unit

In LCA, the functional unit is a measure of the purpose of the studied system, and it provides a reference by which the inputs and outputs can be related. This enables the comparison of two essentially different systems. The definition of the functional unit is intricately linked to the goal of an LCA study. It was observed that ~42% of the reviewed studies used 'kg of hydrogen produced' as the functional unit (Fig. 3). While some studies provided results considering hydrogen as an energy carrier and therefore recorded functional unit as 'energy produced in MJ or kWh'. Very few

studies reported 'distance travelled in km' as a functional unit when hydrogen was utilised as fuel for vehicles. The choice of different functional units for the same product, i.e. hydrogen, shows the challenges associated with comparing LCA models.

#### System boundary

In LCA, the system boundary definition profoundly impacts the materials, processes and emissions considered for evaluation. As such, system boundary limits can also considerably influence the calculation of environmental impacts (Collotta et al. 2019). The two commonly studied kinds of system boundary for hydrogen production are 'cradle-to-gate' or 'well-to-pump' that includes processes only until production and 'cradle-to-grave' or 'well-to-wheel', which incorporates emissions during end use as well.

The generalised system boundary used for conducting the LCA of hydrogen production and consumption includes: (1) raw materials and primary energy sources such as natural gas, coal, biomass, nuclear energy and water; (2) the hydrogen production processes, for instance, water electrolysis and thermochemical processes. Some processes may also consider hydrogen purification as a subsystem to the production; (3) storage of hydrogen in underground caves or compressed tanks; (4) transportation of hydrogen in liquified or compressed gaseous form using trucks and tube trailers or pipelines; (5) emissions during end use such as by hydrogen trains or generation of power using hydrogen; and (6) finally, waste treatment processes from these systems such as emissions to land, air and water (Fig. 4).

During the review, we observed that studies employed an array of processes and limits in system boundary for conducting LCA of hydrogen production and consumption (Table 3). There were only a handful of studies that considered emissions during the use phase. However, given the increasing interest in using hydrogen as a clean energy carrier, it is important to consider the emissions during the use phase and conduct LCAs that present 'well-to-wheel' estimates.

#### Allocation approaches

The allocation approach refers to both 'partitioning' and system expansion/substitution method. The allocation approach has been identified to significantly control the values obtained for environmental impacts (Finnveden et al. 2009). Allocation approaches are required because the life cycle of a product can consist of many multifunctional processes. Therefore, it is imperative to allocate the environmental impacts between the different coproducts generated by the same process in a justified manner.



**Table 2** Life cycle assessment tools and databases used in the reviewed studies

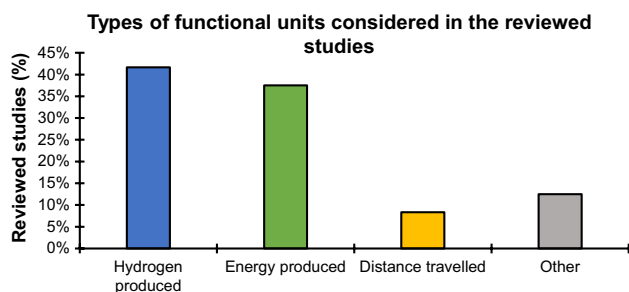
Reference	Region	Functional unit	Feedstock	Process	Product use	LCA tools	Database
Alviani et al., 2021	Japan	1 MJ of energy produced	Aluminium waste materials and water from hot springs	Hydrothermal reaction	Hydrogen for fuel cell vehicle	n/i	n/i
Booto et al., 2021	Norway	1 km of distance travelled	Water and electricity	Steam methane reforming and electrolysis	Comparison of: hydrogen for trucks (fuel cell electric truck), conventional trucks, battery electric truck	OpenLCA 1.7	Ecoinvent 3.4 and International Life Cycle Data System
Bui et al., 2021	Great Britain	Energy produced	Biomass	Comparison of biomass conversion pathways: pulverised biomass for electricity, biomass for combined heat and power, biomass gasification and water–gas shift technology for hydrogen	Biohydrogen for energy production	Not included	Not included
Cvetković et al., 2021	Serbia	Energy produced	Cattle manure and corn silage	Anaerobic digestion followed by steam reforming	Biohydrogen for energy production	Not included	Not included
Desantes et al., 2020	Europe	Life of each vehicle considering 150,000 km as the common life	Water and electricity	Electrolysis and steam methane reforming	Hydrogen for mid-size passenger vehicles	Not included	Greenhouse gases, Regulated Emissions and Energy use in Transportation 2019
Kerscher et al., 2021	Germany	1 kg hydrogen	Natural gas	Electron beam plasma methane pyrolysis	Hydrogen	GaBi 9.2	GaBi and energy extension databases
Kim et al., 2021	Republic of Korea	1 kWh of electricity	Natural gas	Molten carbonate fuel cell system	Hydrogen for heat and electricity	Not included	Expert communications
Li et al., 2021	China	1 kg hydrogen	Comparison of: natural gas, water, water via Cu-Cl, coal	Comparison of: natural gas steam reforming, water electrolysis via wind power, thermochemical water splitting via Cu-Cl cycle, coal gasification	Hydrogen	Not included	Not included
Liu et al., 2021	China	Carbon intensity per km travelled by vehicle	Fossil fuel, electricity	Comparison of use of hydrogen fuel cells for heavy-duty trucks in China for years: 2020, 2030, 2050	Hydrogen fuel cells for heavy-duty trucks	Not included	Not included

Table 2 (continued)

Reference	Region	Functional unit	Feedstock	Process	Product use	LCA tools	Database
Logan et al., 2020	UK	Carbon emissions during operation of trains per passenger	Natural gas coal	Comparison of use of hydrogen and electricity for trains in the UK for years: 2017, 2050	Hydrogen for trains	Not included	Transport Energy and Air Pollution Model
Reaño and Halog, 2020	Philippines	1 kg hydrogen	Comparison of: rice husk, rice straw, corn cob, corn stalk, sugarcane bagasse, sugarcane leaves or tops, coconut shell, coconut husk	Gasification	Biohydrogen	OpenLCA 1.10	Ecoinvent 3.6
Reaño, 2020	Southeast Asia	1 kg hydrogen	Rice husk	Comparison of: alkali water electrolysis, gasification, dark fermentation	Biohydrogen	OpenLCA 1.10	Ecoinvent 3.5
Sadeghi et al., 2020	Iran	1 kg hydrogen	Water and solar power	Comparison of: steam methane reforming, coal gasification, photovoltaic electrolysis, solar thermal electrolysis	Hydrogen	Not included	The United States Energy Information Administration
Sako et al., 2021	Japan	1 kWh of energy	Water and photovoltaic power	Comparison of: Battery assisted, Conventional hydrogen production	Hydrogen	Not included	Ecoinvent 3.5
Sanchez et al., 2021	Colombia	1 kWh of energy produced in a low-temperature proton exchange membrane fuel cell	Comparison of: Ethanol Sugarcane press-mud	Comparison of electricity production using hydrogen from Sugarcane press-mud, Anhydrous ethanol from sugarcane	Biohydrogen for energy	Aspen Plus 9	Ecoinvent 3.4
Barceiß et al., 2019	Germany	1 kg hydrogen	Water and electricity	Comparison of: Steam methane reforming, Proton exchange membrane water electrolysis for years 2017 and 2050	Hydrogen	SimalPro	Ecoinvent 3.3
Chen et al., 2019	n/i	1 kWh of net power	Lignite	Integrated supercritical water gasification of coal	Hydrogen for power	Aspen Plus 8.4 and SimalPro 9	Ecoinvent 3
Cortés et al., 2019	n/i	1 tonne of alcoholic waste	Alcoholic waste stream	Catalytic steam reforming	Biohydrogen	SimalPro 8.5.2	Ecoinvent

Table 2 (continued)

Reference	Region	Functional unit	Feedstock	Process	Product use	LCA tools	Database
Fernández-Dacosta et al., 2019	Netherlands	1 GJ of fuel produced	Water and electricity	Comparison of the use of fuels for transport: Dimethyl ether produced from CO <sub>2</sub> , Methanol produced from CO <sub>2</sub> , Hydrogen produced from steam methane reforming, Hydrogen produced from water electrolysis using renewable energy	Hydrogen for the transportation sector	Aspen Plus	Ecoinvent 3.3
Li et al., 2019	China	1 kg hydrogen	Coal	Coal direct chemical looping hydrogen generation	Hydrogen	Not included	Not included
Ozturk and Dincer, 2019	n/i	1 kWh of energy		Brayton air standard cycle and Rankine steam cycle	Hydrogen for energy	SimaPro	Ecoinvent
Siddiqui and Dincer, 2019	USA	1 kg hydrogen	Comparison of: Coal, Biomass, Methane, Methanol, Ethanol, Water	Comparison of: Coal gasification, Biomass gasification, Methane reforming, Methanol reforming, Ethanol reforming, Water electrolysis via electricity	Hydrogen	GaBi	GaBi
Valente et al., 2019	Spain	1 kg hydrogen	Comparison of: Biogas Electricity	Comparison of: Biomass gasification, Steam methane reforming	Hydrogen	Not included	Product social impact life cycle assessment database. GreenDelta 2017
Karaca et al., 2020	North America	1 kg hydrogen	Nuclear energy	Comparison of: Conventional water electrolysis, High-temperature water electrolysis, 3-, 4- and 5-step CuCl cycles	Hydrogen	SimaPro	Ecoinvent 3



**Fig. 3** Types of functional units used in the life cycle assessment studies reviewed in the present work (N=24)

### Life cycle inventory analysis

Life cycle inventory analysis includes data collation for all the inputs and outputs for processes within the system boundary. In general, the more the processes included in the system boundary, the more complex, challenging and cumbersome is the inventory analysis. This also explains the fact that many studies did not include all the processes ranging from raw material acquisition to end-of-life management (summarised in Table 3). The two different kinds of data to be collected for an LCA study are: (1) foreground data for foreground systems which includes primary data that can be easily modified or improved and (2) background data for background systems typically comes from Life Cycle Inventory databases (Silva et al. 2020). Background systems support the foreground systems. Table 2 details the databases/data sources incorporated in LCA studies on hydrogen production such as Ecoinvent, expert communications, Greenhouse gases, Regulated Emissions and Energy use in Transportation.

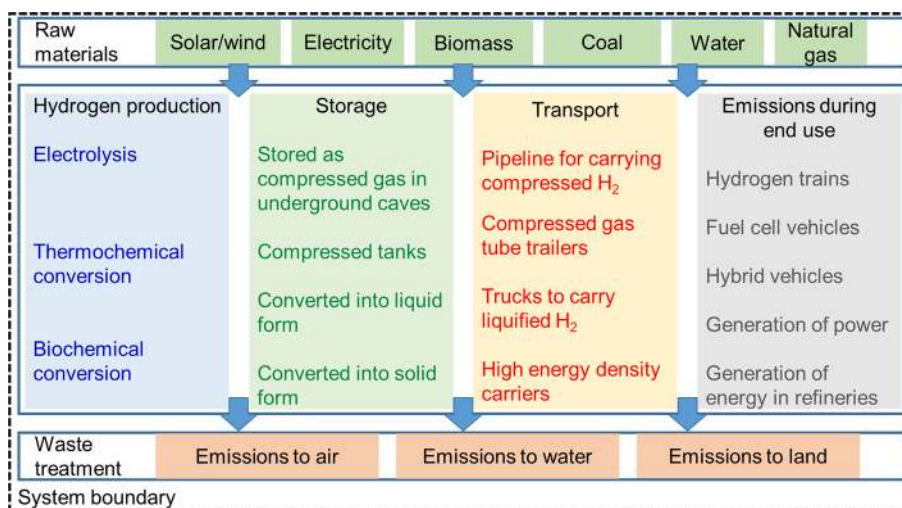
## Environmental impacts assessment

### Midpoint and endpoint indicators

Global warming potential due to emissions of greenhouse gases and depletion of fossil fuels was the centre of the attention in the environmental indicators for hydrogen production, with 100% of the studies accounting for either of these two categories (Table 4). More than half (54%) of the reviewed studies computed environmental impacts in categories that go beyond global warming potential and net energy use/performance. These environmental impacts included but were not limited to acidification, eutrophication, abiotic depletion, marine, freshwater and terrestrial ecotoxicity, and human toxicity.

Global warming potential expressed as kg CO<sub>2</sub> equivalent relates to greenhouse gas emissions; abiotic depletion recorded in kg Sb equivalent is linked to depletion of minerals, peat and clay; acidification reported in kg SO<sub>2</sub> equivalent is due to the emission of acidifying substances; eutrophication measured as kg PO<sub>4</sub><sup>3-</sup> equivalent is due to release of nutrients; particulate matter formation calculated as PM 2.5/PM 10 equivalent relates to the emission of PM 2.5 (particulate matter with ≤ 2.5 μm in diameter) and/or PM10 (particulate matter with ≤ 10 μm in diameter). Photochemical oxidation (commonly called as ‘summer smog’) occurs in stagnant air, in the presence of pollutants such as NO<sub>x</sub>, non-methane VOCs and others. Ozone layer depletion evaluates the global loss of ozone gas caused by trichlorofluoromethane (CFC-11) of the same mass. Land use calculated in m<sup>2</sup> is categorised as the transformation of urban, agricultural and natural land. Damage to terrestrial, freshwater and marine ecosystems is measured by ecotoxicity potential. Finally, human toxicity is caused due to the potential human health impacts of carcinogenic and non-carcinogenic pollutants.

**Fig. 4** Generalised system boundary used for conducting life cycle assessment of hydrogen production and consumption. This includes various raw materials such as solar, wind, biomass, coal, water and natural gas



**Table 3** Production and utilisation phases studied in the life cycle assessment studies

Reference	Raw materials production	Inter-operational transportation	Production	Purification	Storage	Utilisation	Emissions from waste treatment
Alviani et al., 2021	x	x	x	x	x	x	x
Booto et al., 2021	x	x	x			x	x
Bui et al., 2021	x	x	x	x	x		
Cvetković et al., 2021	x	x	x				x
Desantes et al., 2020	x	x	x	x	x	x	
Kerschler et al., 2021	x		x				
Kim et al., 2021	x	x	x				
Li et al., 2021	x	x	x				
Liu et al., 2021	x	x	x	x	x	x	
Logan et al., 2020						x	
Reaño and Halog, 2020	x	x	x	x			
Reaño, 2020	x	x	x				
Sadeghi et al., 2020	x	x	x	x	x		x
Sako et al., 2021	x	x	x	x			
Sanchez et al., 2021	x	x	x	x	x	x	
Bareiß et al., 2019	x	x	x	x			
Chen et al., 2019	x	x	x				
Cortés et al., 2019	x	x	x				
Fernández-Dacosta et al., 2019	x	x	x				x
Li et al., 2019	x	x	x		x		
Ozturk and Dincer, 2019	x	x	x				
Siddiqui and Dincer, 2019	x	x	x				
Valente et al., 2019	x	x	x	x	x		x
Karaca et al., 2020	x	x	x				

The midpoint categories are aggregated to present results as endpoint categories such as human health, damage to ecosystem quality in the form of loss of species and resources depletion (Osman et al. 2021b). It is argued that the environmental impacts should be presented as midpoint categories to prevent oversimplification or misinterpretation of environmental impacts (Kalbar et al. 2017). This is because endpoint indicators entail weighting of impacts. Evidently, only one study was identified that presented environmental impacts for both midpoint and endpoint indicators (Ozturk and Dincer 2019).

### Uncertainty and sensitivity analysis

Uncertainty arises in LCA studies due to sparse and imprecise nature of the available data and model assumptions (Cherubini et al. 2018). It is, therefore, imperative to consider and compute these uncertainties quantitatively to reach transparent, robust and trustworthy decisions.

There has been a vast development on the methods to imbibe these uncertainties in LCA models such as parameter variation and scenario analysis, classical statistical theory (e.g. probability distributions and tests

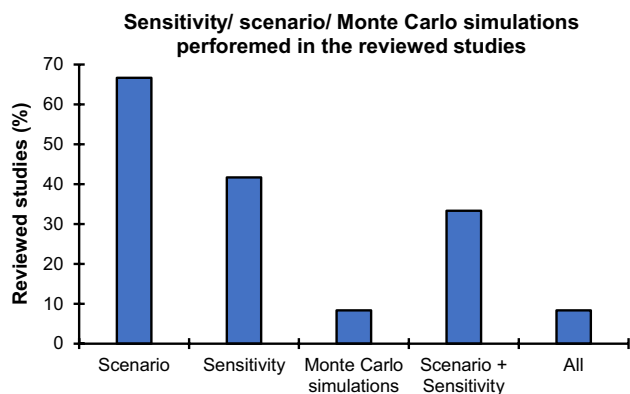
of hypothesis); Monte Carlo simulations, bootstrapping and other sampling approaches; nonparametric statistics, Bayesian analysis, fuzzy theory; and the use of qualitative uncertainty methods (Finnveden et al. 2009).

This review recorded that 67% of the studies used scenario analysis to account for parameter uncertainty (Fig. 5). Together with comparative studies mentioned in (Table 2) and scenario analysis in Fig. 5, this value reaches 96%, i.e. all but one study performed comparative and/or scenario analysis (Cvetković et al. 2021). This can be attributed to the dearth of the data and the serious effort required to conduct an LCA of biohydrogen production via anaerobic digestion (Cvetković et al. 2021). Furthermore, it was noted that 8% of the studies employed Monte Carlo simulations to propagate parameter uncertainties in the model.

Sensitivity analysis is conducted to distinguish processes in the hydrogen production chain that contribute to the burdensome environmental footprints. Relatedly, if environmental impacts are to be minimised, these will be the processes where future research should focus on (Al-Muhtaseb et al. 2021). 42% of the studies reviewed here conducted sensitivity analysis.

**Table 4** Environmental impacts studied in life cycle assessment studies in the context of hydrogen production

Environmental impacts	Reference
Global warming potential	Alviani Hirano et al. 2021, Bareiß De La Rua et al. 2019, Booto Aamodt Espegren et al. 2021, Bui Zhang et al. 2021, Chen Xu et al. 2019, Cortés et al. 2019, Desantes Molina et al. 2020, Fernández-Dacosta Shen et al. 2019, Kerschler Stary et al. 2021, Kim, Kim et al. 2021, Li Liu et al. 2019, Li Yao et al. 2021, Liu Mauzerall et al. 2021, Logan Nelson et al. 2020, Ozturk and Dincer 2019, Reaño and Halog 2020, Reaño 2020, Sadeghi Ghandehariun et al. 2020, Sako Koyama et al. 2021, Sanchez Ruiz et al. 2021, Siddiqui and Dincer 2019, Valente Iribarren et al. 2019
Net energy use, depletion of fossil fuels	Alviani Hirano et al. 2021, Bui Zhang et al. 2021, Cortés et al. 2019, Cvetković et al., 2021, Kim, Kim et al. 2021, Reaño 2020, Sanchez Ruiz et al. 2021, Fernández-Dacosta Shen et al. 2019, Li Liu et al. 2019, Ozturk and Dincer 2019, Siddiqui and Dincer 2019, Valente Iribarren et al. 2019
Abiotic depletion potential	Booto Aamodt Espegren et al. 2021, Chen Xu et al. 2019, Karaca Dincer et al. 2020, Kim, Kim et al. 2021, Ozturk and Dincer 2019, Sako Koyama et al. 2021, Sanchez Ruiz et al. 2021
Acidification potential	Booto Aamodt Espegren et al. 2021, Chen Xu et al. 2019, Cortés et al. 2019, Karaca Dincer et al. 2020, Kim, Kim et al. 2021, Li Yao et al. 2021, Reaño 2020, Sanchez Ruiz et al. 2021, Ozturk and Dincer 2019, Siddiqui and Dincer 2019, Valente Iribarren et al. 2019
Eutrophication potential	Booto Aamodt Espegren et al. 2021, Chen Xu et al. 2019, Cortés et al. 2019, Kim, Kim et al. 2021, Li Yao et al. 2021, Reaño 2020, Sanchez Ruiz et al. 2021, Ozturk and Dincer 2019, Siddiqui and Dincer 2019
Ozone layer depletion potential	Booto Aamodt Espegren et al. 2021, Bareiß De La Rua et al. 2019, Chen Xu et al. 2019, Cortés et al. 2019, Karaca Dincer et al. 2020, Ozturk and Dincer 2019
Photochemical oxidant formation potential	Booto Aamodt Espegren et al. 2021, Bareiß De La Rua et al. 2019, Chen Xu et al. 2019, Cortés et al. 2019, Ozturk and Dincer 2019, Siddiqui and Dincer 2019
Metal depletion potential	Bareiß De La Rua et al. 2019
Particulate matter formation potential	Bareiß De La Rua et al. 2019, Cortés et al. 2019, Siddiqui and Dincer 2019
Land use	Ozturk and Dincer 2019
Terrestrial ecotoxicity potential	Bareiß De La Rua et al. 2019, Cortés et al. 2019, Ozturk and Dincer 2019, Reaño 2020
Freshwater ecotoxicity potential	Cortés et al. 2019, Ozturk and Dincer 2019, Sanchez Ruiz et al. 2021
Marine ecotoxicity potential	Cortés et al. 2019, Ozturk and Dincer 2019
Human toxicity potential	Booto Aamodt Espegren et al. 2021, Bareiß De La Rua et al. 2019, Chen Xu et al. 2019, Cortés et al. 2019, Karaca Dincer et al. 2020, Ozturk and Dincer 2019, Sanchez Ruiz et al. 2021, Siddiqui and Dincer 2019

**Fig. 5** Details of the scenario, sensitivity and Monte Carlo simulations (to propagate uncertainty) conducted in the reviewed studies (N=24). Scenario analysis was conducted in 67% of the reviewed studies

## Interpretation of results

This stage of the LCA includes making interpretations, drawing conclusions and distinguishing the processes that can be improved to increase the environmental feasibility of the system. This stage could also involve presenting and communicating results to stakeholders. Table 5 summarises key findings from the reviewed studies.

## Key findings and recommendations for future life cycle assessment studies

Life cycle assessment is a complex tool that sits at the interface between science, engineering and policy. Despite this inherent complexity, it is recognised as a comprehensive tool to evaluate environmental impacts associated with products and processes. We reviewed LCA studies published from 2019 to 2021. This section draws recommendations for policymakers to create a sustainable hydrogen economy and LCA practitioners to conduct future studies.

**Table 5** Key findings reported in the reviewed life cycle assessment studies

Reference	Findings
Alviani et al., 2021	This study used aluminium waste materials and hot spring water to produce hydrogen for use in buses. The LCA estimates demonstrated environmental impacts for 1 MJ of hydrogen transport fuel produced as follows. Global warming potential = -195 g CO <sub>2</sub> eq/MJ Net energy use = 984 kJ/MJ
Booto et al., 2021	This study compared battery electric, conventional diesel-based and hydrogen trucks for environmental impacts. Global warming potential for battery electric truck = 0.286 kg CO <sub>2</sub> eq/km Global warming potential for conventional truck = 0.907 kg CO <sub>2</sub> eq/km Global warming potential for hydrogen fuel cell truck = 0.477 kg CO <sub>2</sub> eq/km
Bui, Zhang et al. 2021	This study evaluated three biomass to bioenergy pathways: (i) pulverised biomass-fired power plants which generate electricity, (ii) biomass-fuelled combined heat and power plants that provide both heat and electricity, and (iii) biomass-derived hydrogen production with carbon capture and storage
Cvetković et al., 2021	Anaerobic digestion followed by steam reforming analysed in this study showed a negative energy balance (with 16,837 GJ). Therefore, demonstrating that this process is energy unsustainable in an environmental context
Desantes et al., 2020	This study compared the use of fuel cells, hydrogen and conventional engines for mid-size passenger vehicles in Europe. The study concluded that the global warming potential for these three engines over the lifetime of the vehicle were: Electric vehicle = 15,000 kg CO <sub>2</sub> eq Hydrogen = 9000 to 49,000 kg CO <sub>2</sub> eq Conventional diesel-based = 24,500 kg CO <sub>2</sub> eq
Kerschler et al., 2021	The life cycle emissions of pyrolysis technologies are in the range of 1.9 to 6.4 kg CO <sub>2</sub> eq/kg H <sub>2</sub> , compared to state-of-the-art technology based on steam methane reforming technology 10.8 kg CO <sub>2</sub> eq/kg H <sub>2</sub>
Kim et al., 2021	Molten carbonate fuel cell system analysed for environmental impacts in this study showed global warming potential as 0.3 kg CO <sub>2</sub> eq/kWh, abiotic depletion potential as 1.90 g Sb eq/kWh, acidification potential as 30.5 g SO <sub>2</sub> eq/kWh and eutrophication potential as 0.01 g PO <sub>4</sub> <sup>3-</sup> eq/kWh. The main cause for the impact was found to be the reforming of liquefied natural gas in the operation stage
Li et al., 2021	This study calculated the environmental impacts of hydrogen production through coal gasification, natural gas steam reforming, thermochemical, water electrolysis via wind-power and thermochemical water splitting via Cu-Cl cycle in China. Global warming potential for all the processes = 0.7 to 17 kg CO <sub>2</sub> eq/kg of H <sub>2</sub> Acidification potential for all the processes = 1.7 to 30.7 g SO <sub>2</sub> eq/kg of H <sub>2</sub>
Liu et al., 2021	This study analysed carbon intensity per km travelled by heavy-duty trucks in China using hydrogen fuel cells for the years 2020, 2030 and 2050. The research concluded that replacing conventional fuels with hydrogen fuel cells for 100% of the heavy-duty truck fleet reduced 63% of greenhouse gas emissions in 2050
Logan et al., 2020	To achieve the UK net-zero objectives, trains should be converted to electric or hydrogen trains, which is impossible for economic or technical reasons. Furthermore, both electric and hydrogen trains produced much lesser carbon emissions per person than conventionally fuelled trains for the years 2017 and 2050
Reaño and Halog, 2020	The study compared global warming potential for 100 years for biohydrogen production using rice husk, rice shell, sugarcane bagasse, sugarcane leaves, corn cob, corn stalk, coconut husk and coconut shell. It was reported that global warming potential varied from -85 kg CO <sub>2</sub> eq/kg H <sub>2</sub> to 110 kg CO <sub>2</sub> eq/kg H <sub>2</sub>
Reaño, 2020	The study compared alkali water electrolysis, gasification and dark fermentation for hydrogen production. The dark fermentation pathway was recorded as the most efficient process based on a net energy ratio of 1.25 and global warming potential as 46 kg CO <sub>2</sub> eq/kg of H <sub>2</sub>
Sadeghi et al., 2020	The total greenhouse gas emissions were 10.28, 11.59, 3.08 and 2.06 kg CO <sub>2</sub> eq for 1 kg H <sub>2</sub> for steam methane reforming, coal gasification, photovoltaic electrolysis and solar thermal electrolysis, respectively
Sako et al., 2021	This study compared the production of battery-assisted and conventional hydrogen production systems. It concluded that the battery-assisted hydrogen production had lower greenhouse gas emissions (0.15 to 0.3 kg CO <sub>2</sub> eq/kWh) compared to the conventional hydrogen production process (0.9 to 1.08 kg CO <sub>2</sub> eq/kWh)
Sanchez, Ruiz et al. 2021	The use of sugarcane press-mud, as a biomass source, had environmental benefits in comparison to the use of anhydrous ethanol from sugarcane molasses as feedstock for power generation using hydrogen as an energy vector. The environmental impacts of 1 kWh of energy production using sugarcane press-mud were observed as: global warming potential = 1.2 kg CO <sub>2</sub> eq, acidification potential = 0.3 kg SO <sub>2</sub> eq and eutrophication potential = 0.01 kg PO <sub>4</sub> eq
Bareiß et al., 2019	The study showed that mainly the composition of the electricity mix determines the impacts like global warming potential for Proton exchange membrane water electrolysis process for hydrogen production. A reduction of the used materials causes only a very little reduction in global warming potential
Chen et al., 2019	This study demonstrated that the lowest global warming potential in all the scenarios considered in the integrated supercritical water gasification of coal was 0.66 kg CO <sub>2</sub> eq/kWh
Cortés et al., 2019	This study assessed alcoholic waste stream for catalytic steam reforming for the production of hydrogen. The catalyst is composed of a sepiolite base with nickel (15% weight) and lanthanum (1% weight). The environmental impacts for the production of 1 kg of H <sub>2</sub> were observed as global warming potential = 9.55 kg CO <sub>2</sub> eq and acidification potential = 4.25 g SO <sub>2</sub> eq

**Table 5** (continued)

Reference	Findings
Fernández-Dacosta et al., 2019	Comparison of the use of four types of fuels for transport: dimethyl ether produced from CO <sub>2</sub> , methanol produced from CO <sub>2</sub> , hydrogen produced from steam methane reforming and hydrogen produced from water electrolysis using renewable energy. The study concluded that none of these four alternative fuel options appears as the clear replacement to gasoline and diesel
Li et al., 2019	Global warming potential and non-renewable energy use for the coal direct chemical looping hydrogen generation process were 9.54 kg eq CO <sub>2</sub> /kg H <sub>2</sub> and 312.02 MJ/kg H <sub>2</sub> , respectively
Ozturk and Dincer, 2019	This study compared the use of hydrogen, oil, lignite, solar power and natural gas to produce 1 kWh of energy. It concluded that hydrogen was the most environmentally efficient fuel with global warming potential as 0.04 kg CO <sub>2</sub> eq/kWh
Siddiqui and Dincer, 2019	The water electrolysis route, coal gasification and biomass gasification routes showed global warming potential of 28.6, 23.7 and 4.4 kgCO <sub>2</sub> eq/kg H <sub>2</sub> , respectively. In addition to this, the ethanol, methanol and methane-based hydrogen production routes are estimated to have 12.2, 17.9 and 13.8 kgCO <sub>2</sub> eq/kg H <sub>2</sub> of global warming potential, respectively
Valente et al., 2019	In terms of global warming potential, biomass hydrogen was a better option than conventional hydrogen, however, to a lesser extent for acidification potential
Karaca et al., 2020	Nuclear-based hydrogen production methods caused global warming potential from 0.48 to 0.71 kgCO <sub>2</sub> eq/kg H <sub>2</sub>

Eq: Equivalent

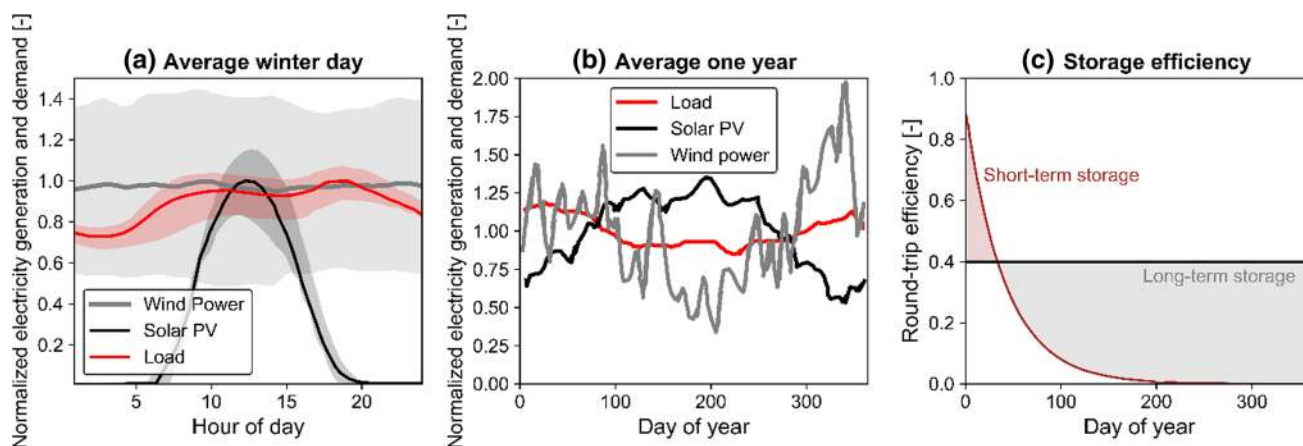
- During the review, no two LCA studies were identified to be similar. Differences in the geographical and temporal span, functional units and system boundaries considered, and environmental impact categories were reported. Therefore, it is recommended that the policymakers pay heed to the modelled processes and extent of the system boundary for making decisions for creating a sustainable hydrogen economy.
- Most of the studies did not encompass processes, inputs and outputs for ‘cradle-to-grave’ LCA analysis. Thus, future studies should conduct ‘cradle-to-grave’ evaluation for robust decision-making.
- About 54% of the reviewed studies computed environmental impacts in categories that go beyond global warming potential and depletion of fossil fuels. It is crucial to assess environmental impacts in more categories. Otherwise, there can be the issue of burden shifting, where hydrogen production processes are developed to mitigate climate change and energy security, however, leading to severe environmental and human health impacts such as acidification, eutrophication and human toxicity.
- Finally, focusing on production pathways, only eight studies were identified that computed environmental impacts for biohydrogen, showing that there is a considerable knowledge gap in production processes utilising bio-based feedstocks.

## Hydrogen underground storage

There are ambitious goals of the Paris agreement for climate change to be met than ever by 2050. However, the continuous increase in carbon dioxide (CO<sub>2</sub>) emission generated by the use and storage of fossil fuels has created a clear demand for alternative sources of clean and renewable energy (Ochedi et al. 2021). Solar and wind energy, however, provide intermittent and volatile power sources (as shown in Fig. 6) that are requiring backup solutions and/or energy storage at scales comparable to their power generation capacity (i.e. longer-term TWh storage solutions). In particular, some industrial sectors are hard to be decarbonised. To help balance the energy supply and demand, a capability of various energy storage technologies, with a dynamic combination of daily, weekly and seasonal storage, can reduce CO<sub>2</sub> emissions per unit of energy provided.

To date, the technical feasibility and economic attractiveness for developing large-scale, lithium-ion-based and seasonal energy storage batteries can be challenging to be implemented and provide an energy supply during high demand times. Such shortfall can be eliminated by storing the excess renewable energy chemically—in the form of hydrogen—in the subsurface aquifers, salt caverns and/or exhausted hydrocarbon reservoirs in the so-called Underground Seasonal Hydrogen Storage (USHS). The usage of hydrogen as an energy carrier can be a promising solution for clean energy because of its non-toxicity,





**Fig. 6** [A] Diurnal time series shows the matching of load, wind and solar of a typical day during the winter season for Europe with 15th and 85th percentiles for each average day time series. [B] Annual time series of weekly averages illustrate the seasonal correlation (i.e. excess/shortage) of load, wind and solar. Electricity generation and

demand normalised over the corresponding average value. [C] Schematic round-trip efficiency for a short-term (e.g. battery, brown line) and long-term (e.g. power to hydrogen, black line) storage technology. The figures were adapted from (2017) and (Gabrielli et al., 2020)

high specific energy and non-CO<sub>2</sub> emission after combustion. The challenge is to find hydrogen storage materials with high capacity. USHS, therefore, can be one of the most promising solutions for offsetting seasonal mismatch between energy generation and demand (Fig. 6), firstly for medium- and long-term storage while increasing contribution to low-carbon energy supply. Despite the vast opportunity provided by USHS, maturity still is considered low, with several uncertainties and challenges (Heinmann et al. 2021).

Hydrogen-based economy requires a large gas transport infrastructure. It has been suggested that existing natural gas pipe networks could be used to transport hydrogen (Melaina et al. 2013; Panfilov 2016). The gases would be transported as a mixture and separated afterwards. Some methods for separating mixtures of methane and hydrogen, particularly gas membrane separation, appear promising (Ockwig and Nenoff 2007).

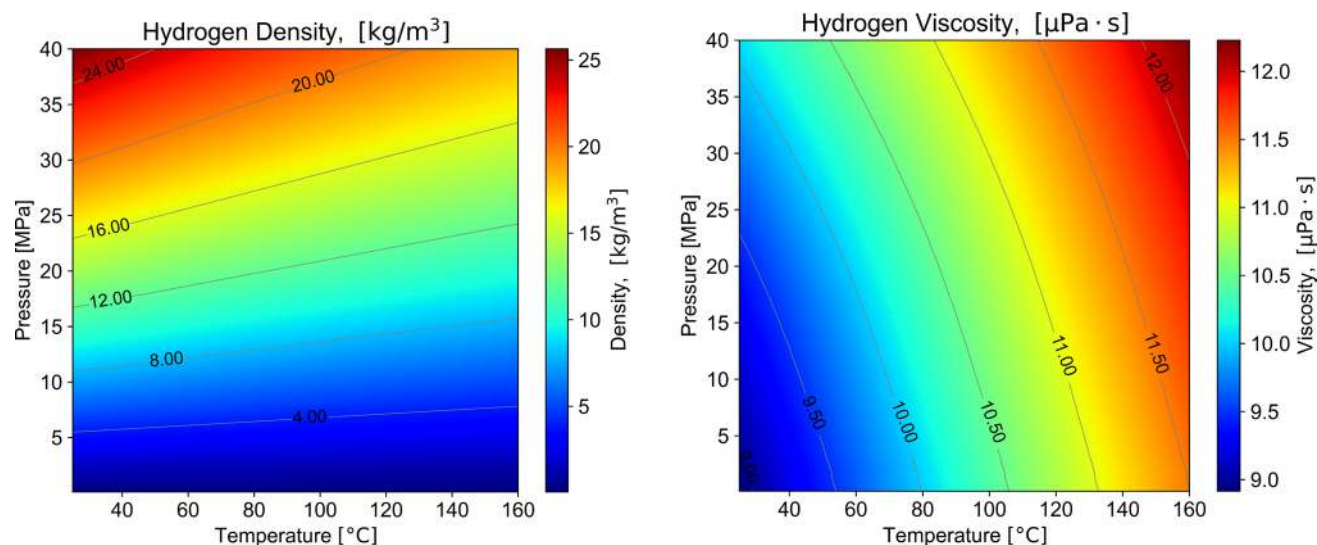
Geologically, underground formations are suitable for storing hydrogen, which may then be used as a carrier of chemical energy produced in times of surplus energy production, stored for several months and ultimately retrieved for re-electrification when it is needed most (Bauer et al. 2013; Bauer et al. 2017). As an illustration of the possible storage potential, a system volumetric capacity (i.e. the Net Energy Density) of hydrogen-based flow battery stores approximately 2.7 kWh/L (NREL) of electrolyte, and hence, an exhausted million-barrel oil field would hold > 3 TWh of electricity. This is equivalent to 30 weeks' output from a large offshore wind farm which is far more than is needed to eliminate the intermittency issues associated with such a facility. Hence, it was proved that only a few offshore gas fields are required to store enough energy as hydrogen to

balance the entire seasonal demand for UK domestic heating (Mouli-Castillo et al. 2021).

### Thermophysical properties of hydrogen

After hydrogen is produced at the surface from one of the technologies, it must be transported to a seasonal storage facility in a liquid or gas phase. Moreover, hydrogen can also be stored on the surfaces of solids (i.e. by adsorption) or within solids (i.e. by absorption) (El-Eskandarany 2020). During the loading cycle, where the power demand is at a peak, hydrogen can be easily re-converted for electrical generation.

Hydrogen can be considered as an ideal gas that may occur in various states over a wide temperature range and even at high pressures. Here, the thermophysical properties of hydrogen at the conditions relevant to the underground hydrogen storage were provided. One of its most important thermophysical characteristics is its low density, making it necessary for any practical application to compress the hydrogen or liquefy it. At intended storage depths, the density and dynamic viscosity of hydrogen are iteratively calculated using equation of state (EOS) and following (Span et al. 2020). Primarily, the hydrogen density (kg/m<sup>3</sup>) mainly increases with increasing pressure while dynamic viscosities (μPa.s) significantly increase with increasing temperature, as shown in Fig. 7. At low temperatures of −262 °C, hydrogen is solid with a density of 70.6 kg/m<sup>3</sup>. At higher temperatures, hydrogen is a gas with a small density of 0.089 kg/m<sup>3</sup> at 0 °C and at a pressure of 1 bar. The extent of hydrogen's liquid state can be presented as a narrow zone between the triple and critical points, with a density of 70.8 kg/m<sup>3</sup> at −253 °C.



**Fig. 7** (left) Density [ $\text{kg/m}^3$ ], (right) dynamic viscosity [ $\mu\text{Pa}\cdot\text{s}$ ] of hydrogen at representative P–T conditions which are typical for Underground Hydrogen Storage system. The calculations were car-

ried out by the authors, using the fundamental properties of Hydrogen as an ideal gas. By the time pressure of  $> 35$  MPa is reached, a deviation of 15% from the real values is expected

**Table 6** Storage density of hydrogen under certain pressures and temperature conditions. (i) liquid storage, (ii) cryo-compressed gas storage and (iii) compressed gas storage

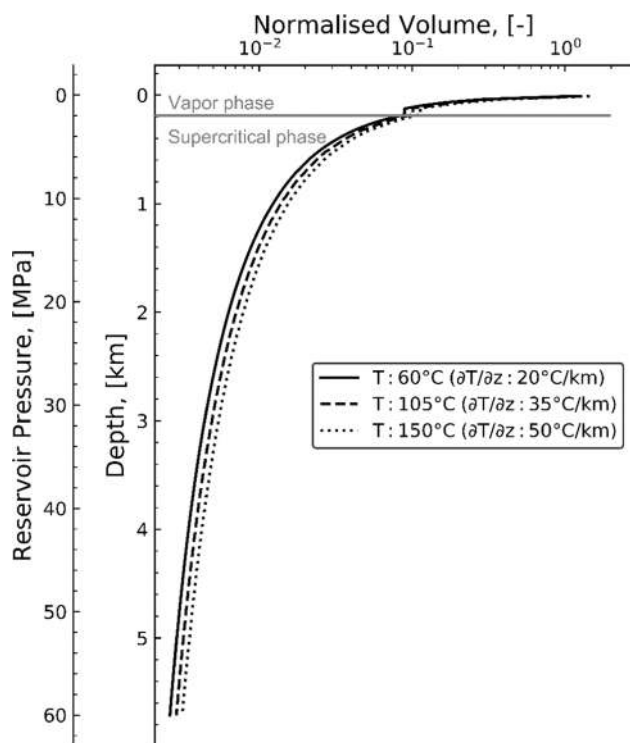
Stored hydrogen phase	Pressure [MPa]	Temperature [ $^{\circ}\text{C}$ ]	Density [ $\text{kg/m}^3$ ]
Case I: liquid storage	0.1	$< -253$	60–70
Case II: cryo-compressed storage *	25–35	$< -253$	$> 70$
Case III: compressed gas storage	5–30	25–130	10–40

\* Cryogenic pressure vessel (tank) is made of carbon fibre-coated metal

Three potential technologies for hydrogen storage, therefore, can be considered according to combinations of pressure and temperature relevant to the storage conditions (Table 6):

- Cryo-compressed hydrogen storage ( $\text{C}_2\text{H}_2$ ) and liquid hydrogen ( $\text{LH}_2$ ) storage: storage of hydrogen as a liquid requires cryogenic temperatures because the boiling point of hydrogen at one-atmosphere pressure is  $-253$   $^{\circ}\text{C}$  with a density of close to  $71$   $\text{kg/m}^3$ . These properties make storing hydrogen under standard atmospheric pressure and temperature extremely difficult due to the high cost and safety issues. Whereas other gases can be liquefied around the standard temperature of  $20$   $^{\circ}\text{C}$ , this is unfortunately practically impossible for hydrogen. Therefore, hydrogen needs compression into cryogenic vessels that can be pressurised to 25–35 MPa. Accordingly, the size of liquid hydrogen requires larger tanks reaching about three times larger than the currently used gasoline tank (El-Eskandarany, 2020).
- For pressure ranges between 5 and 30 MPa and temperature between 25 and 130  $^{\circ}\text{C}$ , hydrogen can safely be stored as a gas in underground geological formations. For USHS, hydrogen must be transported to a wellhead for underground storage. The hydrogen must then be compressed to be injected at sufficient pressure to enter the geological formation at the in situ pressure and temperature. Different potential geological storage sites for USHS are shown in Fig. 10 and will be discussed in more detail in the following sections.

Additionally, pressurised hydrogen gas takes a great deal of volume compared with, for example, gasoline with equal energy content—about 30 times bigger volume at 10 MPa gas pressure (El-Eskandarany 2020). USHS basically implies the reduction of the enormous volume of hydrogen gas due to the reservoir pressure gradient (Fig. 8). One kilogram of hydrogen in ambient temperature and at atmospheric pressure occupies a volume of  $11$   $\text{m}^3$ .



**Fig. 8** Normalised volume of hydrogen at the pressure–temperature (over the range of geothermal gradients) conditions plotted as a function of depth. Grey horizontal line at 800 m marks the minimum depth recommended for hydrogen injection, where it can be found as a supercritical phase at pressure and temperature conditions relevant for USHS (above 1.3 MPa)

### Fluid dynamics of hydrogen in a brine-saturated porous medium

In the context of the USHS system, the cyclic injection of hydrogen into (and possible retrieval from) a brine-filled permeable formation is part of multi-phase flow problems that have been studied extensively (Hashemi et al. 2021; Liebscher et al. 2016). In this case, a two-phase hydrogen–brine system is immiscible—the fluids are separated by a capillary interface. Likewise, the CO<sub>2</sub> geological storage, an important first approximation to the behaviour of the hydrogen–brine system, is found via applying a group of dimensionless ratios and solubility (and hence its mobility) that analyse the dynamics of two-phase immiscible flow systems (Ringrose et al. 2021). Viscous/capillary ( $N_{vc}$ ) and gravity/viscous ( $N_{gv}$ ) ratios are, respectively, the characteristic time ratios for fluid to flow in the transverse direction due to capillary and gravity forces to that in the horizontal direction due to viscous forces using the assumption of (Zhou et al. 1997). The two fluids here are assumed to be vertically segregate due to the gravity and density difference. Both ratios can be formulated in Eqs. 7 and 8 as follows:

$$N_{vc} = \frac{v_x \mu_{nw}}{k_{av}} \frac{\Delta z^2}{\Delta x} \frac{1}{\frac{dP_c}{dS_w}} \quad (7)$$

and

$$N_{gv} = \frac{\Delta \rho g k_{av}}{v_x \mu_{nw}} \frac{\Delta x}{\Delta z} \quad (8)$$

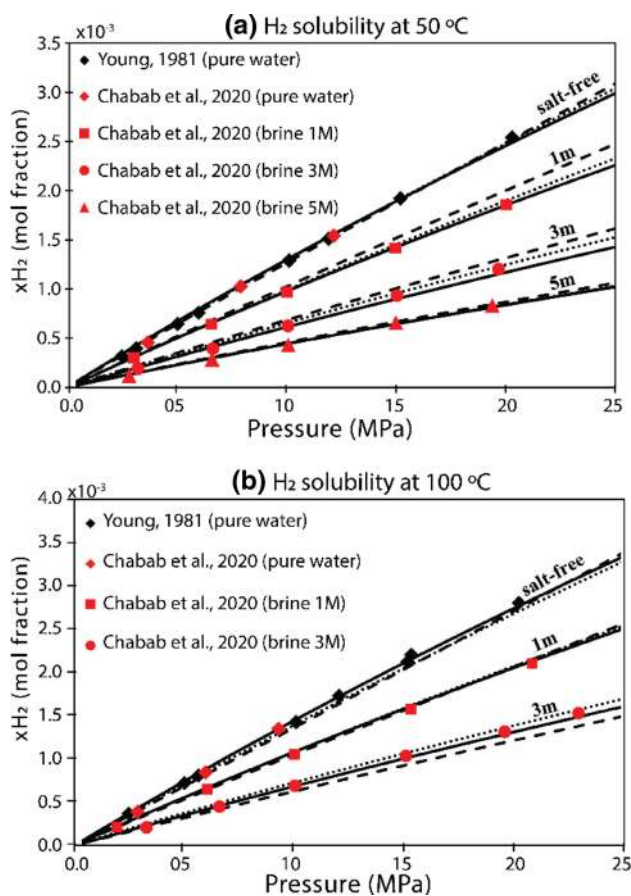
where  $u_x$  is the total flow velocity in the horizontal ( $x$ ) direction,  $\Delta x$  and  $\Delta z$  are the system dimensions,  $\mu_{nw}$  is the viscosity of the non-wetting phase (hydrogen),  $k_{av}$  is the average permeability,  $\Delta \rho$  is fluid density difference,  $g$  is the acceleration due to gravity and  $(dP_c/dS_w)$  is the capillary pressure gradient as a function of wetting-phase saturation.

Around the injection/production wellbore, viscous-dominated conditions are expected to occur due to the high-pressure gradient (Ringrose et al. 2021). However, within the reservoir and away from the injection/production wellbore region, gravity-dominated conditions are expected to occur. Such ratios, therefore, can be used to expect the fluid dynamic behaviour of the hydrogen–brine flow system and determine which factors are likely to be most critical, particularly when assessing large-scale macroscopic fluid flow, where the capillary and gravity forces become important enough to be not neglected.

Another important factor for USHS is the solubility of hydrogen in the resident formation fluid (water/brine). Therefore, forecasting the phase equilibria (solubility of hydrogen in brine and water content in the hydrogen-rich phase) under the geological storage conditions (i.e. at different temperatures, pressure and molality) is necessary for the study of hydrogen mobility and reactivity, as well as the control, monitoring and optimisation of the storage. Based on new experimental datasets, Chabab et al. developed predictive models to estimate the water content in the hydrogen-rich phase and precisely capture the salting-out effect on hydrogen solubility (Fig. 9) (Chabab et al. 2020).

### Large-scale hydrogen geological storage

A promising solution to help balances the energy supply from renewable intermittent sources and demand is hydrogen as an energy carrier for clean energy and must be accompanied by energy storage systems. The benefits of using hydrogen are because of its non-toxicity, high specific energy and non-CO<sub>2</sub> emission after combustion. However, the challenge is to find hydrogen storage materials with high capacity. Large-scale underground storage of natural gas has been practised successfully for many decades, with a global total of 413 billion standard cubic metres (BSCM) of natural gas storage accommodated in depleted gas fields (80%), underground aquifers (12%), and engineered salt caverns



**Fig. 9** Solubility of hydrogen in pure water as well as the brine of different molalities (up to 5 M), as a function of pressure (up to 25 MPa), and at the temperature of 50°C [a] and 100°C [b]. The symbols represent experimental results from the literature (Chabab et al., 2020). The solid, dotted and dashed lines represent the hydrogen solubilities calculated by the e-PR-CPA, SW and geochemical models, respectively. The figure is modified from Chabab et al., (2020)

(8%) (Perry 2005), as shown in Fig. 10. Here, these types of underground hydrogen storage systems have been considered (Lord et al. 2014; Panfilov 2010).

### Depleted hydrocarbon reservoirs

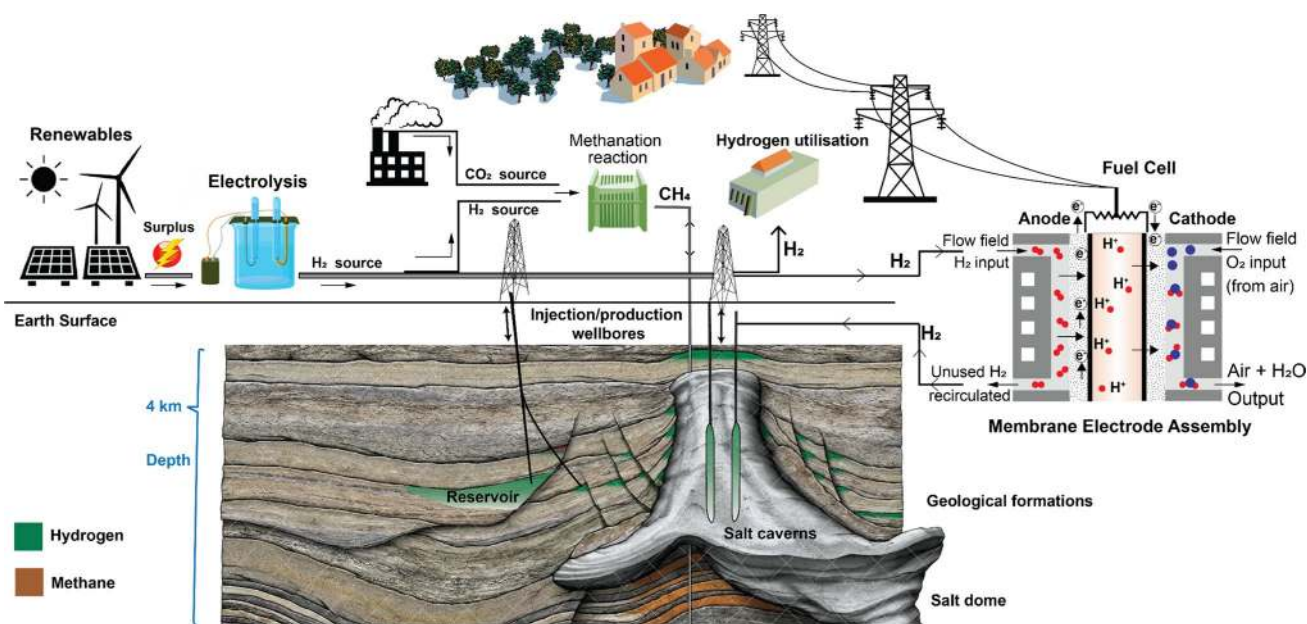
More often than not, depleted hydrocarbon reservoirs are appealing targets for USHS because of their storage capacity, proven seal, previous knowledge of reservoirs characterisation and existing infrastructure (i.e. natural gas pipeline network). Nevertheless, various physical, chemical and microbial processes are associated with USHS in hydrocarbon reservoirs (Heinemann et al. 2021) (summarised in Fig. 10).

While one can transfer know-how and technology from underground natural gas storage and underground carbon storage, some of the challenges USHS faces are peculiar. In both compressed gas and liquid forms, the low density

of hydrogen makes the seasonal storage of hydrogen in porous media (and possible retrieval) problematic. With a mass–density ratio of less than 0.01 compared to water for most relevant subsurface storage conditions,  $H_2$  is very light. Consequently, an  $H_2$  plume would experience strong buoyancy forces (i.e. the stronger the buoyancy forces, the higher the potential for hydrogen leakage), and water upconing towards the extraction borehole may occur (Heinemann et al. 2021; Sainz-Garcia et al. 2017).

- This limitation is felt most strongly during the hydrogen retrieval from the subsurface. The gas saturation around the production well required to keep a gas well flowing is of major concern since it will impact and reduce the production and ultimately will kill the well. The thinner the hydrogen plume will be, the lower gas saturation and the higher accumulation of resident formation brine in the downhole. Therefore, the dynamics of the USHS system require a wellbore model capable of describing/predicting the conditions (pressure and temperature) in the extraction borehole as the fluid(s) flow up (or the liquid accumulation at the bottom of) the borehole.
- Water upconing is the change in the hydrogen–water contact profile due to drawdown pressures. This phenomenon can be seen as the name implies: a cone of water formed below the perforations. One way to avoid upconing during  $H_2$  production is the use of a cushion gas (Kim et al. 2015; Oldenburg 2003), usually a cheaper and denser gas like nitrogen ( $N_2$ ), which helps prevent water flooding of the gas plume when  $H_2$  is being produced. This concept is well known in underground natural gas storage and has previously been proposed for USHS (Cao et al. 2020).

Additionally, it is important to note that USHS involves cyclic hydrogen injection (i.e. during power surplus) into and withdrawal (i.e. during energy demand) from the geological formations, where changes in the reservoir pressure may induce fatigue in the caprock and lowering the fracturing pressure at which hydrogen commences to leak through a seal rock. Therefore, assessing the sealing capacity to hydrogen (or hydrogen column height) will be crucial to keeping the risk of the potential upward leakage of hydrogen through the sealing caprock at a minimum. Seal rocks have fine pore and pore throat sizes that, in turn, generate hydraulically tight low-permeability caprocks with high capillary threshold pressures. High threshold pressures, together with wettability and interfacial tension (IFT) properties, determine the final column height that a seal can hold, thereby affecting the ultimate reservoir storage volumes. Compared to the underground natural gas storage, higher capillary entry pressures are expected to occur for hydrogen due to its higher interfacial tension (Hassanpouryouzband et al. 2021; Naylor et al. 2011). Therefore, hydrogen can be stored at a higher



**Fig. 10** Schematic diagram of different processes which are associated with hydrogen production using electrolysis, seasonal storage in geological formations and/or salt caverns, utilisation for ammonia production and re-electrification of hydrogen using fuel cells. The

figure shows different potential storage mediums for the hydrogen in the underground geological formations: reservoir/aquifer and salt caverns. The dimensions are not to scale

pressure in the reservoir than methane, with a reduced risk of geomechanical failure.

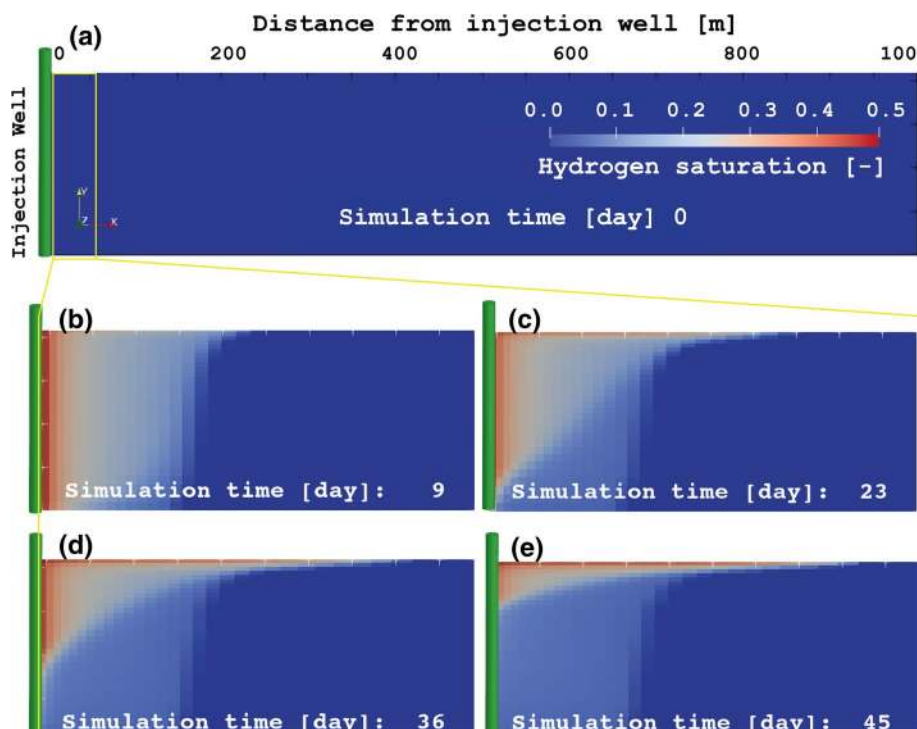
On the hydrogen injection into a storage reservoir, a very small fraction of hydrogen will dissolve into the formation fluids (Chabab et al. 2020), and water vapour may contaminate the hydrogen phase due to chemical disequilibrium. Hydrogen losses through diffusion need to be considered, as the diffusion ability of hydrogen is several times more than that of  $\text{CO}_2$  and methane, to such an extent that hydrogen can travel between the structures of ice-like crystals (Hasanpouryouzband et al. 2020).

In order to show the influence of the large density difference (Fig. 11) between the injected gas (hydrogen) and the resident formation fluid (brine) on the hydrogen plume migration during the seasonal storage period, we numerically simulate the injection of 10-ton kg of hydrogen over 10 days and its storage for 35 days. We used the numerical simulator PorousFlow Module, open-source software for solving parallel tightly coupled nonlinear THM processes in porous media (Wilkins et al. 2021; Wilkins et al. 2020). It is based on the MOOSE framework (Gaston et al. 2009) and its internal architecture relies on state-of-the-art libraries for finite element analysis (Kirk et al. 2006) and nonlinear iterative algebraic solvers (Balay et al. 2019). The simulation results are shown in Fig. 11. It is shown from the simulation standpoint that the leakage rate of hydrogen is going to be the biggest challenge due to the very high mobility of hydrogen, the small molecule size, the high dispersion

rate and the large density difference between the hydrogen and brine. Therefore, a proper tightness assessment of the caprock above the reservoir is required to prove its effectiveness for any possible hydrogen leakage. In addition, we propose expressly storing  $\text{H}_2/\text{CH}_4$  gas mixtures to improve the density contrast with the water. The mixed gas can, upon demand, then be extracted and transported in the same natural gas pipelines.

Subsurface microorganisms, including methanogens, sulphate reducers, homoacetogenic bacteria and iron(III) reducers can make use of  $\text{H}_2$  as an electron donor, which may lead to an unwanted accumulation of biomass in the vicinity of the injection borehole and/or loss of  $\text{H}_2$  (Ganzer et al. 2013; Hagemann et al. 2015a). The local rate of the biochemical reactions depends on the number of the particular microorganism (Hagemann et al. 2015b). Hence, an important problem for the modelling of USHS is the description of microbial growth and decay functions. Microbial conversion of hydrogen can only occur if the hydrogen is in the aqueous phase. A mixture of hydrogen with another gas means it will have a lower partial pressure and hence lower solubility in water. It was stated that if the temperature of the formation is higher than  $122^\circ\text{C}$  or the salinity is higher than 5 M NaCl, the hydrogenotrophic microbial activity becomes highly unlikely (Thaysen and Katriona 2020). Hence, if a storage reservoir is hot enough, one can combine hydrogen storage with  $\text{CO}_2$ , since methanogenic microbial activity will

**Fig. 11** Hydrogen–brine displacement in an idealised 2D horizontal cross section (i.e. geological storage formation). The injection wellbore is located at the left-hand side of the simulated domain. The subfigures are showing only the first 50 m horizontal distance from the injection well with 10× horizontal exaggeration. The horizontal exaggeration is 10x. [A] the reservoir is fully saturated with brine (i.e. before the hydrogen injection start). The migration of the hydrogen phase after [B] 9 days, [C] 23 days, [D] 36 days and [E] 45 days



be limited by the temperature constraint. Further, a high-pressure environment is toxic for some microorganisms.

Considering the deep depleted gas-condensate reservoirs, the risks are minimised here due to the presence of well-defined geological traps related to previously formed gas reservoirs. Unfortunately, the risk of migration from the target storage formation cannot be eliminated completely, particularly due to the re-pressurisation and change of the stresses and the long-term well integrity issues of the casing and cement.

### Salt caverns

Another underground storage medium, which could be used under certain conditions and locations, is the usage of salt caverns as high-pressure gas storage facilities (Fig. 10) (Gabielli et al. 2020; Hassanpouryouzband et al. 2021; Pudlo et al. 2013; Foh et al. 1979). Based on energy storage capacity (GWh) and discharge timescale, storing hydrogen in salt caverns can afford utility-scale, long-duration energy storage to meet the market need to shift excess off-peak energy to meet dispatchable on-peak demand. Salt caverns can hold substantial promise due to the self-sealing nature of the salt and the ability to customise the size and often shape of the caverns (Lord et al. 2014). However, the inaccessibility of the salt caverns in the area where hydrogen production is can be a limiting factor.

Salt caverns can be artificially constructed in the salt formation (or salt dome) by injecting water through an access wellbore, dissolving the salt and generating large volumes of brine in the so-called solution mining process. This process is associated with retrieving a large quantity of brine which requires disposal in an eco-environmental way. Finding suitable disposal repositories for brine disposal can be economically problematic due to higher costs for constructing longer pipelines which eventually may slow down or even hinder the permitting process. During the hydrogen withdrawing from the caverns under constant pressure, part of this saturated brine can be injected into the caverns to maintain the caverns' pressure and stability. Cushion gas, therefore, is not needed under these operating conditions (Foh et al. 1979; Taylor et al. 1986).

Compared to depleted oil and gas reservoirs, the key advantages for storing hydrogen in salt caverns are: (1) salt surrounding the caverns is highly impermeable and virtually leakproof where the only possibility for gas loss is escaped through leaky wells (Lord et al. 2014). (2) Salt does not react with hydrogen (Bünger et al. 2016). (3) Withdrawal of 'discharge' of hydrogen is highly flexible in rate, duration and volume with lower cushion gas requirements to avoid rock breakage. (4) Caverns are a mature, financeable storage technology that has been successfully used to store compressed gases for over 75 years with possible extensions for USHS.

The city of Kiel's public utility, as an illustration, has been storing town gas with a hydrogen content of 60–65% in a gas cavern with a geometric volume of about 32,000 m<sup>3</sup>

and a pressure of 8–16 MPa at a depth of 1330 m since 1971 (Kruck et al. 2013; Carpetis, 1988) estimated the hydrogen storage capacity for cavern volume of 500,000 m<sup>3</sup> and a casing shoe depth of 1000 m a pressure range of 180 to 60 bar is suitable of 4.0 Mio kg hydrogen (47 Mio m<sup>3</sup>(st)) and a cushion gas of 2.2 Mio kg (26 Mio m<sup>3</sup>(st)). For an economic prospect, the total installed costs, including wellbore drilling, compressors and gas treatment, were estimated to be about € 100 million (Michalski et al., 2017). Compared to energy storage in Li-ion batteries with a cost of 100 €/kWh, USHS in salt caverns offers a significant cost reduction potential in the total investment cost by a factor of 100.

Storage of hydrogen in the form of methane (natural gas) may be a preferable alternative for overcoming the storage problems associated with storing pure hydrogen in geological formations. When there is a surplus of renewable energy in the summer, hydrogen can be produced through water electrolysis. Furthermore, when this hydrogen and carbon dioxide combine in the methanation reaction, methane is produced, which can then be stored in a geological reservoir for winter use. This could be accomplished through a methane reforming reaction followed by using a fuel cell to generate electricity that can be fed into the power grid.

In short, hydrogen storage in a geological medium can offer a viable option for utility-scale, long-duration energy storage, allowing the hydrogen economy to grow to the size necessary to achieve net-zero emissions by 2050. While the operational experience of storing town gas in salt caverns provides considerable proof of its viability and operational best practice, full-scale deployment of USHS has yet to be evaluated for any associated risks and public acceptance of viewpoints, similar to the potential for induced seismicity.

## Hydrogen utilisation

### Fuel and power systems

Globally, the heat generated from domestic as well as industrial activities contributes by 33 and 50% of the carbon dioxide emissions and universal energy consumption rate, respectively (Dodds et al. 2015). The majority of gaseous emitted by the conventional burning process of natural gas are implicated in numerous environmental contamination issues (i.e. greenhouse gaseous emissions). The primary source of carbon dioxide emissions was energy consumption, with a global emissions rate of 33.1 gigatonnes in 2018, mainly resulting from the burning of fossil fuels. Contrarily, applying hydrogen gas as an alternative fuel to natural gas has proved to be an efficient pathway to reduce greenhouse gaseous emissions. Once it is generated from renewable energy sources, as shown in Fig. 1, it can directly participate in the decarbonisation process in the energy sector thanks to

its reacting nature, whether combusted or utilised in the fuel cell. The hydrogen is currently produced by conventional (non-renewable sources) of 18%, 30% and 48% from coal, heavy oil/naphtha and natural gas, respectively, which was negatively responsible for releasing about million 560 tonnes of carbon dioxide per year (Lui et al. 2020).

Moreover, given the costly natural gas employed throughout the power-producing framework (i.e. requires a huge area to store), hydrogen appears to be a viable option as a fuel feeding to gas turbines (Bicer and Khalid 2020). The utilisation of hydrogen in the central heating system instead of natural gas offers numerous merits: comparable operational activity and an increased heat generation rate with minimal harmful emissions (Dodds et al. 2015). Several factors, such as the Wobbe index, should be considered before forwarding hydrogen to various appliances. Generally, Wobbe index values differ considering the chemical composition of the gas. The Wobbe index number of pure hydrogen is about 48 MJ/m<sup>3</sup>; it falls within the permissible natural gas integrity extent for the vast majority of burners (Zachariah-Wolff et al. 2007). Supplying the operating system with a fuel beyond the Wobbe index band can negatively result in some operational problems (i.e. incomplete combustion and burner overheating). Clearly, attributing to the hydrogen's higher combustion velocity compared with the natural gas fuel, advanced burners with specialised technical specifications must be operated with hydrogen as a fuel feed stream.

Furthermore, the overabundant electricity generated from power facilities can be transformed into hydrogen, which can be either directed to the existing natural system (direct consumption) or chemically converted into chemicals used in different industrial aspects (Collet et al. 2017). Besides, hydrogen can be used individually in the aerospace industry or in combination with oxygen as propellants. The mentioned liquid mixture (oxygen and liquid) generates a large amount of energy and makes it more suitable for space applications. Because of releasing water during hydrogen combustion, in addition to its high efficacy compared with gasoline, these characters qualify it to be employed as an automotive fuel (Gurz et al. 2017).

### Hydrogen employment in power systems

Hydrogen is enormously used to store and transport energy in a variety of power applications, typically illustrated in Fig. 1 and discussed as follows (Parra et al. 2019):

#### Storing of energy and auxiliary services

Given the hydrogen's high storing efficacy, hydrogen-based energy storage has gained traction for storing energy over a medium/long term and in auxiliary services in the last decades. It can meet energy storage requirements over

a broad timescales to avoid any defect (shortage) that may occur between the product and the demand (required) of energy (Al Shaqsi et al. 2020). Recently, renewable energy production has grown rapidly; however, certain renewable energy supplies are sporadic and seasonally dependent. As a result, the produced renewable energy should be stored in a dependable form that is resistant to the fluctuation in those energy sources (Mehrerjedi et al. 2019). In particular, the most popular types of energy storage are: (1) power-to-power, (2) power-to-heat and (3) power-to-gas (Widera 2020). Hydrogen, in comparison, has a large energy storing capacity, a great storing time and flexibility. It has the ability to reduce energy volatility and absorb the surplus of energy production. Practically, it can deal with the economic and seasonal variations issues. Hydrogen can exceptionally balance between the resultant and required energies by storing the surplus energy when the production rate exceeds the required one as well as in times at which the electricity's price is minimal and reuse it in the opposite cases. Contrarily, hydrogen can be forwarded to generate electricity in the high energy demand.

Moreover, the storing capacity of hydrogen is higher than batteries, as it may range to weeks or months, unlike batteries that may extend (limited) for hours (Bocklisch 2016). Otherwise, hydrogen can be subjected to transform renewable resources to produce energy during different climatic conditions in different seasons. The storage capacity of hydrogen is estimated to reach up to megawatt-hours (1000 Kilowatts hours), even terawatts-hours, which is considered a high value by considering that of batteries (i.e. kilowatts hours). A slew of hydrogen power storage plants has been commenced worldwide, showing the technology's potency for the large scale. Examples of power plants established to produce and store hydrogen are Underground Sun Storage, Orsted and SoCalGas in Austria, Denmark and USA, respectively (Home | SoCalGas, <https://www.socalgas.com>).

In the Underground Sun Storage, the energy derived from wind and solar renewable resources is stored beneath the earth's surface. Referring to the difficult storing of the produced energy from renewable resources, the rest released power is reprocessed into hydrogen via electrolysis process and conserved for the futuristic challenges. The findings of the plant outlines revealed that it has the efficiency to equilibrate the basic energy requirements in line with the various seasonal variations. Other projects were established to face the shortage between the system supply and demand. Orsted plant was designed to operate the electrolyzers by subjecting the oversupply of energy generated from wind farms to them. Another project launched by SoCalGas on campus succeeded in directly converting the produced hydrogen from the solar electric system into methane inside a bioreactor.

Besides, hydrogen is hugely accounted as an assistant tool for providing the energy sector (grid) with the necessary services such as frequency maintenance and voltage strengthening via electrolyzers and fuel cells (Bird et al. 2016). In the HAEOLUS facility (Haeolus, <https://www.haeolus.eu/>), the oversupply of wind generation is directly fed into an electrolyser to generate hydrogen, which is subsequently forwarded into fuel cells to be used later for various purposes (utilities, data transmittance, systems controlling and others) (Larscheid et al. 2018). Another form of energy storage can be achieved by regulating the grid frequency near its normal value (50–60 Hz) by injecting or consuming energy in a coordinated manner to maintain the gap between the product and the required power. Numerous regulation reserves have been installed in different European grid systems. Commonly, frequent containment and restoration reserves have been used to handle the frequencies through the distributed control systems. The first mentioned controlling scenario supplies a steady feed stream in case of occurring a sudden corruption in frequency in a very short period, whereas the latter can tolerate a longer corruption beyond the 30 s. The twice services can be attained via electrolyzers and fuel cells by incrementing or decreasing their power setpoints related to frequency signals (Alshehri et al. 2019).

Besides, hydrogen-based equipment can contribute to voltage support by adjusting their power factor to meet the local voltage support requirements, which can be accomplished using inverter or rectifier monitoring systems (Alshehri et al. 2019). Some troubles such as blackout can occur in power plants, which was conventionally faced using a diesel Genset. The use of fuel cells may have the advantage to realise this scope given its no emissions and noiseless nature. These studies imply the profitability of hydrogen scaling up in the power sector.

### Power-to-gas

Power-to-gas is a process in which electrical energy is used to generate a combustible gas. Since hydrogen is thought to be a combustible gas with a large power density, power-to-hydrogen technologies are increasing (Eveloy and Gebreegiabher 2018). Because of the combustibility nature of hydrogen, it has been inserted into gas applications. The hydrogen generated from the electrolyser can be converted into methane by the methanation process, which is either pumped to the natural gas grid operating system or stored to achieve the financial budget for the energy market (Gondal 2019). By the literature, numerous pilot projects have been commenced worldwide with the highest establishment rate of 85% in Europe, followed by the USA and Japan (Thema et al. 2019). Among different European countries, Germany constructed a power-to-gas plant with a maximum production capacity of (40–100 megawatts) to be directed for



industrial purposes, and it will pump in the natural gas grid operating system from 2022 (Romeo et al. 2020).

Furthermore, several power-to-gas infrastructures have been installed in the regions rich in solar and wind renewable resources. A realistic study is displayed in the HAEO-LUS project (north of Norway). Chiefly, its core idea was based on using 2.5 Megawatts proton exchange membrane electrolyser to transform the produced wind power generated from wind farms into hydrogen, which can be consumed in various aspects. HyCAUNAIS project displays the viability of running a resilient power to gas facility in conjunction with the methanation approach by equipping a nominal 1 megawatts electrolysis area to produce hydrogen, which was methanated and inserted into natural gas grid operating system or combined with biomethane generation area from landfill biogas (HYCAUNAIS – Storengy – Europe en BFC. <https://www.europe-bfc.eu/beneficiaire/hycaunais-storengy/>).

### Fuel cells

Lately, fuel cells have gained worldwide attention as efficient and environmentally friendly energy generators. Practically, they are integrated electrochemical devices widely used to convert the delivered chemical energy into its electrical counterpart via redox reactions (Yuan et al. 2021). Regarding their efficacy for energy generation, they can be served as energy carriers. Fuel cells are composed of two electrodes (i.e. anode and cathode) separated by electrolytes responsible for the migration of ions between electrodes (Ogawa et al. 2018). There are numerous types of fuel cells such as alkaline fuel cell, direct carbon fuel cell, direct methanol fuel cell, microbial fuel cells, molten carbonate fuel cells, phosphoric acid fuel cell, proton exchange membrane fuel cells and solid acid fuel cells.

Table 7 displays different types of fuel cells with their operational conditions and efficiency%. During system operation, hydrogen is passed to the anode while oxygen is passed to the cathode. At the anode, the hydrogen molecules are split into protons and electrons by a catalyst. The positive hydrogen particles can pass through the membrane to the cathode side, but the negative cannot. However, electrons change their path by being forced to the circuit and generating electric current. At the cathode, the hydrogen protons, electrons and oxygen combine to produce a water molecule which is the end product of this reaction. Among different types of fuels (i.e. hydrocarbons and chemical hydrides), applying hydrogen in fuel cells is eco-friendly because it does not expel any pollutants (Psoma and Sattler 2002). It works within low temperatures ranges comparing with the internal combustion engine. As mentioned before, the end product of the hydrogen-based fuel cell is water, whereas the end products of diesel/natural gas-based fuel cells are

carbon dioxide and greenhouse gases (Xu et al. 2021). The main differences between fuel cells and traditional batteries are presented as follow: (1) operational mode of fuel cells is mostly like the traditional batteries, but the latter requires an electrical powering to run, (2) batteries can store hydrogen, unlike fuel cells that can provide a continuous electricity supply wherever hydrogen (fuel) and oxygen (oxidising agent) are available from outside sources. In addition to the mentioned differences, the batteries electrodes are steadily consumed during their extended usage, which entirely differs (not found) in the fuel cells (Spingler et al. 2017; Aydın et al. 2018).

### Co-generation and tri-generation distribution systems

Interestingly, fuel cells can be employed to optimise the efficiency of different power systems and reduce the overall production cost of these processes in several aspects, including co-generation systems (i.e. heat + power/cold + power) or tri-generation systems (i.e. cold + heat + power). Co-generation is the sequential generation of two different forms of beneficial energy from a primary single source (fuel cells). In that case, the electricity generated from fuel cells is used to meet the electrical demand, and the released heat is directed towards the heating activities. As a result, total efficiency will be about 95%. Systematically, co-generation fuel cell systems consist of different components, including fuel processors, power suppliers, heat recovery unit, energy (thermal/electrochemical) storage unit, control devices, additional apparatus (i.e. pumps) and stack. Commercially, a large number of facilities have been launched to improve the performance of co-generation systems. Different co-generation projects were erected around the world. In Japan, the plant installed by the ENE-FARM project (300,000 units/2018) simultaneously supplied the home with electricity and heat necessary for daily activities by using proton exchange membrane fuel cells ranged from 0.3 to 1 kilowatt. Initially, liquefied petroleum gas feedstock streams are fed into a reformer, where they are converted into hydrogen, which is further combined with oxygen inside the fuel cells to produce water, electricity and heat used later for various residential purposes (Yue et al. 2021). Recently, the manufacturing of micro-co-generation fuel cells has grown in Europe. Besides, more than 1000 micro-combined heat and power fuel cells were launched in 10 European countries between 2012 and 2017. The primary European plant for a micro-co-generation fuel cell was the ENE. Field project (ene.field. <http://enefield.eu/>). An LCA study was successfully performed for the mentioned project, and simply it revealed that co-generation fuel cell was environmentally in nature compared with other gas boilers and heat pumps strategies considering its less greenhouse gaseous emissions.

**Table 7** Types of fuel cells with their operational conditions and % efficiency (Ogawa et al., 2018; Edwards et al., 2008; Wilberforce et al., 2016)

Fuel cell	Fuel	Electrolyte	Temperature	Catalyst	Electrical efficiency (%)	Applications
Microbial fuel cells (MFCs)	Organic materials	Ion exchange membrane	20–60	Carbon supports on biocatalysts or platinum	15–65%	Wastewater treatment and biosensors
Alkaline fuel cells (AFCs)	Hydrogen	Potassium hydroxide solution, anion exchange membrane	70–130	Nickel/silver supported on carbon	40–55%	Military and aerospace applications
Phosphoric acid fuel cells (PAFCs)	Hydrogen	Phosphoric acid in silicon carbide	175–210	Platinum supported on carbon	40–55%	Combine heat and power production, medium- to large-scale power production
Proton exchange membrane fuel cells (PEMFCs)	Hydrogen	Solid polymer membrane (Nafion) (low temp) and Nafionpolybenzimidazole doped in phosphoric acid (high temp)	60–110	Carbon-supported platinum (low temp) and platinum ruthenium catalysts (high temp)	40–55%	Mobile applications
Solid oxide fuel cells (SOFCs)	Methane	Solid yttria stabilised zirconia	500–1000	Nickel–yttria-stabilised zirconia composite or strontium-doped lanthanum manganese	40–72%	Heat and power generation and small- to large-scale power production
Molten carbonate fuel cells (MCFCs)	Methane	Alkali carbonates in lithium aluminate	550–650	Lithiated nickel or nickel–chromium	50–60%	Large-scale power production
Direct methanol fuel cells (DMFCs)	Liquid methanol water solution	Solid polymer membrane (Nafion)	60–120	Carbon supports used on platinum or a combination of platinum and ruthenium	50–70%	Mobile applications
Direct carbon fuel cells	Carbonaceous materials	Solid Yttria stabilised zirconia, molten carbonate, molten hydroxide	600–900	Carbon-based materials, lanthanum manganese doped with strontium	70–85%	Combined heat and power production

PACE was another project, firstly started in 216, whereas about 2800 of combined heat and power fuel cells are fabricated. Briefly, the overall development in the electrical efficiency through the two inspected projects were 60 and 95%, respectively (Home - PACE Pathway to a competitive European fuel cell micro-cogeneration market. <https://pace-energy.eu/>).

Tri-generation strategy is an improved strategy of cogeneration in which a single primary source achieves the required cooling by thermally driven equipment. The working principle of heat pumps mainly stands on producing cooling from a thermal source. Typically, this can be achieved by using condenser and evaporator types of equipment. The gas released from absorbent/adsorbent is cooled down in the condenser and converted into a liquid by releasing its heat (refrigeration process). Then, the cooled down fluid continues to an evaporator, whereas it is evaporated by losing its contained heat. Significantly, the tri-generation fuel cells simultaneously reduce carbon emissions and enhance energy efficacy (Yue et al. 2021). Fong and Lee (2014) reported that employing a 593 kilowatts solid oxide fuel cell and absorption chillers, the carbon emissions were notably decreased by about 50% with an increase in the energy efficacy up to 75% (Fong and Lee 2014). A simulated 339 kilowatts solid oxide fuel cell combined with a combustor and a heat recuperation system proficiently recovered about 267 kilowatts of heat with an efficacy of 84%. Besides, they announced that 339 kilowatts solid oxide fuel cells provided with an absorption chiller generated about 303.6 kilowatts of cold with an efficacy of 89% (Yu et al. 2011).

## Transportation sector

Compared with conventional battery-powered powertrains, vehicles based on hydrogen fuel (hydrogen-fuelled vehicles) represent a promising solution to surpass them. Globally, the sales rate of hydrogen-fuelled vehicles is anticipated to be 3% and enhanced up to 36% in 2030 and 2050, respectively (Path to Hydrogen Competitiveness: A Cost Perspective - Hydrogen Council. <https://hydrogencouncil.com/en/>). Currently, innumerable vehicles companies are developing their operating system to be hydrogen-based, attributing to its dependability and quality. Toyota has evolved Mirai fuel cell vehicles by using proton exchange membrane fuel cells with a volume power density and maximum power productivity of 3.1 km/L and 144 kilowatts, respectively. The hydrogen-fuelled vehicles can be driven by different forms of hydrogen (i.e. liquid and compressed). The compressed (high pressurised) hydrogen is the most appropriate form in the vehicles storage system of Clarity and NEXO; hydrogen-based fuel cell vehicles developed by Honda and Hyundai companies, respectively. At the same time, liquid hydrogen operates Hydrogen 7 vehicle improved by BMW

company (Yue et al. 2021). Moreover, regional multi-unit trains powered by hydrogen have been entered into service in Europe and are projected to gain more economic benefits. Approximately 30% of presently employed diesel fleets may be phased out in the future (Study on the use of Fuel Cells and Hydrogen in the Railway Environment - Shift2Rail. <https://shift2rail.org/publications/study-on-the-use-of-fuel-cells-and-hydrogen-in-the-railway-environment/>).

Among different modes of transportation, the aviation division is regarded as the fastest transportation mode with anticipated annual growth in air traffic. The most common aircraft fuel is kerosene. Various aviation fuels often display a set of specifications, such as resistance to corrosion and severe temperature changes (Tzanetis et al. 2017). It is worth noting that petroleum accounts for the majority of the fuel used in the aviation sector. To improve energy preservation and reduce the negative environmental effects of fossil fuels, alternative, less harmful fuels such as liquid hydrogen are developed and thought to be eco-friendly. Table 8 presents some variations in the physicochemical properties between hydrogen and kerosene fuels. Refrigerated hydrogen fuel can be potentially better than kerosene as aviation fuel. It emits fewer greenhouse gaseous emissions and is easily produced from a variety of sources. Aside from that, the operating hydrogen-fuelled aircraft is characterised by minimal maintenance costs, long lifetime engines, high energy content and better combustion.

Furthermore, some constraints may arise during hydrogen utilisation as aviation fuel, such as depressed ignition energy, high flammability and the possibility of unburned traces forming that promotes metal embrittlement. Furthermore, the hydrogen admission with the onboard technology instead of inserting into the grid commercially allows its manufacturing companies to resell it (Nanda et al. 2017). The National Renewable Energy Laboratory manifested that the hydrogen cost in the mentioned case ranges from 3 to

**Table 8** Physicochemical properties of kerosene (jet fuel) and hydrogen (Nanda et al., 2017; Contreras et al., 1997)

Physicochemical properties	Kerosene	Hydrogen
Chemical formula	$C_{12}H_{26} - C_{15}H_{32}$	$H_2$
Density (Kg/m <sup>3</sup> )	0.8	0.071
Boiling point	167–266	–252.7
Melting point	–50	–259.2
Vol% for combustion limit	1.1–3.3	13–65
Combustion heat (kJ/kg)	42.8	120
Flame temperature	2126.9	2026.9
Vaporisation heat (J/g)	360	440
Standard heat of formation (kJ/mol)	–208.4	0
Minimum ignition energy (MJ)	0.25	0.02
Thermal efficiency (MJ/kg)	42.9	120.0

10 USD/Kg, while the most traded hydrogen cost is about 13.99 USD/Kg. To sum up, liquid hydrogen presents admirable efficacy as an aviation fuel for reducing greenhouse gaseous emissions, resulting in a significant improvement in air quality. Furthermore, by using hydrogen-based aviation fuels, over-reliance on traditional fuels could be decreased. The total cost of aircraft powered by liquid hydrogen is predominately associated with the cost of production and storage technologies (Eichman et al. 2012).

Recently, the global navigation movement in terms of maritime shipping has become increasingly important in the movement of different types of goods worldwide, which is in line with tremendous industrial progress in various fields. Unfortunately, this, in turn, led to an increase in the consumption of conventional fuels (i.e. diesel and heavy fuels). Regrettably, the pollution created by ships significantly implicates about 2.5% of the universal greenhouse gaseous emissions. Furthermore, bunkering activities broadly contribute to the leakage of heavy fuels in the aquatic environment, consequently posing a threat to the ecosystem. It was announced that carbon dioxide emissions associated with shipping activities release about 3.3% of the global emissions (Vogler and Sattler 2016). Other gaseous emissions such as nitrogen oxide and sulphur oxide are also associated with shipping activities. Accordingly, the maritime industry seeks more environmentally alternative fuels than conventional ones to overcome these obstacles (Prussi et al. 2021). Numerous suitable substitutes in different states, gas (i.e. hydrogen, propane) and liquid (i.e. bio-oil, methanol and ethanol) are used to compensate for the usage of traditional fuels (Al-Enazi et al. 2021; Abou Rjeily et al. 2021). Among them, hydrogen can be employed in maritime activities in two routes: (1) internal combustion engines and (2) fuel cells (Banawan et al. 2010). Relatively, fuel cells meet the energy requirements needed by ships sailing for long distances travelling and supply the ancillary energy requirements of larger ships in contrast to the other battery-powered ones. Numerous studies have been conducted to assess the feasibility of using hydrogen in maritime activities. Deniz and Zincir (2016) stated that hydrogen had a durable, safe and bunker capability criterion, qualifying as a favourable fuel for shipping. Although they reported that liquefied natural gas has the preference to be used as an alternative fuel, they recommended more research studies on utilising hydrogen as an effective alternative fuel (Deniz and Zincir 2016).

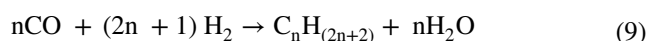
## Production of hydrocarbon fuels

### Production of hydrocarbon fuels via Fischer–Tropsch pathway

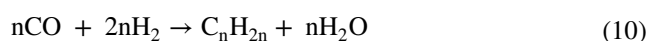
Syngas (synthesis gas), a mixture of carbon monoxide and hydrogen, is a product of different thermochemical conversion processes (i.e. pyrolysis, gasification and others)

and can be utilised by two scenarios: (1) direct fuel or (2) transformed into transportation fuels via Fischer–Tropsch synthesis process and syngas fermentation (Wainaina et al. 2018). The two strategies are categorised as gas-to-liquid transformation strategies that can generate hydrocarbon fuels and alcohols based on syngas feedstock stream (Gruber et al. 2019). Normally, the Fischer–Tropsch strategy (exothermic) operates at 200–350 °C and 1.5–4 MPa for reaction temperature and pressure, respectively (Okolie et al. 2019). Majorly, it comprises three main stages: (1) syngas production, (2) syngas treatment and (3) transforming into hydrocarbon fuels associated with their upgrading. Besides the production process of transportation fuel, other valuable products (i.e. paraffin, naphtha and others) can be produced. Significantly, the as-produced green fuels based on the Fischer–Tropsch process have numerous advantages over petroleum-based fuels. They have excellent burning characters, elevated smoking points and free of heavy contaminants. The physicochemical properties of resultant fuels depend heavily on reaction conditions (i.e. reactor type, heating rate, residence time and others) (Sun et al. 2017). The given equations from (Eqs. 9, 10, 11, 12, 13, 14 and 15) explicates the synthesis of different products (i.e. alkanes, alkenes, oxygenated products, methanol, ethanol and dimethyl ether via the Fischer–Tropsch process by participating in hydrogen. The hydrogen/carbon monoxide ratio is a critical controlling parameter in the Fischer–Tropsch synthesis process (Bermudez and Fidalgo 2016). Different types of catalysts (i.e. copper-based catalysts) can be used to optimise the yield of the Fischer–Tropsch process.

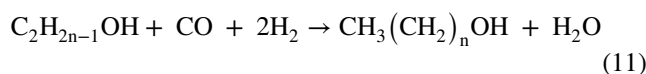
Synthesis of alkanes:



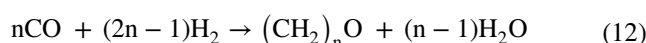
Synthesis of alkenes:



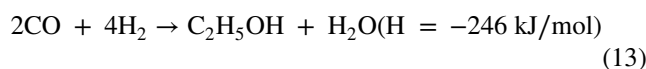
Synthesis of alcohols:



Synthesis of carbonyl:



Synthesis of ethanol:



Synthesis of methanol:

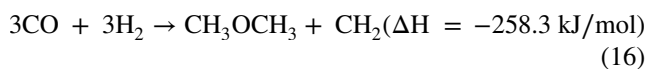


A ratio of  $H_2/CO$  of 2:1 is preferable for the synthesis of hydrocarbon fuels via water–gas shift reaction as given in Eq. 15:



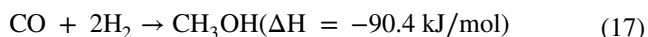
Dimethyl ether is admirably commonly realised as an efficient alternate for diesel fuel (Kim and Park 2016). Distinctly, numerous physicochemical features characterise liquefied petroleum gas, such as anti-corrosive, anti-carcinogenic, less nitrogen oxide and carbon monoxide emissions during its burning, less engine noise and high cetane number (Dincer and Bicer, 2020). In general, dimethyl ether can be produced by (1) direct route (combined single step of methanol synthesis and dehydration) or (2) indirect route (separated methanol synthesis and dehydration steps) as shown in Eqs. (16, 17 and 18) (Gogate, 2018):

Direct route (single step):

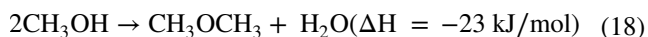


Indirect route (two steps):

Synthesis of methanol:



Dehydration of methanol:



### Production of hydrocarbon fuels via Syngas fermentation pathway

Syngas fermentation (biorefining) pathway is regarded as the interconnection between the biochemical and thermochemical scenarios (Thi et al. 2020). It produces value-added products (i.e. alcohols) from syngas by flexibly employing several groups of microorganisms at different reaction temperatures of 37–40 °C and 55–90 °C for mesophilic (i.e. *Clostridium autoethanogenum*) and thermophilic (i.e. *Moorella thermoacetica*), respectively. During the process, the feedstock of syngas can be simply converted into alcohols (i.e. ethanol) via two subsequent stages via (1) producing acetyl coenzyme A and then (2) its transformation into ethanol. Other alcohols and chemicals (i.e. acetate, butanol and formate) can be synthesised by acetogenic bacteria (Park et al. 2017). Regarding several operational advantages characterised to syngas fermentation such as (1) no necessity for using costly pretreatment step, (2) process' versatility with different biomass composition, (3) independent on the hydrogen/carbon monoxide ratio in the feedstock upstream, (4) high selectivity of as-used microorganisms and (5) moderate (ambient) working parameters with no necessity for

catalysts usage or its poisoning trouble, they support it over the Fischer–Tropsch process. However, there are some operational challenges such as (1) weak solubility of the gas in the liquid state, (2) complicated bioreactor design, (3) existence of impurities and (4) low yield of production. Briefly, integration between different thermochemical, biochemical and hydrothermal routes can effectively compensate for the shortage of individual techniques and maximise productivity (Rigueto et al. 2020).

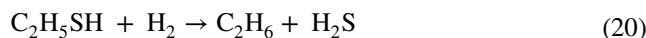
### Refining of crude oil and petroleum products

Commercially, hydrogen is conceived as an upgrading (improving) agent for crude oil products and petroleum distillates in terms of hydrocracking and hydroprocessing and processes. The hydrocracking process is defined as treating heavier hydrocarbons with hydrogen to simultaneously split them into lighter derivatives and enhance the hydrogen/carbon ratio (El-Sawy et al. 2020). In hydroprocessing, various heteroatoms such as nitrogen, sulphur, oxygen and heavy metals are majorly captured from petroleum products via different treatment processes named: hydrodenitrogenation (Dasgupta and Atta 2020), hydrodesulphurisation (Han et al. 2018), hydrodeoxygenation (Yfanti and Lemonidou 2020) and hydrodemetallisation (Rana et al. 2020), respectively, as displayed in Eqs. (19–21).

Hydrodenitrogenation:



Hydrodesulphurisation:



Hydrodeoxygenation:



This can be achieved by reacting the upstream feedstock (heavy oils and petroleum products) with hydrogen through catalytic reaction, resulting in removing these contaminants and saturating the aromatics (C–C) bonds. The elimination process of these contaminants from feedstocks directly contributes to fuel upgrading because they deactivate the as-used catalysts due to their adsorption on the surfaces of the catalyst (blocking of active catalyst sites). Recently, the appeal for inserting hydrogen in hydroprocessing has been increasingly growing (Al Obaidi et al. 2018). From the environmental point of view, the key cause of this pattern is the combination of strict environmental legislation governing gaseous greenhouse emissions and other particulate contaminants, as well as product quality specifications. Generally, numerous upgrading techniques are directed to improve the physicochemical properties of heavy oils by decreasing

their viscosity and carbon/hydrogen ratio at the same time (Misra et al. 2017).

### Production of ammonia

Ammonia is deemed one of the essential chemicals largely employed in industrial fertiliser activities with huge global production rates. The biggest ammonia production plant has projected to achieve a daily capacity rate of 3300 metric tons (Brightling 2018). Broadly, ammonia can be introduced as fertiliser in the agriculture sector. Additionally, it is provided to various industries such as polymers processing, explosives, refrigerant, pharmaceuticals, gas sensors and fuel cells. The ammonia synthesis process is promoted by the catalytic reaction between hydrogen and nitrogen elements through the Haber process (Arora et al. 2018). It is performed in the as-designed reactor under operating conditions of 20–30 Mpa and 300–500 °C for pressure and temperature, respectively, using KOH-promoted finely divided iron catalysts with the required energy of 2.5 EJ (Tolod et al. 2019).

Moreover, the hydrogen addressed to the ammonia synthesis process is primarily derived from steam gas reforming, which is not regarded as environmentally friendly. Accordingly, there is an increasing interest in other green and sustainable ammonia synthesis pathways, such as electrochemical hydrogen manufacturing techniques and photocatalytic nitrogen fixation (artificial photocatalysis). The distinctiveness of the electrochemical ammonia synthesis pathways routes is controlled by the employed energy sources. Hydrogen can be generated from water employing an electrolysis process using renewable green sources (i.e. wind and solar energy) and, hence, reduce harmful greenhouse gaseous emissions (Bicer and Dincer 2017).

### Metallurgical industries

Generally, hydrogen can produce oxy-hydrogen flames in industrial metallurgical activities and act as a reducing agent to obtain metals from their ores. During the oxy-hydrogen flames synthesis process (exothermic reaction), hydrogen is allowed to react with oxygen at very high temperatures (3000 °C) to produce oxy-hydrogen flames, used later for cutting and welding working on non-ferrous metals (Polverino et al. 2019). Otherwise, hydrogen is reliably characterised by its high ability to recover (reduce) metals from the aqueous solutions of their salts (hydrogen reduction). The metals may be powdered for later metallurgical usage or incorporated into a composite material. Chemically, hydrogen can interact with the

elements of periodic tables in three ways: (1) ionic bond formation between the elements of Ia and IIa groups, (2) interstitial solid solution between the elements of VIa, VIIa and VIII groups and (3) metallic bond between elements of IIIa, Iva and Va groups. Additionally, the electrostatic shielding phenomenon can be generated by attributing the hydrogen's capability to capture free electrons and the self-trapping of metals. Likewise, the small particle size of hydrogen effectively facilitates the process of metal–hydrogen interaction (Agrawal et al. 2006).

### Conclusion

To ensure the long-term production of clean and green hydrogen, it is crucial to conduct a critical assessment of various production routes and their environmental impacts, as well as seasonal storage and utilisation options. Hydrogen is produced from either fossil-based or renewable feedstocks; however, each route has advantages and disadvantages. The current hydrogen colour coding is imprecise, assuming that green hydrogen always has lower carbon emissions than blue or grey hydrogen, which is not always accurate.

Water electrolysis is gaining momentum; however, meeting 24% of energy demand with hydrogen in a 1.5-degree scenario of climate change mitigation will necessitate massive amounts of additional renewable electricity generation. In this scenario, approximately 31,320 terawatt-hours of electricity would be required to power electrolyzers, which is more than is currently produced globally from all sources combined. Furthermore, > \$11 trillion in manufacturing, storage and transportation infrastructure would be needed. The affordability and accessibility of freshwater is one side of the coin, and the proximity of these two supplies, i.e. renewable energy and freshwater, is the other. Water electrolysis research priorities include lowering the capital cost of electrolysis technology, locating water resources, find utilisation routes for the produced oxygen and increasing the efficiency of the process.

In terms of biomass gasification, the economic feasibility of producing hydrogen from biomass must be closely related to the availability and affordability of raw materials in the surrounding area. The main characteristics of the supply materials are the biomass physicochemical properties, distribution and hydrogen rate. Because biomass feedstocks differ greatly in structural composition and shape, all of these factors must be considered when combining the feedstock with the appropriate conversion technology. In conclusion, there are challenges associated with the most common hydrogen generation routes, such

as steam methane reforming, water electrolysis, coal or biomass gasification, methane pyrolysis with or without carbon capture and storage technology.

To understand advances in evaluating environmental impacts due to hydrogen production, we performed an intensive critical review of 24 life cycle assessment studies published from 2019 to 2021, including methods and findings. The important methodological approaches and key findings observed were:

1. No two life cycle assessment studies were identified to be similar. There were differences in the geographical and temporal span, functional units and system boundaries considered, and environmental impact categories assessed. Therefore, it is recommended that the policy-makers pay heed to the modelled processes and extent of the system boundary for making decisions for creating a sustainable hydrogen economy.
2. Many life cycle assessment studies did not encompass processes, inputs and outputs for ‘cradle-to-grave’ analysis. Thus, future research should pay more attention to ‘cradle-to-grave’ evaluation for robust decision-making.
3. In addition to the global warming potential and depletion of fossil fuels, environmental impacts in more categories for hydrogen production processes must be evaluated.

Furthermore, large-scale energy storage is key in securing the energy supply chain for the next energy transition using electrolysis-generated hydrogen. The Underground Seasonal Hydrogen Storage (USHS) holds great potential to overcome the natural temporal fluctuations inherent in renewable energy production at the scale required to achieve net-zero by 2050. The selection of geological porous media for USHS should be based on a comprehensive geological investigation that includes an assessment of their utility on both a basin and regional scale, fluid flow behaviour of hydrogen in brine-saturated subsurface reservoirs, an assessment of storage capacity, the safety of long-term storage, geochemical and biological reactions triggered by hydrogen injection, the geomechanical response of the subsurface to hydrogen storage and other measures. The discussed procedures can lead to informed decision-making in terms of forecasting best-operating strategies and ensuring safe and efficient hydrogen storage installation. Further research to integrate the theoretical studies with existing experimental USHS trials is required to minimise the uncertainty that might be associated with the feasibility of large-scale hydrogen storage. Finally, blending the need with the various utilisation routes such as fuel production, ammonia production, metallurgical industries and power systems is crucial in the hydrogen economy.

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## Declarations

**Conflict of interest** The authors declare that no conflict of interest.

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