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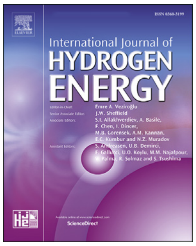

Advancements in hydrogen production, storage, distribution and refuelling for a sustainable transport sector: Hydrogen fuel cell vehicles

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HIGHLIGHTS

- Technological advancements of hydrogen production, storage and transport are reviewed.
- Emissions and economic aspects of production, storage, transport and fuelling are analysed.
- Refuelling protocols, temperature rise issue and its control strategies are discussed.
- Recent progress of mobile hydrogen refueller is explored.
- Challenges of hydrogen production to fuelling and future research directions are presented.

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ABSTRACT

Hydrogen is considered as a promising fuel in the 21st century due to zero tailpipe CO_2 emissions from hydrogen-powered vehicles. The use of hydrogen as fuel in vehicles can play an important role in decarbonising the transport sector and achieving net-zero emissions targets. However, there exist several issues related to hydrogen production, efficient hydrogen storage system and transport and refuelling infrastructure, where the current research is focussing on. This study critically reviews and analyses the recent technological advancements of hydrogen production, storage and distribution technologies along with their cost and associated greenhouse gas emissions. This paper also comprehensively discusses the hydrogen refuelling methods, identifies issues associated with fast refuelling and explores the control strategies. Additionally, it explains various standard protocols in relation to safe and efficient refuelling, analyses economic aspects and presents the recent technological advancements related to refuelling infrastructure. This study suggests that the production cost of hydrogen significantly varies from one technology to others. The current hydrogen production cost from fossil sources using the most established technologies were estimated at about \$0.8–\$3.5/kg H_2, depending on the country of production. The underground storage technology exhibited the lowest storage cost, followed by compressed hydrogen and liquid hydrogen storage. The levelised cost of

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the refuelling station was reported to be about \$1.5–\$8/kg H_2, depending on the station's capacity and country. Using portable refuelling stations were identified as a promising option in many countries for small fleet size low-to-medium duty vehicles. Following the current research progresses, this paper in the end identifies knowledge gaps and thereby presents future research directions.

Nomenclature table

Units	
\$/US\$	United States dollar
kWh	Kilowatt hour
TWh	Terawatt hour
GW	Gigawatt
kW	Kilowatt
MJ	Megajoule
MW	Megawatt
Symbols	
ρ	Density
C	Specific heat
M	Mass
R^2	Coefficient of correlation
Abbreviations	
APRR	Average pressure ramp rate
CCCS	Coal with carbon capture and storage
CCS	Carbon capture and storage
CFD	Computational fluid dynamics
CG	Coal gasification
CHSS	Compressed hydrogen storage system
CNG	Compressed natural gas
DOE	Department of Energy
GHG	Greenhouse gas
GIS	Geographic information system
HFCEVs	Hydrogen fuel cell electric vehicles
ICEVs	Internal combustion engine vehicles
ISO	International Organisation for Standardisation
LPG	Liquid petroleum gas
MC	Mass and thermal Capacity
MOF	Metal organic framework
PEM	Proton exchange membrane
SAE	Society of Automotive Engineers
SMR	Steam methane reforming
SOC	State of charge
SWG	Supercritical water gasification
TA	Ambient temperature
WTW	Well to wheel

1. Introduction

Global energy consumption is increasing rapidly due to population growth and economic development activities happening around the world. Until now, fossil fuels have remained as the major energy source in the world and shared more than 84% of global primary energy consumption [1] as shown in Fig. 1(A). Oil accounts for nearly one-third of total energy consumption followed by coal and natural gas. However, excessive use of fossil fuels and associated environmental emissions are of major concern to policymakers and the scientific and general communities [2]. Global CO_2 emissions from energy consumption increased by 6% in 2021 when compared to that in 2020, and the total amount reached 36.3 billion tonnes, of which nearly one-fourth was emitted from the transport sector [3]. The combustion of coal released 42% of global CO_2 emissions, followed by oil and gas, as presented in Fig. 1(B). Therefore, a global energy transition towards renewable energy is necessary for decarbonising the dominant CO_2-releasing sectors, particularly the transport sector, to achieve the net-zero CO_2 emissions target by 2050 [4].

Hydrogen is a potential emerging alternative to fossil fuels with a zero-greenhouse gas (GHG) emissions footprint. Hydrogen can be used to power vehicles and for electricity and heat generation, as well as in many other typical industrial applications, including ammonia and methanol production, steel production, petroleum refining, metal treatment and fertiliser production. Hydrogen has approximately 3.1 and 3.2 times higher gravimetric energy density compared to gasoline and diesel fuels, respectively [6]. Transport and heat and electricity production are the most emerging sectors of hydrogen application [7,8]. Global hydrogen consumption in 2021 was about 94 million tonnes, a ~5% increase compared to the previous year and is expected to increase to ~130 million tonnes by 2030 to meet longer-term net zero targets [5]. Approximately 43% of global hydrogen was consumed by refining sectors in 2021, followed by ammonia production (~36%), methanol production (~16%) and the remaining ~5% for other sectors' uses [5].

The demand for hydrogen, particularly in the transport sector, is increasing rapidly due to the commercialisation of

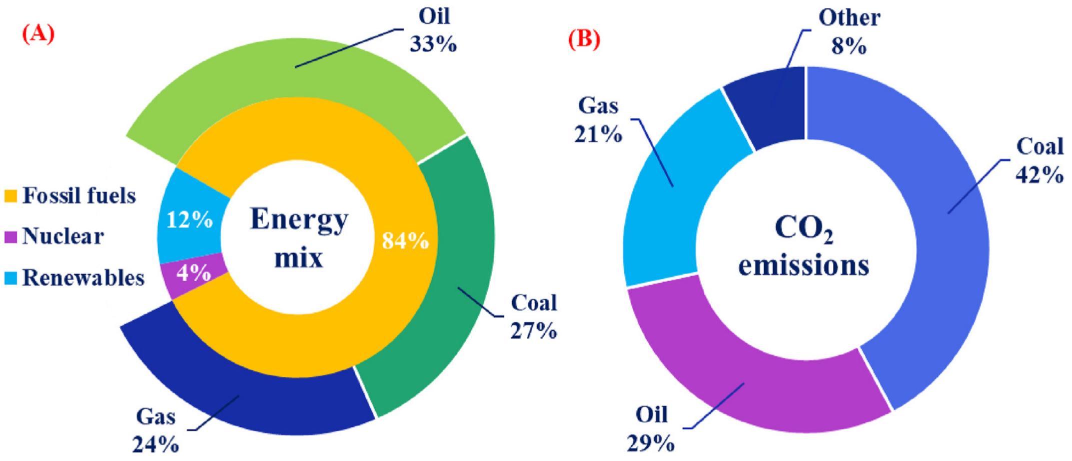


Fig. 1 – (A) Global primary energy consumption and (B) CO_2 emissions by fuel type (data from [1,5]).

hydrogen fuel cells, the growing market for hydrogen fuel cell vehicles and zero tailpipe GHG emissions from hydrogen-powered vehicles [9]. In 2020, the total investment in hydrogen fuel cell vehicles related projects was around $1500 million, which is expected to increase in coming years. Hydrogen consumption by the transport sector (30 kilo tonnes) in 2021 was ~1.6 times and ~2.3 times higher than 2020 and 2019, respectively [5]. Global hydrogen demand in the transport sector is eventually expected to increase to 8 million tonnes by 2030 [5]. However, considering the global consumption of diesel and gasoline in the transport sector in 2020, it was estimated that approximately ~159.7 million tonnes and ~172.4 million tonnes of hydrogen are required to completely substitute the use of gasoline and diesel, respectively, in the transport sector [10].

Hydrogen can be produced at the central production facility or at the refuelling station site to feed into the vehicles, as shown in Fig. 2. The refuelling network with a central production plant requires additional storage systems and transport facilities to deliver hydrogen at the refuelling stations compared to an on-site production network. However, the reliability of on-site production and cost of hydrogen production, which is affected by the scale of economy, can be the limiting factors of pathway 2. Though fossil-based hydrogen production is a proven technology, further improvement or development of low-cost and environmentally clean production technology is of recent interest [11]. Additionally, high production costs, high storage requirements and costs, high distribution costs and safety concerns during storing, transmission and utilisation are identified as the major limitations of hydrogen utilisation in the transport sector [12,13].

Hydrogen refuelling station infrastructure will be essential for a breakthrough in the commercialisation of a hydrogen-powered automotive sector. The capacity of the hydrogen storage system of hydrogen fuel cell car is typically within the range of ~5–6.3 kg at a pressure of 70 MPa which is sufficient for a driving range of about 400–750 km, depending on the type of car [14,15]. However, on-board hydrogen storage capacity using multiple tanks can be up to ~55–60 kg for the heavy-duty hydrogen truck for a driving range of ~400–560 km, depending on the truck type and payload. The physical dimension of a gaseous hydrogen tank of weight ~167.8 kg and maximum storage capacity of 7 kg H_2 at 35 MPa is ~1.37 m long and ~0.66 m internal diameter [16]. On the other hand, a ~2.34 m long hydrogen tank (~382.4 kg) with an internal diameter of ~0.67 m can store a maximum of 17.2 kg H_2 at 51.7 MPa. Therefore, a small refuelling station of capacity 150–200 kg/day can fill up ~24–40 hydrogen fuel cell cars per day. However, in practical scenario, the station can refuel more cars, as the customers would not let the fuel tank run too low. A SunLine Transit Agency hydrogen refuelling station of 900 kg/day on-site production capacity with 35 MPa can feed 30–35 hydrogen fuel cell buses on normal services [17]. On the other hand, a H2 Mobility brand extra-large size station with 2.5 tonnes/day average hydrogen throughput is enough for fuelling 40 heavy-duty vehicles of capacity 60 kg H_2 per fill, considering 10–15 min for fuelling time [18]. Up to 2021, approximately 729 hydrogen refuelling stations have been in operation throughout the world, which is ~121% higher than

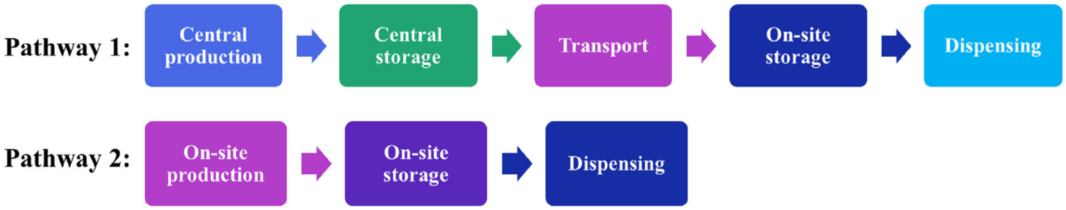


Fig. 2 – Pathways of hydrogen production to consumption in the transport sector.

in 2017 [19]. However, the high capital costs of refuelling stations, along with some technical challenges, including refuelling time and efficient control strategies, remain the major bottlenecks of the hydrogen-based transport sector.

The hydrogen-powered transport sector is a rapidly growing sector, and thereby, extensive research is going on to advance the relevant technologies and reduce associated costs and emissions. Several review papers [20–27] have been published on hydrogen production and storage that comprehensively discuss technologies, covering their process description, major exergy factors, benefits and drawbacks. However, only a few review papers [28–31] on the hydrogen refuelling aspects have been published that covered the deployment of hydrogen refuelling stations in different countries with policies, major components, design and challenges for dissemination with very limited focus on cost analysis. However, several other aspects such as cost economics and hydrogen production-related emissions, delivery technologies, technical challenges of refuelling and their control strategies, refuelling protocols, economic assessment and advanced refuelling stations are required to be reviewed thoroughly to benchmark and improve the technologies, which are the aims of this paper.

In this paper, Section 2 presents technological advancements in hydrogen production along with cost and associated emissions; Section 3 discuses hydrogen storage and delivery technologies with cost economics in relation to vehicle applications; Section 4 presents the progress of hydrogen refuelling technology, which includes the investigation and analyses of temperature rise issue during refuelling with its control strategies, standard refuelling protocols, economic aspects and the emerging refuelling technologies; finally, Section 7 of the paper suggests several future research directions.

2. Hydrogen production

2.1. *Production routes*

Hydrogen can be produced from various resources, including water, organic waste and fossil fuels such as natural gas and coal, through different thermochemical, electrochemical and biochemical processes, as depicted in Fig. 3. The chronological advancement of hydrogen technologies can be found in Supplementary Materials (Fig. S1). Dependent on primary energy source and production process, hydrogen can be classified as grey, blue, green, brown, black and turquoise [32]. Grey hydrogen is produced through steam reforming of natural gas or coal gasification (CG), while hydrogen produced from natural gas or biomass via steam methane reforming (SMR) with carbon capture and storage (CCS) is termed as blue hydrogen. Green hydrogen is produced from the electrolysis of water using renewable electricity. Hydrogen produced from the gasification of brown coal and black coal is known as brown hydrogen and black hydrogen, respectively. The hydrogen produced via methane pyrolysis is called turquoise hydrogen. The process descriptions of hydrogen production technologies have been widely reviewed [20–27], and thereby, in contrast to those previous literatures, this section focuses on the systematic and critical discussions of the current status and prospects of the technologies for future deployment. A comprehensive comparison of different technologies in terms of process (conditions and technical), environmental and economic aspects using the information from the literature is presented in Table 1.

The SMR process is the most established and matured technology, which uses natural gas as an energy source accounting for ~62% of the total global hydrogen production of

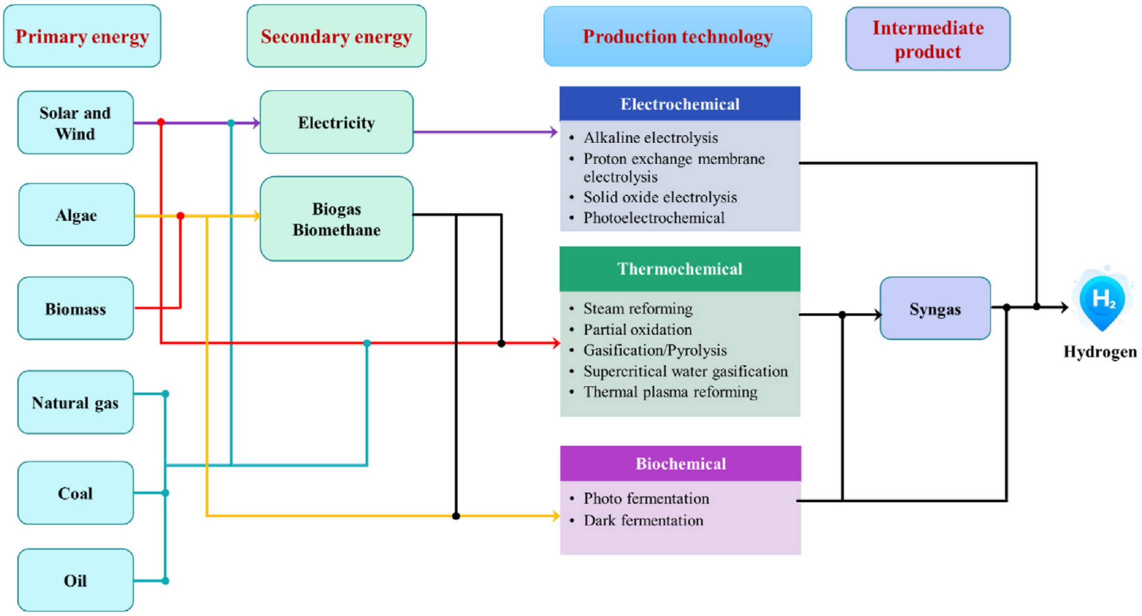


Fig. 3 – Classification of hydrogen production technologies with different energy sources.

Table 1 – Hydrogen production technologies [20–27,35–44].

Technology	Operating conditions	Technical aspects	Economic aspects	Environmental aspects	Energy requirement	Process development
Steam reforming	• Temperature 700–850 °C • Pressure of 3–25 bars • Steam to carbon ratio 3.5	• High-purity H_2 production • Deactivation of catalyst due to coke deposition	• High cost of precious metal catalyst	• CO_2 emissions due to fuel combustion	• High energy consumption	• Mature technology and full commercial phase
Gasification	• Operating temperature 800–900 °C	• Highly efficient process • Corrosion due to slug formation • Deactivation of catalyst • Impurities and tar in H_2	• High capital cost • High operating cost	• CO_2 emissions	• High energy requirement due to high-temperature operation	• Large-scale investigation
Supercritical water gasification	• Temperature 350–600 °C • Pressure 22.12 MPa	• Further optimisation is required for high yield	• High cost of feedstock harvesting • High capital cost due to high temperature and pressure operation	• CO_2 emissions	• High energy requirement	• Lab-scale investigation
Partial oxidation	• Operating temperature 1150–1500 °C	• Deactivation of catalyst due to coke deposition	• Economically attractive due no heat requirement • No cost of catalyst	• CO_2 emissions	• No heat requirement	• Mature technology and full commercial phase
Plasma reforming	• Operating temperature >2000 °C • High degree of dissociation and ionisation	• High electrode erosion • High conversion efficiency	• No costly catalyst required	• CO_2 emissions	• Extremely high energy requirement for plasma generation	• Lab-scale investigation
Proton exchange membrane electrolysis	• Temperature 50–90 °C • Pressure 15–30 bar	• Simplicity in design • High current density	• High cost of membrane and catalyst material	• Acidic environment	• High electricity requirement for electrolysis due to low operating temperature	• Early commercial phase
Alkaline electrolysis	• Temperature 60–90 °C • Pressure 2–10 bar	• Low current density	• No requirement of a catalyst • Low capital cost	• Corrosive electrolyte environment	• High electricity requirement for electrolysis due to low operating temperature	• Full commercial phase
Solid oxide electrolysis	• Temperature 500–1000 °C • Pressure <30 bar	• High energy efficiency • Bulky design of the system	• No requirement of a catalyst • High capital cost	• Environmental-friendly technology	• High heat energy requirement • Low electricity requirement for electrolysis	• Research and development
Photo fermentation	• Ambient conditions	• Slow process	• Low cost due to low operating temperature and pressure	• CO_2 neutral	• High energy requirement for enzymes	• Pilot-scale
Dark fermentation	• Ambient conditions	• Simple reactor design • Low yield and slow process	• Low cost due to low operating temperature and pressure	• CO_2 neutral	• Low energy consumption	• Pilot-scale

94 million tonnes in 2021 [5]. The reforming of naphtha shares about 18% of global hydrogen production. The SMR has a higher feedstock to hydrogen conversion efficiency (Fig. 4(A)) with an energy efficiency of more than 85% (Fig. 4(B)). The steam reforming technology typically consumes 159.6–165 MJ to produce 1 kg hydrogen [33]. CG has been in use for many decades by the fertiliser and chemical industries. CG exhibits similar conversion efficiency to biomass gasification; however, it is benefited with higher energy efficiency than biomass gasification (Fig. 4). Feedstock to hydrogen conversion in gasification technology significantly depends on feedstock type and their physico-chemical properties, as coal-biomass blending has shown significant enhancement in conversion efficiency due to the inherent catalytic species of biomass, which facilitate the water-gas shift reaction [34]. CG typically requires significantly higher energy (~271 MJ/kg H_2 production) compared to that of SMR. Currently, 19% of global hydrogen demand is fulfilled by CG technology. Partial oxidation converts gaseous or liquid hydrocarbon fuels into hydrogen, which can also be employed for hydrogen production from low-value wastes from refinery industries [35]. A partial oxidation-based hydrogen production plant has been in operation at the Pernis refinery in the Netherlands since 2005 [5]. Recently, significant interest has grown in low-emission technologies, as several technologies are at laboratory to pilot-scale investigation stage.

Biomass gasification (i.e., steam gasification, supercritical water gasification (SWG)) has been identified as a potential alternative to CG for renewable hydrogen production due to the abundancy and renewability of biomass. The major challenges of biomass-based hydrogen production include inconsistency of biomass properties and reliable supply chain of biomass. The current global biomass potential is estimated about 181.5 billion tonnes, which is equivalent to ~122 trillion m^3 gaseous hydrogen (approximately 0.672 m^3 gaseous hydrogen can be produced from 1 kg biomass [45]). Therefore, biomass-based hydrogen production can play an important role for shifting from fossil-based hydrogen production. Biomass gasification consumes nearly same energy as of CG. Recently, the Motehydrogen has started building the first commercial biomass gasification-based hydrogen production plant in California, and it is expected to start producing hydrogen in 2024 [46]. Advanced Biofuels Solutions Ltd is expected to start producing 500 tonnes/year hydrogen and 1500 tonnes/year synthetic natural gas from 8000 tonnes/year waste wood gasification in 2022 at Swindon in the UK. Eni has planned to develop municipal solid waste and non-recyclable plastic gasification plant of capacity 100 kg H_2 per year. Hydrogen can also be produced from fermentation process using biomass as feedstock [47–50]; however, this technology is associated with low conversion (Fig. 4(A)) and slow reaction kinetic issues.

Electrolysis is considered as a promising clean hydrogen production technology in the long run. The electrolysis process typically requires approximately 9 L of water to produce 1 kg hydrogen and 8 kg oxygen as a by-product [51]. In order to produce the hydrogen demand of 94 million tonnes in 2021 entirely from electrolysis, it will require 1.3% of global water consumption by the energy sector, indicating the future prospects of the electrolysis process for bulk hydrogen production. It was also estimated that replacing the current global consumption of oil, natural gas and coal (~162,000 TWh per year) with green hydrogen from electrolysis requires 1.8% of the current global water consumption, which is ~25% of current global annual wastewater production [52]. The energy requirement for electrolysis process varies between 26.9 and 90.3 kWh/kg H_2 production, depending on the types of electrolyser (i.e., alkaline, polymer electrolyte membrane, high-temperature solid oxide, and molten carbonate electrolyser) used and operating temperature [33,53]. Typically, the energy efficiency of electrolyser varies between 40 and 70%, depending on the electrolyser types (Fig. 4(B)) with highest efficiency of PEM electrolyser than the other types. However, Hysata's newly designed capillary-fed electrolysis system

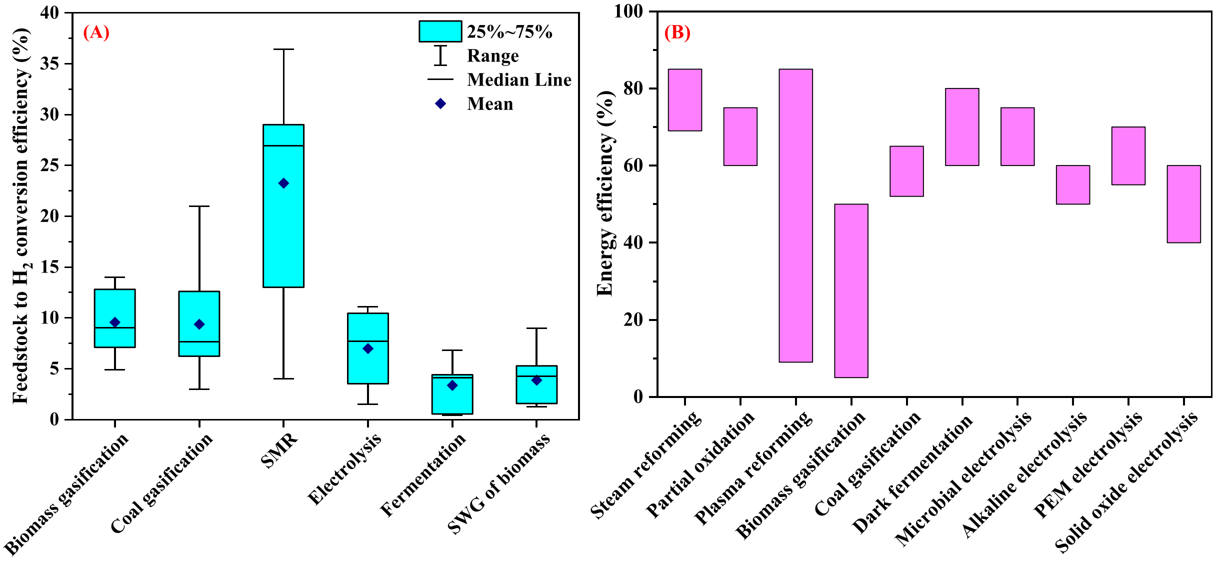


Fig. 4 – (A) Hydrogen production efficiency (data from [33,34,36,49,50,59–65]) and (B) energy efficiency of hydrogen production technologies (data from [4,26,66–70]).

operates at the world's highest efficiency of 95% [54]. Sunfire's solid oxide electrolysis system can achieve 84% efficiency [55], considering the low heating value of hydrogen whereas, the efficiency of FuelCell Energy developed solid oxide electrolyser is reported as ~90% [56].

Currently, less than 0.1% of total hydrogen is produced via electrolysis; however, electrolysis is eventually expected to produce around 51% of global hydrogen demand [5]. In 2021, total installed electrolysis capacity was more than 20 GW, including more than 200 MW addition in 2021 and expected to install 720 GW by 2030 [57]. The Ningxia Baofeng Energy has developed world's largest alkaline electrolyser in China which shared 75% (150 MW) of total addition in 2021 [58]. Baofeng Energy is producing 23.7 million tonnes of hydrogen annually from this plant and has planned to build 150 MW electrolysis plant every year until 2040. Sunfire has built world's largest solid oxide electrolysis system of capacity 2.6 MW in 2022 [57]. Norsk e-fuel will start building a 25 MW solid oxide electrolysis system in 2023 at Mosjøen in Northern Norway. Anion exchange membrane electrolysers are at development stage and so far, Enapter and Alchemr have developed only kW scale unit. In order to achieve the global target, the European Union has set a goal to produce 40 GW of power via electrolysis by 2030 under the European Union Hydrogen Strategy-2020; and, Chile has aimed to produce 25 GW under their National Green Hydrogen Strategy-2020 [5].

2.2. *Production and capital costs*

The major problem with hydrogen utilisation is the need for a production method which has low production costs and associated emissions. The current production cost of hydrogen from SMR and CG is \$0.75–3.49 and \$0.90–2.11 per kg (Fig. 5(A)), respectively, depending on the carbon capture technology. The lower production cost for SMR and CG than other technologies is due to the availability of natural gas and coal. Despite the fact that CG technology has higher capital costs than the steam reforming process, the fuel (i.e., coal) cost is much cheaper than natural gas [32,36], resulting in lower hydrogen production costs in the case of CG. The cost of feedstock shares the majority of plant operating cost. For instance, Katebah et al. [71] reported that material costs contributed ~70% of the total operating costs of SMR plant, whereas Li and Cheng [72] estimated more than 60% of operating costs of CG plant as material costs.

The plant capital cost significantly changes with plant capacity. Fig. 5(B), developed based on the correlation from [73], shows the variation of capital cost with plant capacity. Although biomass gasification has similar capital cost to CG technology, it shows higher production cost due to the lower energy efficiency [69], resulting in an increase in operating cost. The capital costs of SMR and CG technologies also significantly increases when carbon capture system is integrated into the process [22]. CG and SMR without CCS exhibited ~8–16% and ~7–18% less hydrogen production cost, respectively, than with CCS technologies [22,66]. A recent study conducted in China reported ~44.6–60.8% higher hydrogen production cost (\$1.44–2.11 vs \$0.90–1.46 per kg H_2) for CG with CCS compared to that of without CCS unit [74]. Lee et al. [75] recently conducted a techno-economic analysis of on-site hydrogen production from SMR (~15.7 tonne H_2 per year) without carbon capture and compared with that of with carbon capture. The study estimated the levelised cost of hydrogen as \$7.09 per kg H_2 without CCS whereas this was \$7.29–7.73 per kg H_2 with CCS, depending on the method of carbon capture. The hydrogen production cost of the biomass gasification process is lower than that of the fermentation process, even though biomass gasification has lower energy efficiency than the fermentation process. This lower cost is most likely due to the availability and low cost of biomass feedstock and higher hydrogen yield of gasification process which compensates the high energy cost of gasification. The energy requirement for gasification can be lowered through the incorporation of low-cost renewable energy as the heat

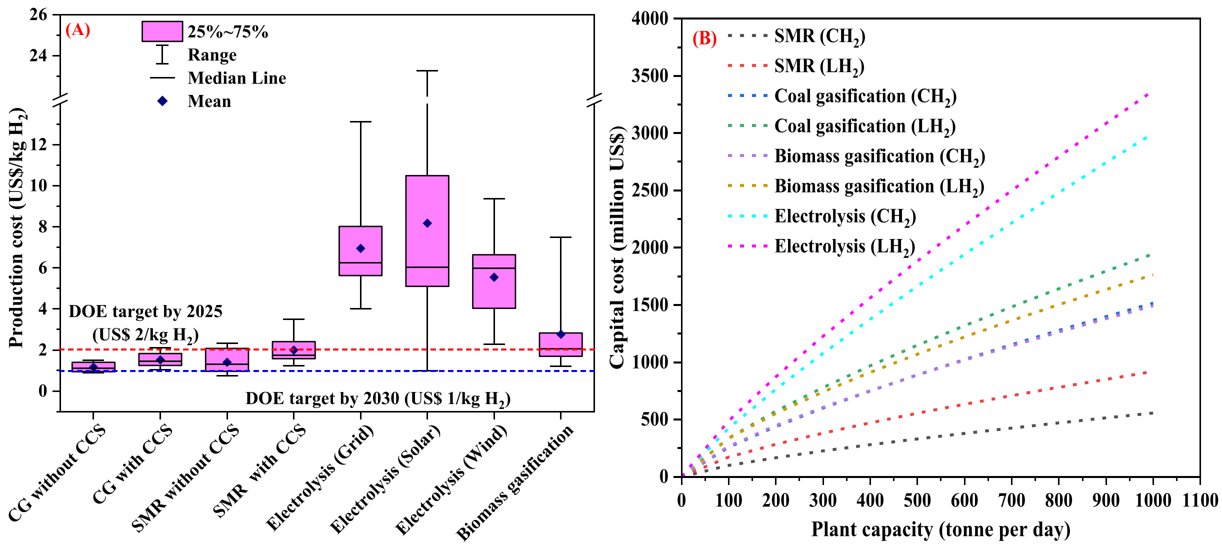


Fig. 5 – (A) Hydrogen production costs per kg of hydrogen for different technologies (data from [22,26,36,39,68,71,74,76–81]) and (B) capital costs variation with plant capacity of different hydrogen technologies (based on the correlation from [73]).

source for the gasification process. However, taking into account the rapid depletion and environmental concerns of natural gas and coal, electrolysis is considered the as leanest way to produce hydrogen.

The capital cost of electrolysis technology widely ranges between \$500 and \$5600 per kW, depending on the types of electrolyser and other installation related costs [5,57]. The current capital cost of alkaline, PEM and solid oxide electrolysers are estimated as \$500–1400, \$1100–1800 and \$2800–5600 per kW, respectively. However, a much lower price of \$750-\$1300 per kW can be obtained in China due to the lower cost of equipment, engineering, acquisition and construction in China. The capital cost of electrolysis is still significantly higher than other technologies. The cost of electrolyser is expected to reduce to \$440-\$500 per kW by 2030 and ~\$300 per kW by 2050. The current cost of hydrogen production via electrolysis varies widely between \$0.98 and \$23.27, with a mean value of \$6.62 per kg H_2 (Fig. 5 (A)), depending on the energy source required for the electrolysis process. The cost of hydrogen production using grid electricity is lower than solar electricity from small scale solar PV; however, the cost is comparable with that of wind electricity. The hydrogen production cost via electrolysis is higher than other technologies and still far away from the DOE 2025 target. This could be due to the higher cost of electricity used in the electrolysis process, as electricity shares about 90% of the total operating cost for the electrolysis [32]. The hydrogen production cost via the electrolysis method can be reduced by optimising the capacity of renewable energy source, reducing the component cost, hybridising solar/wind with other low-cost sources and effectively integrating into the electrolysis process. The cost can be further reduced if the benefits from carbon credit is considered in the estimation. Therefore, the cost of hydrogen production significantly varies from technology to technology as well as from country to country. This variation in production cost is likely due to the discrepancy in the assumption of capital and operational cost, system configuration and country-specific energy mix, taxes and subsidy policies.

2.3. *Emissions associated with production*

Even though hydrogen is considered as a clean fuel, the production process of hydrogen may associate with significant GHG emissions. As of 2021, approximately 99% of global hydrogen is produced from fossil fuels, including natural gas, coal and oil via naphtha reforming, which emit a substantial amount of CO_2, indicating the unsuitability of these routes towards net zero emissions targets. Total CO_2 emissions from hydrogen production activities around the world were about 0.9 billion tonnes in 2021 [5]. Therefore, hydrogen production from low-emission technologies is expected to increase to 24 million tonnes by 2030. CG, which contributes about 19% of global hydrogen demand, is associated with a huge GHG emissions and ranging between 11.5 and 32.2 kg CO_{2}-eq/kg H_2 without any CCS system, as shown in Fig. 6. Despite the high hydrogen production and energy efficiency of SMR, the key issue of this process is its high GHG emissions (7.1–13.8 kg CO_{2}-eq/kg H_2) [82,83]. Taking the average of emissions values from the literature, it is estimated that CG emits ~2 times higher GHG emissions (22.7 vs 11.2 kg CO_{2}-eq/kg H_2) than SMR technology. Kothari et al. [84] also reported ~4 times higher CO_2 emissions for hydrogen production from CG compared to SMR. Additionally, CO_2 emissions from partial oxidation of hydrocarbons was estimated as ~2.4 times higher than SMR (29.3 vs 12.4 kg CO_2/kg H_2). However, the integration of CCS technology with fossil-based hydrogen production approaches can reduce the GHG emissions by storing and reusing CO_2 instead of emitting to the atmosphere. For instance, Verma and Kumar showed a reduction of GHG emissions from 18 kg CO_{2}-eq/kg H_2 to 0.91 kg CO_{2}-eq/kg H_2 when carbon was captured using CCS technology after CG [85]. Li et al. [74] reported ~52.3–74.6% carbon footprint reduction with CCS technologies for hydrogen production from CG in China. Dufour et al. [86] exhibited ~67% less GHG emissions for SMR with CCS when compared to SMR without CCS. However, fossil-based technologies with CCS for hydrogen production are in the development stage and require additional capital and maintenance costs.

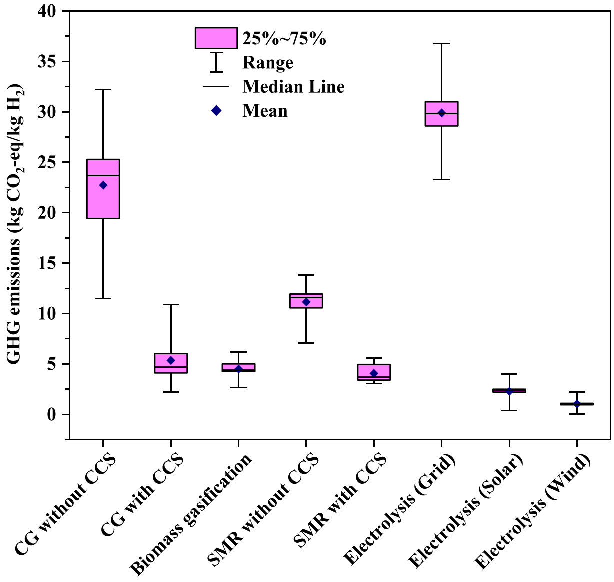


Fig. 6 – GHG emissions from major hydrogen production technologies (data from [62,72,74,82,83,87–93]).

Several companies have retrofitted hydrogen production plant with CCS system and some of them have started building new plant equipped with CCS. As of 2022, seven hydrogen production plants with CCS are in fully operation and another 33 plants are in different stages of development particularly in UK, USA, Europe and Canada. Air Products has retrofitted two SMR plants with CCS at Valero Refinery in Port Arthur, Texas which are in operation since 2013 to capture ~1 million tonne CO_2 every year [94]. Air Products is also building world's largest blue hydrogen production facility of capacity more than 21.24 million m^3 hydrogen per day with CCS in Louisiana, USA which will be in operation in 2026 with a target to sequester more than five million tonne of CO_2 per year. Additionally, another new hydrogen production facility from natural gas with CCS is expected to operate in Canada in 2024 which will capture >95% of CO_2 produced in the plant. Shell Canada retrofitted a bitumen refinery plant located in Alberta, Canada with CCS in 2015 and until 2021 captured six million tonnes of CO_2 produced from SMR of 900 tonnes hydrogen per day production capacity [95]. Shell is expected to build another new hydrogen facility with CCS in Alberta which will capture 0.75 million tonnes CO_2 per year from the Scotford refinery and chemicals plant. Great Plains synfuel plant in North Dakota, USA is producing 1.3 kilo tonnes of hydrogen per day since 2000 from brown CG with CCS whereas, Coffeyville petroleum coke gasification plant in Kansas, USA started hydrogen production in 2013 and is producing 200 tonnes of hydrogen per day with CCS [96]. Under the Alberta Carbon Trunk Line project in Alberta, Canada, NorthWest is producing ~240 tonnes of hydrogen per day at the Alberta Sturgeon Refinery plant from asphaltene residue gasification with CCS and ~800 tonnes of hydrogen per day at the Agrium fertiliser facility from SMR with CCS.

Biomass gasification-based hydrogen production technology shows a GHG emissions ranging between 2.67 kg CO_2-eq/kg H_2 and 6.2 kg CO_2-eq/kg H_2, which is lower than CG without CCS but similar to CG with CCS. The low emissions from biomass gasification could be due to the compensation of CO_2, produced from the gasification process, by the amount of CO_2 required for the biomass growth. The electrolysis process, using electricity from the grid, emits extremely high GHG emissions (23.3–36.8 kg CO_2-eq/kg H_2) [82,83]. The higher GHG emissions from electrolysis using grid electricity is due to the fossil fuel-based electricity generation. Renewable electricity-based electrolysis exhibits significantly lower GHG emissions than grid electricity-based electrolysis process, even lower than the gasification and SMR with CCS. Therefore, energy-mix for electrolysis requires to be optimised considering the high cost and low emissions from the renewable electricity.

3. Hydrogen storage and transport

3.1. Storage and transport technologies

In contrast to gasoline, gaseous hydrogen requires four times more volume to store equivalent energy to that of gasoline [97–99]. Therefore, one of the key challenges for the global adoption of hydrogen and enabling the global hydrogen economy is the development of a safe, low-cost, efficient and high-density hydrogen storage and transport technology [26,100]. Stored hydrogen can be used for transport sector, portable power, stationary power or system backup applications. Hydrogen can be stored in a solid, liquid or gaseous state through several technologies, including gas compression, liquefaction and solid-state storage [101], as shown in Fig. 7. Hydrogen can be delivered from the central production facility to the refuelling station through various transport modes, such as pipelines, tube trailers, rail, trucks and ships, depending on the storage state (i.e., gaseous, liquid and solid) and availability of the transport mode.

Gas compression and liquefaction of hydrogen are well-established technologies for hydrogen storage and are widely used in commercial sectors; however, these technologies are associated with a number of issues, such as the high-pressure requirement for compressed hydrogen, a high risk of leakage and explosion, boil-off losses and high energy requirements for liquefaction [101,102]. In order to address the issues of liquid or compressed gas hydrogen, Lawrence Livermore National Laboratory has developed cryo-compressed hydrogen storage system that can store hydrogen as cold compressed hydrogen, liquid hydrogen or hydrogen in saturated liquid and vapor phase at cryogenic temperature and 250–350 atm pressure [103,104]. In underground storage, hydrogen is typically stored in naturally occurring porous rocks such as depleted natural gas deposits, depleted oil deposits and aquifers and in artificially created salt caverns [105]. Underground storage of hydrogen requires a unique geological structure and ample space and is linked with safety hazards, project economics, legal and other technical issues [106,107]. The UK stores about 830 tonnes of hydrogen at 5 MPa in three underground salt caverns of 400 m depth for producing ammonia and methanol by nearby industrial plants [105]. The USA has two underground domal salt caverns of capacity 2560 tonnes and 3720 tonnes hydrogen in Texas operated by Conoco Phillips and Praxair, respectively. Hydrogen storage via compression consumes ~1.7–6.4 kWh/

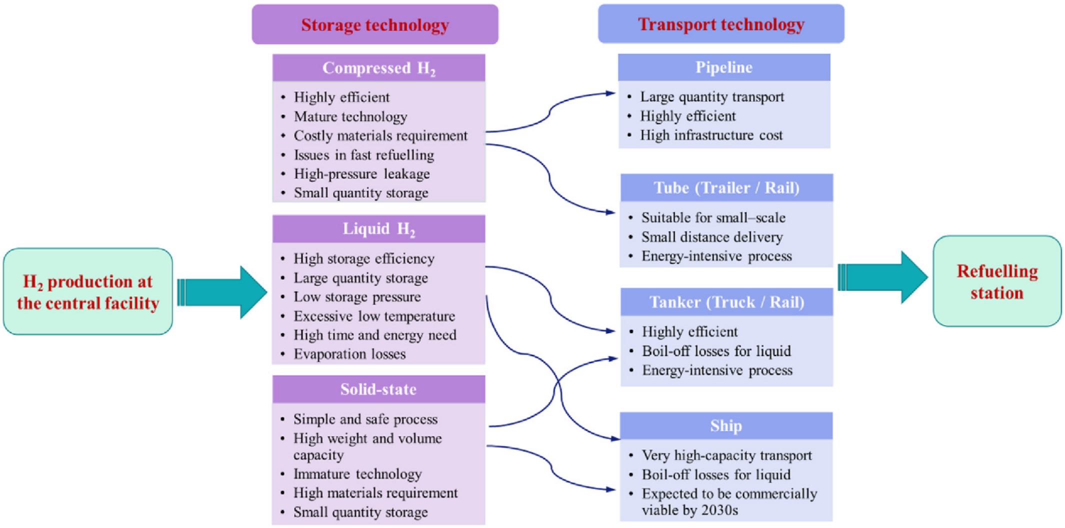


Fig. 7 – Hydrogen storage and transport to refuelling station for vehicles.

kg H_2 with an energy storage efficiency of ~81–95%, depending on the types of compressors [70,108]. On the contrary, liquefaction of hydrogen requires ~10–13 kWh/kg H_2, equivalent to an energy efficiency of ~61–70%, depending on the size of liquefaction unit.

Adsorption/absorption-based solid-state hydrogen adsorption technologies have received significant research interest as a long-term option in recent times due to their high reliability, volumetric efficiency and safety [8,102]. Hydrogen can be stored at a pressure between 1 and 10 MPa, depending on the adsorbent properties [109]. Hydrogen absorption via chemical reaction using various metals, their alloys and metal hydrides at ambient conditions showed excellent storage capacity (0.7–18 wt%) [110–112]; however, the lack of reversibility and complexity in the extraction of hydrogen due to strong metal-hydrogen interaction are key challenges [113]. Additionally, a significant amount of energy is required to release the hydrogen and estimated about 2.8–10.3 kWh/kgH_2, depending on the metal [114]. The storage of hydrogen in liquid organic hydrogen carriers is still a relatively new and has numerous advantages over conventional storage systems [115–117]. However, they require high temperatures and are associated with high catalyst costs and carrier costs. Metal organic framework (MOF) exhibited up to 7.5 wt% hydrogen uptake, while zeolite showed a maximum hydrogen uptake of 2.6 wt% [118]. The major challenges of MOFs are their poor thermal conductivity and extremely high material cost. Additionally, the formation of metal hydrides, very low-temperature operation and possible environmental pollution from metal at high concentrations are the limiting factors of MOFs-based hydrogen adsorption process [119]. The hydrogen uptake of activated carbon was reported between 0.5 and 8.9 wt%. Petroleum and coal-derived carbon nanotubes, nanosphere and nanofibers have experimentally exhibited 3–7 wt% hydrogen uptake [120]; however, the preparation of these carbon-based materials is associated with fossil fuel utilisation and energy-intensive processing, and the materials contain mostly mesopores and macropores. Hydrogen uptake of carbon nanomaterials can also be further enhanced up to 20 wt% by doping transition metals; metal doping can form metal hydrides, leading to poor reversibility of adsorption [121]. Therefore, considering the needs of modern society and emerging issues, the development of a new, low-cost and environmentally benign material-based high-performance ambient temperature hydrogen storage system is essential for the future hydrogen economy.

Most recently, lignocellulosic biomass-derived carbon-based materials have emerged as a promising material for hydrogen storage due to their renewability, high surface area, high surface functional groups and abundant micropores with no metal hydrate formation. Lignocellulosic biomass is a low-cost and sustainable material, which consists of 30–50% cellulose, 15–35% hemicellulose and 10–30% lignin [122]. Of the 181.5 billion tonnes of biomass currently being produced in the world, only about 8.2 billion tonnes are used for paper and pulp, biorefinery, fodder, energy and material purpose, indicating a huge potential for biomaterial-based hydrogen storage systems [123]. Paper and biorefinery industries use cellulose and hemicellulose, leaving more than 100 million tonnes of lignin waste per year, which can also be used due to their high pore structure, specific surface area and conductivity [124–126]. Porous carbon materials produced from biomass exhibited promising hydrogen storage capacities with about 3.8–6.4 wt% hydrogen uptake [127–129].

The benchmarking of the hydrogen uptake capacity of different materials with the DOE target (Fig. 8) suggests that carbon-based materials have already achieved DOE's 2025 target. However, the hydrogen storing capacity of these materials was estimated at cryogenic temperature and high pressure, which does not meet the hydrogen storage target set by the DOE, USA. At ambient temperature, hydrogen uptake capacity drops significantly below the DOE target. In this case, metal hydrides can be a promising alternative, though they also have some other challenges. However, further in-depth research is required to increase the capacity to justify the associated challenges.

3.2. *Hydrogen storage costs*

Hydrogen storage at central production facility or at refuelling station (i.e., off-board) and in the vehicles (i.e., on-board) is a challenge for a wide driving range. Therefore, the levelised cost of storage is one of the critical factors which is required to be taken into account when selecting a suitable method for hydrogen storage [135]. According to a report published in 2020 by BloombergNEF [136], the levelised cost of small-scale hydrogen storage for a short period (i.e., daily) as a pressurised gas is about \$0.19/kg H_2, which increases up to \$1.9/kg H_2 for large volume seasonal storage. Abdin et al. [137] estimated the levelised storage cost of compressed hydrogen for a 5000 tonnes storage system capacity in 2020 and the costs for a daily and 4-monthly storage cycle were reported as ~\$0.33 and ~\$25.20 per kg of H_2, respectively. On the contrary, daily hydrogen storage cost in salt caverns was ~\$0.14/kg of H_2. Tzimas et al. [138] also reported a significantly higher storage cost per kg H_2 for a long-term storage cycle (1 month)

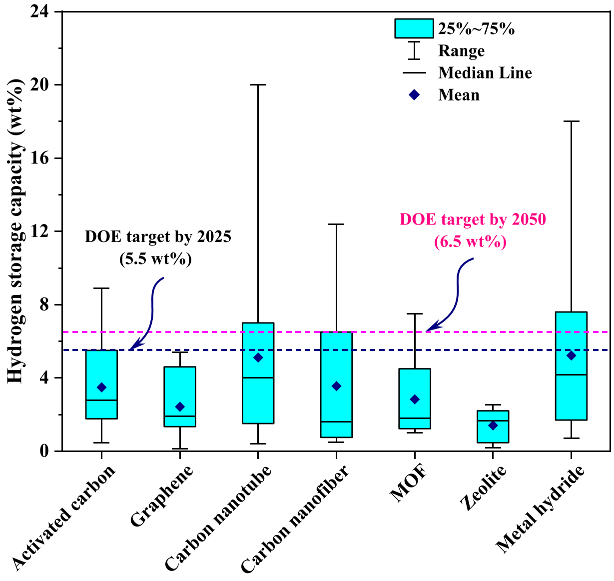


Fig. 8 – Benchmarking of hydrogen uptake capacity of different materials with DOE target (data from [112,118,130–134]).

compared to that of short-term storage (1–3 days) for both compressed gaseous and liquid hydrogen storage. Hydrogen storage cost per kg H_2 also reduced with an increase in storage capacity. The cost of small-to-medium volumes of liquid hydrogen storage for days-to-weeks is approximately $4.57/kg H_2. A well to wheel (WTW) economic analysis in the context of India showed that the levelised cost of underground storage was less when compared to other technologies and ordered as underground < compressed hydrogen < liquid hydrogen < metal hydride [139]. Hawkins [140] also reported a similar order as the storage cost was $0.12–0.30/kg H_2, $1.00–1.50/kg H_2, $0.15–0.60/kg H_2 and $0.40–4.00/kg H_2 for underground, liquid hydrogen, compressed hydrogen and metal hydride storage method, respectively, depending on the storage capacity and duration. The lower levelised cost of underground hydrogen storage was due to its significantly lower capital and operating costs compared to those of other storage systems. The capital cost of underground hydrogen storage includes the costs of the compressor, well construction, site preparation and cushion gas. The cushion gas shares more than 79% of the capital cost for the depleted gas deposit (79%) or saline aquifer (88%), while the site preparation accounts for 72% of the capital cost in a salt cavern, because salt cavern is made artificially [141]. The total capital cost of underground hydrogen storage was estimated to be ~$40.1–89.6 million, depending on the storage capacity (2486–2868 tonnes) and storage site types (salt caverns, depleted oil and gas deposits, saline aquifers and hard rock caverns) [142]. However, Papadias and Ahluwalia [143] demonstrated that the capital cost of underground hydrogen storage could be significantly reduced by increasing the storage capacity. Lord et al. [142] found depleted oil and gas deposits as the most economically attractive ($1.23/kg H_2) storage system compared to other underground storage options such as aquifers (1.29 $/kg H_2), salt caverns (1.61 $/kg H_2) and hard rock caverns ($2.77/kg H_2). Chen et al. [141] also estimated the lowest hydrogen storage cost for depleted gas deposits ($1.15//kg H_2). However, higher storage cost was observed for saline aquifer ($3.28//kg H_2) than salt cavern ($2.76//kg H_2), which contradicts the findings of Lord et al. [142]. This could be due to the variation in assumptions considered in the economic model.

Hydrogen storage system cost significantly depends on the types of storage technology, storage mode (i.e., onboard or off-board), storage capacity and the production volume of the storage system. Several companies, including Linde, NPROXX and INOXCVA have manufactured off-board bulk liquid hydrogen storage systems and reported the system cost between ~$39 and ~$115 per kg H_2, depending on the storage capacity (~0.8–4.8 tonnes of H_2) of the system [144]. According to the on-board storage system breakdown cost analysis, carbon fiber is the largest cost category which shares >45% of the system cost, respective of hydrogen storage capacity of the system (i.e., light-duty or heavy-duty storage) and system production volume [145–147]. The costs of 5.6–10.4 kg on-board compressed and liquid hydrogen storage systems considering 500,000 units/year production was estimated as ~$12–23.7 and ~$5–10 per kg H_2, respectively [148–150]. An analysis, conducted in 2021, reported that the cost of on-board storage system with an available storage capacity of 60 kg H_2 at 70 MPa and 100,000 units/year system production volume is $383 per kg H_2 for heavy-duty Class 8 long haul trucks [144]. Ahluwalia et al. designed an on-board liquid hydrogen storage system of 86.8 kg useable H_2 capacity with a 1057 km driving range for Class-8 heavy duty trucks and estimated the cost of the system in 2016 considering a production volume of 100,000 units/year [151]. The cost of the storage system was $174–183 per kg H_2 which is ~53% lower than the cost of compressed hydrogen storage system for same vehicle as reported by Houchins and James [144]. The costs of 40 kg on-board cryo-compressed hydrogen storage system with 5000 units/year for fuel cell bus were $333 and $366 per kg H_2 for 35 MPa and 50 MPa storage, respectively which was significantly lower than that of compressed hydrogen storage system ($466 per kg H_2) [152]. Paster et al. also reported lower cost of cryo-compressed storage system compared to compressed hydrogen storage but higher than liquid storage [150]. The storage system cost significantly reduced with an increase in production of storage units [146,147] due to economy of scale in capital cost. However, the on-board hydrogen storage system cost of all available technologies except liquid storage for light-duty hydrogen fuel cell electric vehicles (HFCEVs) is still far away from the DOE target as shown in Fig. 9.

3.3. Hydrogen transport costs

Hydrogen transport from a central production facility to the refuelling station play an important role in the economic feasibility of a hydrogen-based transport system. The transport through pipelines is considered as the most economical mode of gaseous hydrogen transport at large quantities due to the low maintenance cost, while trucks and rails are considered feasible for liquid hydrogen transport, depending on the availability [148,153–155]. Hydrogen pipelines typically are of 0.25–0.3 m in diameter and operate at 1–3 MPa. Tube trailers are designed to transport 300–500 kg hydrogen at 20–60 MPa while tankers for road transport and ship tankers can carry

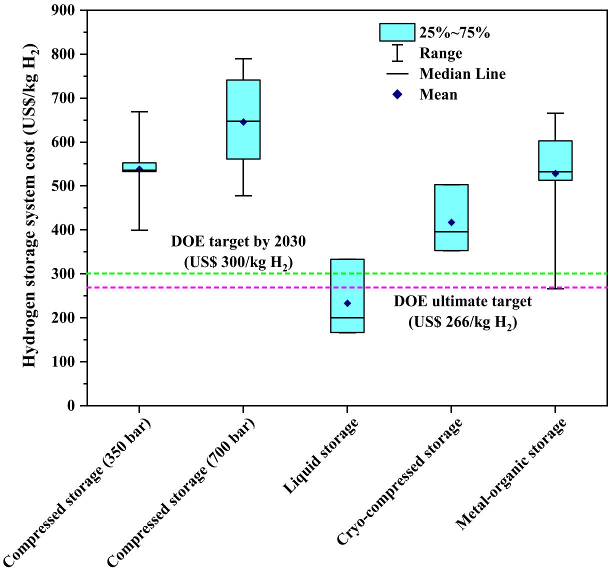


Fig. 9 – On-board hydrogen storage system (yearly production: 500,000 units) cost for light-duty HFCEVs (data from [148–150]).

400–4000 kg and 10 million kg hydrogen, respectively [140]. Currently, over 5000 km of pipelines are in operation for hydrogen transport, of which more than 90% are located in the USA and Europe with the longest pipeline of 400 km owned by Air Liquide in Europe [156]. Germany is transporting hydrogen with a flow rate of 8900 kg/h at 2 MPa pressure through 210 km pipeline. The cost of hydrogen transport through pipelines was reported as ~$1.6/kg H$_2$ in North Africa for a distance of 3300 km and $0.59/kg H$_2$ in the USA in 2017 [148]. In the UK, the transport cost of hydrogen through the pipeline (gaseous hydrogen), tube trailer (gaseous hydrogen), truck (liquid hydrogen) and ship (liquid hydrogen) was estimated to be $0.1–1/kg H$_2$/100 km, $0.5–2/kg H$_2$/100 km, $0.3–0.5/kg H$_2$/100 km and $1.8–2/kg H$_2$/100 km, respectively [140]. The cost of liquid hydrogen transport from Africa to Europe via ship was expected as $3.1–3.6 per kg H$_2$ in 2017 [148]. Simbeck and Chang [157] estimated higher per km transport cost of per kg hydrogen through gas pipeline followed by gas tube trailer and liquid tanker truck. However, hydrogen transport cost per kg H$_2$ per km depends on the hydrogen flow rate through pipeline and distance to be delivered. The transport cost ($/kg H$_2$/km) of both gaseous and liquid hydrogen via truck, rail or ship reduces sharply up to a certain distance following exponential relationship with distance and then decreases insignificantly with distance (Fig. 10). For gaseous hydrogen transport, truck is cheaper than rail for a short distance (<300 km) but expensive for long distance (>300 km) transport [148]. Liquid hydrogen transport via rail can be cheaper than truck for a large quantity of hydrogen due the large capacity per railcar. Transport distance has insignificant impact on gaseous hydrogen transport through pipeline; however, significantly reduces from $320.4 per kg H$_2$ to only $0.1 per kg H$_2$ when the hydrogen flow rate is increased from 5 kg H$_2$/h to 45,359 kg H$_2$/h [148]. Therefore, gas pipelines can be effective to deliver hydrogen for both short and long distance with high flow.

3.4. *Emissions associated with storage and transport*

Hydrogen storage and transport via road using diesel or gasoline as fuel is associated with a considerable amount GHG emissions. Li et al. [91] investigated the effect of different hydrogen transport options on the overall GHG emissions of hydrogen production from CG with or without CCS in China. The study estimated that the transport of 1 kg hydrogen produces 36.9 gCO$_2$-eq and 12.3 gCO$_2$-eq for a road transport distance of 300 km and 100 km respectively, irrespective of CCS system. Considering the emissions from storage, the total GHG emission was estimated about 718 gCO$_2$-eq/kg H$_2$ for 100 km distance. The sensitivity analysis for a distance range of 100–500 km suggested that GHG emissions increased linearly with the increase in transport distance. Road transport of hydrogen exhibited a higher GHG emissions followed by rail transport and pipe transport for any transport distance. Hren et al. [158] compared the GHG emissions of different storage and transport pathways of gaseous and liquid hydrogen and reported that the storage and transport can contribute 35% of total GHG of hydrogen production pathway. Considering the underground salt cavern storage, transport of gaseous hydrogen via pipeline emitted 50–500 gCO$_2$-eq for a transport distance of 100 km and this was 83–665 gCO$_2$-eq for road transport. The higher GHG emissions for road transport is associated with the higher compression energy consumption in the case of road transport. Liquid hydrogen storage in tank and transport via road produced 336–3332 gCO$_2$-eq/kg H$_2$ for the same distance as of gaseous hydrogen. Akter et al. [159] also estimated a lower GHG footprint for pipeline transport route of gaseous hydrogen compared to that of road transport. With salt cavern storage, pipeline transport emitted ~108 gCO$_2$-eq/kg H$_2$ whereas road transport using tube trailer produced ~361 gCO$_2$-eq/kg H$_2$ for 100 km distance. Liquid hydrogen with tank storage is associated with ~255 gCO$_2$-eq/kg H$_2$.

4. Hydrogen refuelling into vehicles

4.1. *Refuelling process and temperature rise during fuelling*

A refuelling station with externally supplied hydrogen consists of a compressor, storage tank, dispenser, electrical and piping control and a safety monitoring system. The compressed hydrogen refuelling process is similar to that of CNG, i.e., the vehicle is filled with overflow from a high-pressure stationary storage tank. The volume and pressure of the

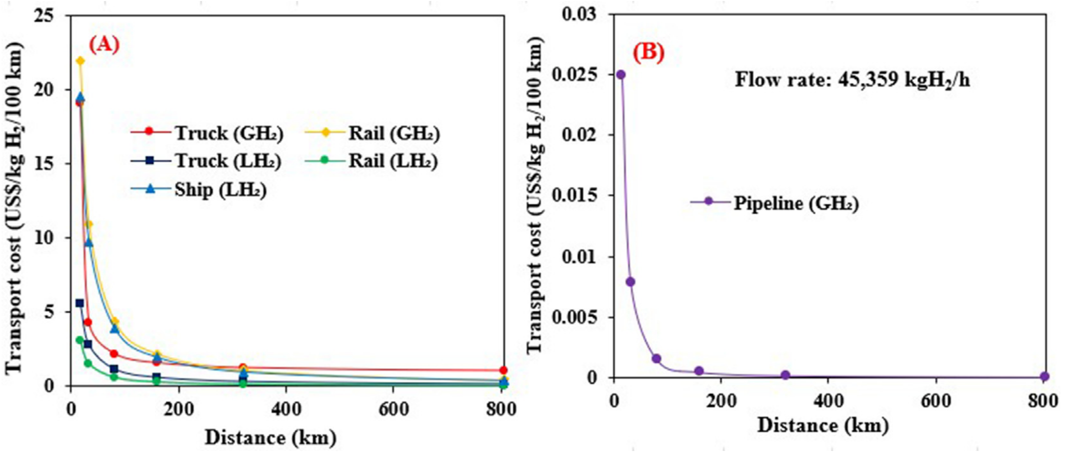


Fig. 10 – Hydrogen transport cost via different technologies (data from [148]).

stationary storage tank are usually kept at least 10 times the maximum expected volume of the fuel tank and 1.2 times the target pressure [160]. According to the SAE J2601 standards, refuelling stations follow a standardised refuelling procedure [160].

- The vehicle is first connected to a high-pressure stationary storage tank through the dispenser.
- The high-pressure stationary storage tank is opened for a short-time initial gas surge, which makes the refuelling line pressure balance.
- This balance facilitates an estimate of the volume of the fuel tank in the vehicle.
- The target pressure is then calculated using these data, and the fuel tank is filled up to the target pressure.
- The intended filling level is typically maintained as <100% in order to avoid overfilling.

In order to be competitive with internal combustion engine vehicles (ICEVs) refuelling, manufacturers of automobiles intended to keep the time for hydrogen refuelling into passenger cars not more than 3 min [160,161], indicating the requirement for fast hydrogen refuelling for the commercialisation of HFCEVs. Therefore, the Society of Automotive Engineers (SAE) developed the SAE J2601 protocol for refuelling 5–7 kg of hydrogen into HFCEV within 3–5 min [162]. The SAE J2601-2 suggested ~20–30 kg of hydrogen refuelling into heavy-duty vehicles within 5–10 min.

During hydrogen refuelling operation, fast filling of high-pressure hydrogen significantly increases the temperature in the hydrogen tank due to the isenthalpic expansion of the high-pressure gas in the dispenser throttling system and fuel tank. This phenomenon of temperature change in throttling is termed as Joule-Thomson effect. The variation in hydrogen refuelling temperature affects the ramp rate of the cylinder and thereby refuelling rate [163]. The increase in temperature during refuelling causes several issues, including the potential failure of the hydrogen storage tank and a reduction in fuel volume in the fuel tank (up to 20% compared with CNG vehicle) with a subsequent reduction in the driving range of the vehicle [164,165]. The temperature rise within the tank after refuelling can be estimated using a simplified thermodynamic model [166–168].

The effects of several process parameters (i.e., initial temperature, initial pressure, mass filling rate and inlet temperature) on the temperature rise have been investigated experimentally and using CFD simulations on Type III and Type IV cylinders. Type III cylinders are made of an inner metal liner which is wrapped with carbon fibre, while Type IV hydrogen cylinders have a non-metallic inner liner made of high-density polyethylene or composite materials with an outer wrapping of carbon fibre [169]. Considering the several benefits such as light-weight and high strength of Type III and Type IV cylinders over Type I and Type II cylinders entirely made of metal, Type III and Type IV cylinders are considered as the most suitable types for automotive applications [170]. For instance, Type IV cylinders have up to 70% less weight and can deliver hydrogen into vehicles at more than 4 times higher capacity compared to those of Type I cylinders.

- **Effects of initial pressure:** An increase in the initial pressure of the fuel tank/cylinder negatively impacts the temperature rise. For instance, Liu et al. [166], Li et al. [171], Kim et al. [172], and Zheng et al. [169,173] performed experimental and CFD simulation studies to investigate the effects of initial cylinder pressure on temperature rise and observed a linear decrease of temperature rise with an increase in initial pressure of the cylinder/fuel tank with a coefficient of correlation (R^2) >0.923 as depicted in Fig. 11(A). A decrease in temperature with an increase in initial pressure is most likely due to flow-restricted hydrogen filling at high initial pressure, resulting in a lower temperature rise. Miguel et al. [174] performed CFD analysis of refuelling for Type III and Type IV cylinders. The initial cylinder pressure had no significant effect on the temperature rise at a fuel delivery temperature of −40 °C. The increase in temperature rise can also be correlated with the instantaneous pressure of the fuel tank/cylinder [166,175,176]; the temperature rise increases nonlinearly with an increase in cylinder pressure with a decreasing rate, and thereby their correlation can be expressed by the logarithmic correlation as shown in Fig. 11(B). A slightly poor fitting was observed in Sadi et al. [175] compared to other studies as can be justified by the comparatively lower coefficient of correlation (R^2) values. Sadi et al. [175] thermodynamically modelled the behaviour of a hydrogen

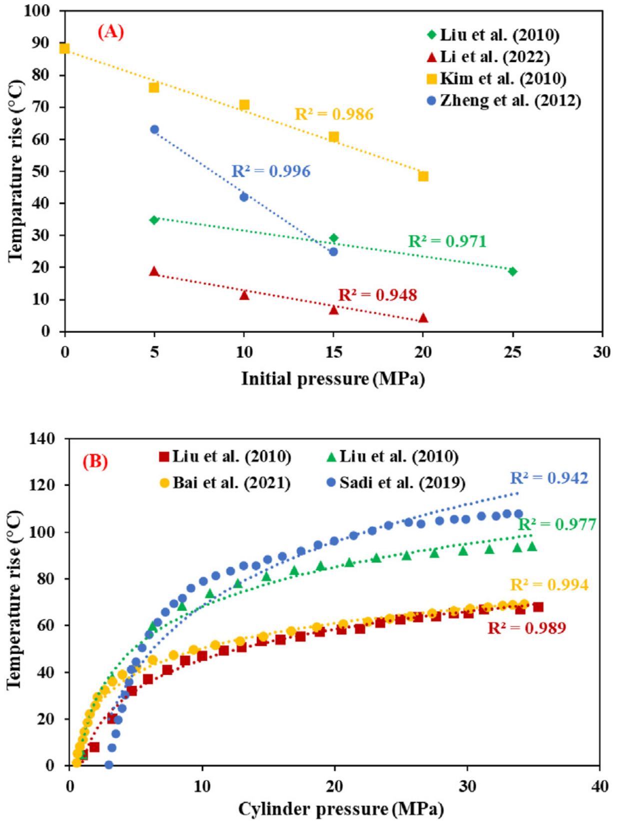


Fig. 11 – (A) Impacts of initial pressure on temperature rise and (B) Correlation between temperature rise and instantaneous cylinder pressure.

cylinder during refuelling, considering the same configuration as experimentally described in Liu et al. [166] with an assumption of adiabatic condition. The slight difference in model temperature rise data from the experimental data could be caused by the model assumptions as the temperature rise is higher for the adiabatic condition during refuelling compared to that of non-adiabatic condition [175].

- **Effects of initial cylinder, inlet gas and ambient temperature:** The initial temperature of the cylinder, inlet gas temperature and the ambient temperature influence the maximum temperature rise in the cylinder during hydrogen refuelling. Miguel et al. [174,177] investigated the thermal behaviour of Type III and Type IV tanks and the role of initial temperature during the refuelling of hydrogen and reported a decrease in temperature rise with an increase in initial temperature, as depicted in Fig. 12. According to the data from these studies, the correlation between temperature rise and initial temperature can be expressed by a linear relationship with the $R^2 > 0.925$. Li et al. [171] and Melideo and Baraldi [178] also noted a linear decrease in temperature rise with an increase in initial cylinder temperature. On the contrary, the inlet gas temperature and ambient temperature exhibited opposite effects on the temperature rise of gas in the cylinder as of the impact of the initial temperature of the cylinder. The temperature rise during hydrogen refuelling increases with an increase in both the inlet gas temperature and ambient temperature and there is a linear correlation between them [169,171,173,179]. Zhao et al. [179] and Zheng et al. [173] observed a 0.3 °C increase in temperature rise during refuelling when the ambient temperature was increased by 1 °C. In another study by Liu et al. [166] reported a small effect of ambient temperature on the temperature rise in the cylinder. Guo et al. [180] investigated the change in cylinder gas temperature during hydrogen refuelling and recorded a 1 °C increase in cylinder gas temperature when the inlet temperature was increased by 1 °C. Refuelling at higher inlet temperature reduces the density of hydrogen and thereby lowers the amount of hydrogen mass into the cylinder, which increases the temperature rise in cylinder gas [167,181].
- **Effects of mass flow rate:** The refuelling time can be shortened by increasing the hydrogen flow rate, which reduces the time for heat transfer from hydrogen and cylinder, leading to an increase in cylinder temperature, even beyond the upper limit of 85 °C [182]. Zhao et al. [179], Li et al. [171], Miguel et al. [177] and Liu et al. [166] noted an increase in temperature rise with an increase in mass flowrate, as shown in Fig. 13. The temperature rise increases nonlinearly with an increase in mass flow rate; however, the temperature rise rate has a decreasing trend. The data from these studies exhibited a logarithmic correlation between mass flow rate and the maximum temperature rise. Zhao et al. [179] performed the numerical simulations for the mass flow rate of 12 g/s, 18 g/s, 25 g/s, 40 g/s, 65 g/s, 130 g/s and 200 g/s; however, they observed an insignificant difference in temperature rise between the flow rate of 130 g/s and 200 g/s.
- **Effects of other parameters:** Other factors such as cylinder types and dimensions can affect the heat transfer and, ultimately, the temperature rise. Miguel at el [174,177]. Experimentally investigated refuelling in Type III and Type IV tanks at different initial temperatures and hydrogen flow rates and observed a higher gas temperature in Type IV tanks. The heat transfer in a Type III tank is high compared to that of Type IV due to the higher thermal conductivity of the aluminium liner (Type III tank) than the plastic liner (Type IV tank), resulting in a lower gas temperature in Type III tanks. Zheng et al. [169] investigated the effects of cylinder dimensions on temperature rise and observed a non-uniform temperature distribution in the larger cylinder compared to the smaller one.

4.2. *Safety standards and protocols*

In order to tackle the safety issues and meet the technical requirements to be competitive with conventional refuelling, the Society of Automotive Engineers developed the SAE J2601 protocol for light-duty gaseous HFCEVs [183]. The SAE J2601

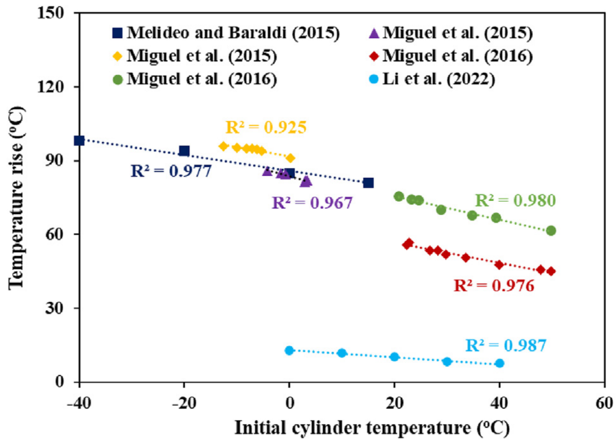


Fig. 12 – Effects of initial cylinder temperature on temperature rise.

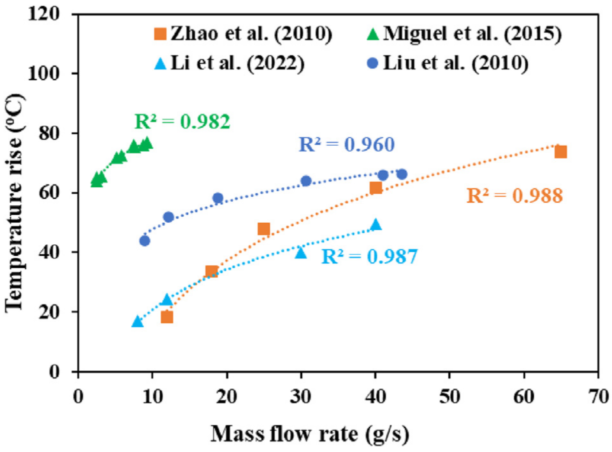


Fig. 13 – Effects of hydrogen flowrate on temperature rise.

recommended refuelling protocol for 2–10 kg of hydrogen storage at 70 MPa and 1.2–6 kg of hydrogen at 35 MPa in the fuel tank with a maximum hydrogen flow rate of 3.6 kg/min. According to the protocol, the hydrogen refuelling temperature limit is between −40 and 85 °C where final gas temperature in cylinder must not be greater than 85 °C, the maximum dispenser pressure limit is 87.5 MPa and the state of charge (SOC) between 95 and 100% as shown in Fig. 14(A). As per the protocol SAE J2601, the gas temperature in the inlet of the cylinder must be within the given limits depending on the SAE J2601 recommended dispensing temperature category (i.e., T40 = −40 °C, T30 = −30 °C, T20 = −20 °C, T10 = −10 °C and TA = Ambient) just after 30 s of refuelling starts [184].

The SAE J2601 protocols suggested two methods: “Table-based method” and “Mass and thermal Capacity (MC) formula-based method” for the refuelling station. The “MC formula-based method” was first developed by Honda and then incorporated into the limits and conditions of the SAE J2601 protocol [162]. This method uses empirical equations for dynamically calculating average APRR for the entire pre-cooling range of −17.5 °C to −40 °C and thereby determining the refuelling rate. The empirical equations are dependent on a number of parameters, including initial pressure, ambient temperature and cylinder capacity. The MC-based protocol uses an adaptive dynamic control strategy to estimate mass average temperature and hydrogen enthalpy based on the measured pre-cooling temperature and, thereby, calculate APRR and end pressure as shown in Fig. 14(B).

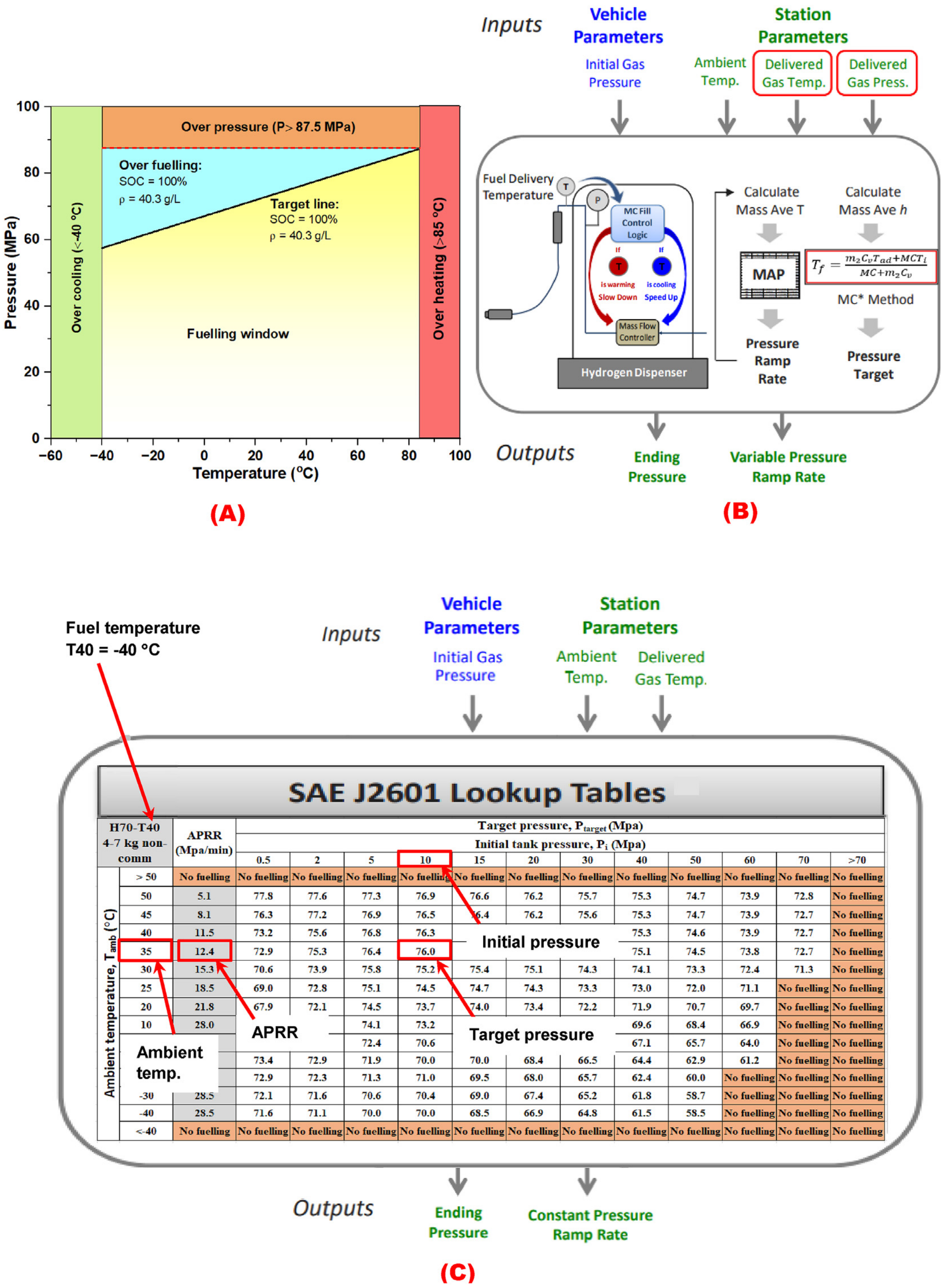
The “Table-based method” controls the average pressure ramp rate (APRR) based on the temperature and pressure data from the refuelling stations. This method includes 44 individual “lookup tables”, which have been developed based on thermodynamic simulation and validated using actual refuelling data from refuelling stations and experimental investigations [162,185]. The “lookup tables” states the constant APRR and end pressure value corresponding to a combination of other parameters such as delivery temperature and pressure (H70 for 70 MPa and H35 for 35 MPa), storage capacity (2–4 kg, 4–7 kg and 7–10 kg), dispenser interface and the outlet temperature (T40, T30 and T20) of the dispenser as illustrated in Fig. 14(C). Additionally, the protocol contains top-off refuelling method (determines top-off pressure and APRR) to enhance the refuelling speed and SOC in the case of low initial pressure (<5 MPa) [186]. The SAE J2601 protocols further aims to include warmer hydrogen delivery temperatures (−10 °C and ambient) for smaller capacity vehicles such as motorcycles [183]. Reddi et al. [162] simulated and compared the fuelling performance of the “Table-based method” and “MC formula-based method” using a physical model and reported a significantly faster fuelling in the case of “MC-based method” compared to “Lookup table-based method”.

The SAE J2601-2 protocol [188] was developed for dispensing gaseous hydrogen into heavy-duty vehicles (i.e., 35 MPa heavy-duty transit buses and trucks) of >10 kg compressed hydrogen storage system (CHSS) capacity. This protocol suggested a maximum hydrogen flow rate of 7.2 kg/min with an ambient temperature between −40 °C and 50 °C. Additionally, the SAE has developed SAE J2799 [189] for the requirements of communications hardware and software, SAE J2600 [190] for the design and testing requirements of connected devices such as fuelling connectors, nozzles, and receptacles and SAE J2719 [191] for hydrogen fuel quality standards. The International Organisation for Standardisation (ISO) has proposed several protocols under ISO TC 197: hydrogen technologies [192] and recently introduced a new protocol in ISO TC 197 for dispensing hydrogen into vehicles with variable sizes and cargo weight [193]. According to the ISO, the CHSS pressure should not be higher than 1.25 times that of the nominal working pressure provided by the manufacturer. China developed national regulatory codes and standards technical guide, GB50516-2010, for the safe operation of hydrogen fuelling stations [194,195]. According to the technical guide, the storage system working pressure in the fuelling station must not exceed 45 MPa and 90 MPa for a charging pressure of 35 MPa and 70 MPa, respectively. Standards Australia adopted eight ISO standards in 2020 with close involvement with the Australian Hydrogen Council for maintaining hydrogen quality and safety and standards of storage, transport and dispensing system in Australia [196]. For instance, AS ISO 19880.5 suggests the requirements of dispensers for hydrogen dispensing at 70 MPa with a temperature between −40 °C and 65 °C. Apart from these, several studies have proposed various qualitative and quantitative risk assessment approaches and assessed the safety hazards and their consequences, such as jet fires, safe distance and failure frequencies of different components in the hydrogen fuelling station [197,198].

4.3. *Strategies for temperature control*

An increase in cylinder gas temperature during fast refuelling can cause safety issues and challenges for the development of HFCEVs [12,199]. The final temperature of the gas in the cylinder and the energy requirement for hydrogen compression can be reduced to meet the SAE standard protocols by controlling the hydrogen flow rate, pre-cooling the inlet temperature and refuelling at a multi-stage initial pressure.

- **Mass flow rate control strategy:** Refuelling time can be shortened by increasing the hydrogen mass flow rate; however, this can cause a temperature rise in the cylinder. Therefore, the mass flow rate is required to be controlled within the boundaries and limits of temperature rise to obtain the optimum refuelling time. Dicken and Mérida [200] and Li et al. [171] noticed a quick temperature rise in the initial stage of refuelling (first quarter of the refuelling time) with a gradual increase in the later stage of refuelling due to the heat transfer between hydrogen gas and cylinder wall for the extended time. Therefore, varying the hydrogen mass flow rate can be effective in controlling the temperature rise during the refuelling instead of keeping a constant flow rate throughout the refuelling process. In the early stage of the refuelling process, the gas temperature can be maintained within the recommended limits by keeping the mass flow rate low and then can be increased in the later stage of the refuelling process. For instance, Wu et al. [201] proposed a 3D CFD simulation model for the fast refuelling of Type III fuel tanks and suggested various time-delayed strategies based on hydrogen flow rate control.



H70-T40 4-7 kg non-comm	APRR (Mpa/min)	Target pressure, P_{target} (Mpa)											
		Initial tank pressure, P_i (Mpa)											
Ambient temperature, T_{amb} (°C)		0.5	2	5	10	15	20	30	40	50	60	70	>70
> 50	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling
50	5.1	77.8	77.6	77.3	76.9	76.6	76.2	75.7	75.3	74.7	73.9	72.8	No fuelling
45	8.1	76.3	77.2	76.9	76.5	76.4	76.2	75.6	75.3	74.7	73.9	72.7	No fuelling
40	11.5	73.2	75.6	76.8	76.3	[illegible]	[illegible]	[illegible]	75.3	74.6	73.9	72.7	No fuelling
35	12.4	72.9	75.3	76.4	76.0	[illegible]	[illegible]	[illegible]	75.1	74.5	73.8	72.7	No fuelling
30	15.3	70.6	73.9	75.8	75.2	75.4	75.1	74.3	74.1	73.3	72.4	71.3	No fuelling
25	18.5	69.0	72.8	75.1	74.5	74.7	74.3	73.3	73.0	72.0	71.1	No fuelling	No fuelling
20	21.8	67.9	72.1	74.5	73.7	74.0	73.4	72.2	71.9	70.7	69.7	No fuelling	No fuelling
10	28.0	[illegible]	[illegible]	74.1	73.2	[illegible]	[illegible]	[illegible]	69.6	68.4	66.9	No fuelling	No fuelling
[illegible]	[illegible]	[illegible]	[illegible]	72.4	70.6	[illegible]	[illegible]	[illegible]	67.1	65.7	64.0	No fuelling	No fuelling
[illegible]	[illegible]	73.4	72.9	71.9	70.0	70.0	68.4	66.5	64.4	62.9	61.2	No fuelling	No fuelling
[illegible]	[illegible]	72.9	72.3	71.3	71.0	69.5	68.0	65.7	62.4	60.0	No fuelling	No fuelling	No fuelling
-30	28.5	72.1	71.6	70.6	70.4	69.0	67.4	65.2	61.8	58.7	No fuelling	No fuelling	No fuelling
-40	28.5	71.6	71.1	70.0	70.0	68.5	66.9	64.8	61.5	58.5	No fuelling	No fuelling	No fuelling
<-40	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling	No fuelling

Fig. 14 – (A) Temperature and pressure window for hydrogen refuelling, (B) J2601 MC-based protocols and (C) J2601 Table-based protocols [183,187] (Table-based method employs static refuelling control whereas MC-based method uses dynamic control strategy. MC is a lumped heat capacitance model, which can be expressed as follows: $MC = M\ in\ kg \times C\ in\ J/kgK$).

The controlling of flow rate exhibited about a 62% reduction in refuelling time compared to that of constant flow rate refuelling without exceeding temperature limits. Li et al. [171] also observed that controlling refuelling at variable rates could effectively lower the gas temperature rise in the fuel tank.

- **Pre-cooling temperature adjustment strategy:** Lowering the temperature of inlet hydrogen through a proper cooling arrangement is the most effective and feasible solution. Pre-cooling can help to keep the maximum gas temperature below 85 °C during high-pressure fast refuelling [160]. Bai et al. [176] investigated a pre-cooling strategy during fast refuelling and noted a linear relationship between the final temperature and pre-cooling temperatures, where the final gas temperature reduced with a decrease in pre-cooling temperature due to a reduction of internal energy of inlet hydrogen through pre-cooling. However, the pre-cooling temperature must not be below −40 °C to maintain the standard fuel delivery temperature boundaries (−40 to 85 °C) [160]. Xiao et al. [202] developed a correlation between the pre-cooling temperature and various parameters, such as initial temperature, initial pressure and refuelling time, using a simplified lumped parameter thermodynamic model. Handa et al. [203] studied an MC formula-based pre-cooling strategy to control the refuelling rate in a fast refuelling process and demonstrated that pre-cooling at −20 °C can achieve the refuelling time target of approximately 3 min. Japan announced their Hydrogen Fuel Cell Strategy Road Map in 2019 and is aiming to develop a next-generation technology by 2022 for refuelling within 3 min with a pre-cooling temperature between −15 °C and −25 °C [204]. Li et al. [171] included a heat exchanger before the fuel tank and investigated a pre-cooling strategy for reducing the gas temperature. The pre-cooling strategy significantly reduced the maximum temperature in the tank; however, the overall process was highly energy intensive.

- **Multi-stage initial pressure strategy:** In order to reduce the energy consumption for hydrogen compression and gas volume at maximum pressure, a refuelling strategy with multi-stage initial pressure has been investigated and received recent interest from refuelling stations [160,205] due to its effectiveness for multiple vehicles and back to back refuelling. In a three-stage refuelling process, the fuel cylinder is filled with hydrogen from three hydrogen stationary storage tanks in the order of low-pressure, medium-pressure and high-pressure instead of a single stationary storage tank [169], which reduces the requirement of hydrogen compression. Fig. 15 presents the temperature and pressure profiles of gas in the cylinder during a multi-stage refuelling process [206]. Multi-stage initial pressure approach during hydrogen refuelling reduces the cylinder gas temperature rise in addition to the lowering compression energy. At the beginning of hydrogen refuelling process, a low-pressure stationary storage tank is used to fill the on-board cylinder, resulting in a sharp increase in the cylinder gas temperature, as a decrease in initial pressure increases the temperature rise [169,173]. When the difference between the active stage pressure and cylinder pressure decreases to a critical value, the cylinder is filled with medium or high-pressure stationary storage tank which reduces the cylinder temperature rise. Because at high initial pressure, the volume of hydrogen rapidly increases inside the cylinder which lower the temperature rise.

Zheng et al. [169] simulated a two-stage refuelling process using CFD simulation and observed the savings in compression energy in Stage 2 due to the lower compression ratio. In another study, Zheng et al. [207] proposed a multi-objective iterative optimisation model for analysing a three-stage refuelling process. The optimised strategy significantly improved the utilisation ratio and refuelling speed. Yu et al. [208] developed a thermodynamic model to study a three-stage cascade storage system for a refuelling station and identified the medium-pressure stage as the main stage for hydrogen refuelling. The higher utilisation ratio was reported for the smaller proportion of low-pressure stage volume. Caponi et al. [209] studied hydrogen compression energy for three-stage cascade storage and compared it with single-stage using a dynamic lumped model in the MATLAB platform. The three-stage process required 10% less energy for hydrogen compression when compared to the single-stage process. Rothuizen et al. [210] exhibited approximately 12% lower compression energy consumption for a three-stage process than for a single-stage process. Gao et al. [205] proposed a dynamic simulation model to optimise energy consumption in a three-stage refuelling process. The optimised three-stage refuelling strategy significantly reduced the compression energy consumption by approximately 55.3% from 21.64 to 9.68 MJ, compared to that of single-stage refuelling. The saving in compression energy consumption for a two-stage refuelling process was about 39.3%. In another study, three-stage and four-stage refuelling strategies with cascade storage reduced energy consumption by 16.5 and 20%, respectively [176]. Therefore, it can be concluded that the higher the stages in the refuelling process, the lower the compression energy consumption; however, higher number of stages can be associated with increased capital cost.

4.4. *Economic aspects of refuelling*

The most mature and industrially common hydrogen refuelling stations can deliver hydrogen to vehicles through a gaseous hydrogen refuelling station or a liquid hydrogen refuelling station. Depending on the configuration of the refuelling station, the capital cost of the station includes the cost of the compressor, chiller/heat exchanger, storage tank, dispenser electrical and piping control [211]. The capital cost of a hydrogen refuelling station ranges between $0.84 million and $7.84 million, depending on the configuration and

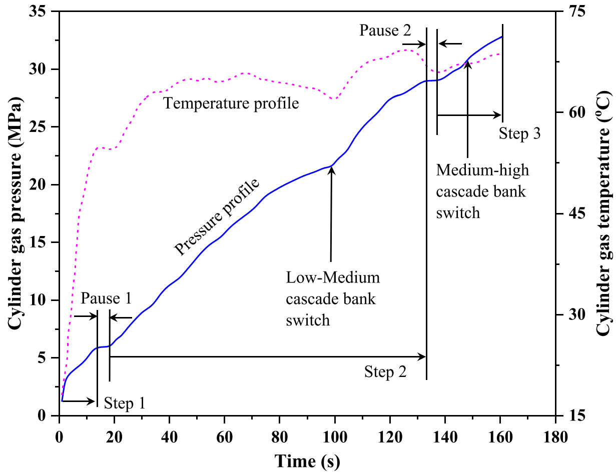


Fig. 15 – Pressure and temperature profiles during filling of a typical hydrogen fuel tank using multi-stage initial pressure (redrawn from Kountz et al. [206]).

infrastructure of the station (which depends on the type of vehicles to be refuelled), capacity (i.e., mass of hydrogen and number of vehicles to be refuelled, number of tube trailer connection points), operation conditions (i.e., delivery temperature and pressure) and country of installation. According to the National Renewable Energy Laboratory report, the investment cost of a refuelling station in Europe is about $1.34 million and $2.05 million for 150 kg/day and 700–1000 kg/day capacity stations, respectively [212]. The capital cost in Japan for the same station capacity is approximately $2.5 and $5.05 million, respectively. According to the Argonne National Laboratory report, the installation cost of a hydrogen refuelling station with a capacity of 250 and 1000 kg/day is approximately $0.84 and $2.05 million, respectively [212]. In another study, the capital cost of a gaseous hydrogen refuelling station (180 kg/day) and a liquid hydrogen refuelling station (350 kg/day) was estimated to be $2 and $2.8 million, respectively [213]. Kurtz et al. [28] reported that the capital cost of 46 refuelling stations was between $0.91 and $4.6 million, with an average cost of $2.2 million. Brown et al. [214] estimated the capital investment of a 180 and 400 kg/day capacity station as $1 and $2 million, respectively. Apart from these, Xu et al. [215] estimated the capital cost and operation/maintenance cost of an on-site SMR hydrogen fuelling station in China and these were $1.04, $4.15 and $7.84 million for 100, 500 and 1000 kg/day capacity, respectively. The annual operation and maintenance cost significantly increased with an increase in the station's dispensing capacity. The estimated annual operation and maintenance cost was $0.16, $0.78 and $1.56 million for 100, 500 and 1000 kg/day dispensing capacity, respectively. Minutillo et al. [216] estimated ~2.89–3.52 times higher capital cost and ~3.29–3.32 times higher operation and maintenance cost for an on-site water electrolysis hydrogen refuelling station of capacity 200 kg/day compared to 50 kg/day, irrespective of electricity mix.

Fig. 16 depicts the breakdown of a hydrogen refuelling station's capital cost. The compressor is the major contributor to the station's capital cost (~18–58%), irrespective of the capacity of the station and region. In 2019, Kurtz et al. [28] estimated that the station equipment, procurement and construction accounted for approximately 84% of the total capital cost. Reddi et al. [211] performed a detailed breakdown cost analysis in 2017 using the hydrogen delivery scenario analysis model and investigated the effect of equipment cost, station capacity, the economics of scale and utilisation capacity on the refuelling cost for a gaseous hydrogen refuelling station and liquid hydrogen refuelling station. The study revealed that for a 500 kg/day gaseous refuelling station in California, the compressor accounted approximately 53% of the total capital cost of the station. The cost of the heat exchanger, booster compressor and evaporator were fairly linear with the cooling capacity of the heat exchanger, compressor motor rating and flow rate of the coolant, respectively. On the contrary, the cost of a refrigeration unit, main compressor and cryogenic storage were in polynomial correlation with refrigeration, compressor motor rating and storage capacity. However, the cost of the refuelling station equipment can be reduced by increasing the annual production [212].

The cost of hydrogen at the dispenser is a major concern for HFCEVs, which includes production, transport and refuelling/dispensing costs. The Department of Energy, USA, has set a target to reduce the transport (excluding production) and dispensing cost of hydrogen to around $5/kg H_2 by 2025 with hydrogen supplied by tanker trucks and ultimately to $2/kg with hydrogen supplied by pipelines from $12-$13/kg in 2017 [217]. In 2017, the hydrogen cost at the dispenser in California was estimated at about $13–$15/kg H_2, including $6–$8/kg H_2 production and transport costs in the United States resulting in $7/kg H_2 for the dispensing cost [211]. An economic study of 68 stations in California conducted by Brown et al. [214] also revealed that the levelised cost of hydrogen at the dispenser, including production and transport cost was between approximately $8/kg H_2 and $19/kg H_2, with an average cost of $10/kg H_2. The maximum despensing cost was estimated about $7.2/kg H_2. Additionally, the cost of hydrogen at the pump for a 70 MPa station is reported as two times higher than that of gasoline, indicating the requirement for low-cost production methods with high production capacity [28]. The dispensing cost for a refuelling station of capacity 200 kg/day in California was estimated at about $6–$8/kg gaseous H_2 and $8–$9/kg liquid H_2 without considering the hydrogen production and transport cost [211]. The hydrogen refuelling cost at a 80% utilisation of the station was reduced by 36.7% from $6.0/kg H_2 to $3.8/kg H_2 when the capacity of the station increased from 200 to 1000 kg/day [211]. The increase in station's capacity increased the capital investment of the station; however, it decreased the station's levelised refuelling cost due to the economies of scale. The station's levelised cost was also reduced with an increase in the production volume of station equipment, irrespective of the capacity of the station.

Combining economies of scale of a large station capacity and high equipment production can reduce the hydrogen dispensing cost to $2/kg H_2, which indicates the future competitiveness of HFCEVs with other options [211]. Minutillo et al. [216] also reported a decrease in the refuelling station's levelised cost with an increase in the station's dispensing capacity, irrespective of the electricity mix. Elgowainy et al. [218] performed an economic analysis of a hydrogen refuelling station using the Heavy-Duty Refuelling Station Analysis Model to estimate the levelised refuelling cost and investigating the effect of various parameters on refuelling cost, as shown in Fig. 17. The refuelling cost increased with an increase in fuelling rate and fuelling pressure for both gaseous and liquid hydrogen dispensing as the faster and high-pressure fuelling required higher capacity and specialised equipment, resulting in higher fuelling cost. The staggered fuelling reduced the fuelling cost for the same fleet size and filled amount when compared with that of back-to-back fuelling. No significant impact of tank type on the fuelling cost was reported in this study [218]. An increase in fleet size and production volume of station equipment positively impacted the fuelling cost, and thereby the fuelling cost can be reduced to $1-$1.5/kg H_2 by increasing the number of HFCEVs and production volume of fuelling components. However, in an early market, the high utilisation capacity of the station, high fleet size and high production volume of station equipment are unlikely and require more research attention.

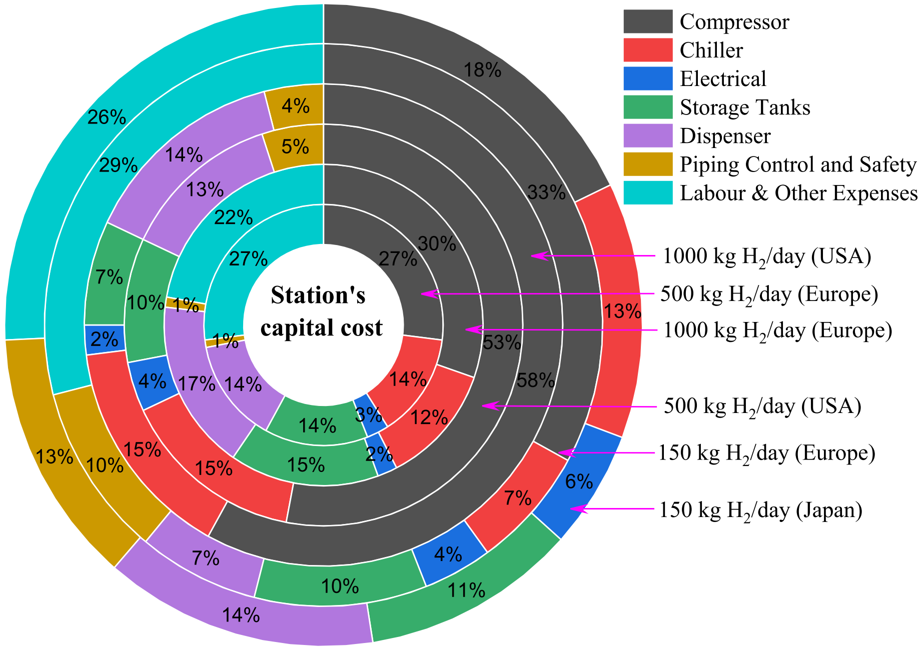


Fig. 16 – Capital cost breakdown of hydrogen refuelling station at different capacities (data from [29,211,212]).

4.5. *Recent technological advancements-mobile/portable refuelling station*

Hydrogen refuelling station infrastructure will be essential for a breakthrough in the commercialisation of a hydrogen-powered automotive sector. However, the current high cost of stationary refuelling infrastructure is considered as one of the major issues of HFCEVs. China has made significant advancements in the hydrogen fuelling stations design based on the national technical guide GB50516-2010 and developed the first mobile fuelling station in 2004 by Tongji University and Shanghai Sunwise Energy System Company [219] to support the early HFCEVs initiatives of China (Fig. 18). At the same time, Niedwiecki et al. [220] patented another design of a portable hydrogen fuelling station. A mobile hydrogen fuelling station is an integrated solution for fuelling infrastructure which offers several advantages such as compact design, low capital and installation cost, fast deployment, low maintenance and reliability over the stationary fuelling station. The design of mobile refuelling station differs from one company to other. The major components of Tongji University and Shanghai Sunwise Energy System Company developed mobile station include interconnected pressure vessel tubes for hydrogen storage, pneumatically driven boosters instead of the compressor (in case of stationary fuelling station) for increasing hydrogen pressure, a nitrogen system for driving boosters, a dispensing system including a control panel and chiller and various safety systems such as temperature and pressure regulator, hydrogen detector, safety valves and rupture discs and emergency control devices [198,221]. Hylium Industries developed mobile refuelling consists of liquid hydrogen tank, pump, liquid hydrogen gasifier, buffer tank and dispensing system [222]. FUELINGAS brand contains compressor, high-pressure storage tank and dispensing system [223]. Following the design of the second generation station in 2007, Tongji University and Shanghai Sunwise Energy System Company developed a third-generation mobile station with improved fuelling efficiency, safety controls and operation protocols, and investigated fuelling performance at the Expo Shanghai 2010 [224]. During this Expo, 196 HFCEVs, including six buses, 90 cars and 100 sight-seeing cars, were fuelled by two portable fuelling stations. The demonstration exhibited that the portable fuelling stations successfully dispensed 15204 kg hydrogen through 20302 times refuelling without any major incidents. The risks and safety issues associated with the refuelling were also assessed during the Expo and reported lower stationary risks than the acceptable criteria [198,221]. Later in 2014, a standard safety and operating protocol (GBT 31139–2014) was developed by China to ensure a safe and fast operation of mobile hydrogen fuelling stations with a refuelling pressure between 15 MPa and 70 MPa [219,225], as shown in Fig. 18. The working pressure of hydrogen storage system must be 1.25–1.5 times of refuelling pressure.

Several companies have launched mobile hydrogen refuelling stations and are providing maintenance and other services related to the refuelling stations. The key technical specifications of the mobile hydrogen refuelling station, developed by some of the leading companies, are listed in Table 2. The Wystrach developed mobile hydrogen refuelling station, “WyRefueler”, was deployed in Europe (Belgium, Germany, France, and the Netherlands) through the “H2Share project” to provide hydrogen refuelling to trucks [226]. The “WyRefueler” was also installed under the “HyBayern” project for refuelling passenger cars and buses. Air Products installed their self-contained portable hydrogen refuelling station of capacity 150 kg hydrogen in several locations in the world [227]. In Australia, the Queensland University of Technology has started

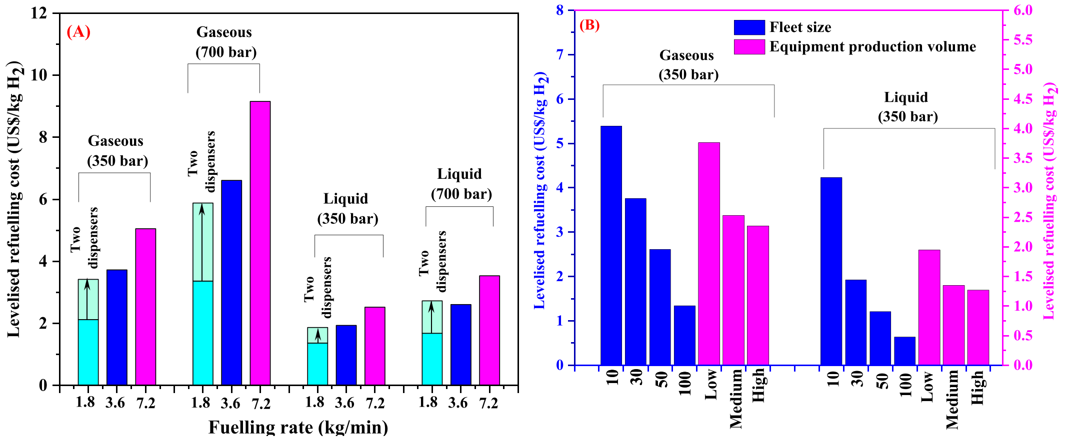


Fig. 17 – Various factors affecting the hydrogen refuelling cost (redrawn and modified from Elgowainy et al. [218]).

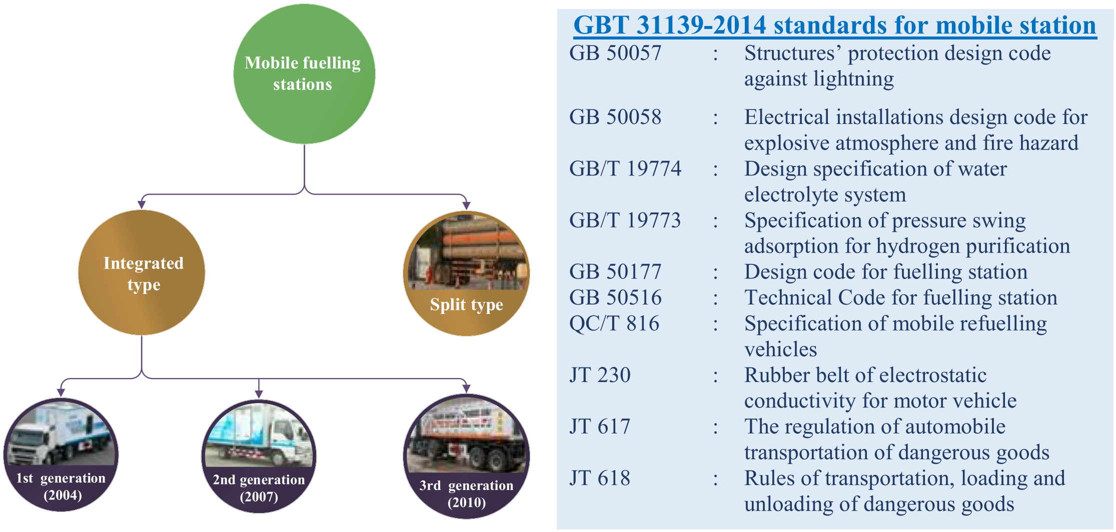
research and development work on a portable refueller in 2021 in collaboration with H2H Energy Pty Ltd and Queensland Government [228]. The MAXIMATOR company designed a portable hydrogen refuelling station with a maximum operating pressure of 105 MPa [229]. OneH2 developed mobile refueller can dispense hydrogen from high-pressure trailer at a dispensing pressure between 35 and 70 MPa [230].

The NanoSUN developed portable hydrogen refuelling station, "Pioneer 145", can refuel 60 delivery vans of 5 kg storage capacity with a refuelling time range of 10–13 min at 35 MPa, which is only seven refuels for heavy-duty trucks of 40 kg capacity with a refuelling time of 20–60 min as shown in Fig. 19(A) [232]. Approximately, 95 cars can be refilled at 70 MPa with a refuelling time range of 10 min. However, the NanoSUN has provided flexibility in the refuelling network (Fig. 19(B)) to include more "Pioneer 145" stations when the fleet size grows. Additionally, the "Pioneer 145" station uses flexible cascade refuelling instead of decant refuelling, which avoids using the compressor and increases the number of full refuels, as shown in Fig. 19(C). The Hylium Industries, a South Korea-based company, has developed the world's first mobile liquid hydrogen refuelling station, which can directly refuel 25 vehicles per day [222]. This refuelling station consists of a liquid hydrogen pump and gasifier (instead of a compressor and chiller for a gaseous hydrogen station), which consumes 70% less energy than a gaseous station. The above demonstration indicates the suitability of portable stations for refuelling the small fleet sizes.

5. Discussions and future research perspectives

Research on hydrogen production to utilisation in the transport sector has received immense attention in recent years, resulting in significant technological advancement. However, there still exist several challenges that are required to be addressed. Therefore, this paper has summarised some suggestions for future research directions as follows:

- The transport of hydrogen from a central production facility to a refuelling station is a major issue as it requires a



GBT 31139-2014 standards for mobile station

GB 50057	:	Structures' protection design code against lightning
GB 50058	:	Electrical installations design code for explosive atmosphere and fire hazard
GB/T 19774	:	Design specification of water electrolyte system
GB/T 19773	:	Specification of pressure swing adsorption for hydrogen purification
GB 50177	:	Design code for fuelling station
GB 50516	:	Technical Code for fuelling station
QC/T 816	:	Specification of mobile refuelling vehicles
JT 230	:	Rubber belt of electrostatic conductivity for motor vehicle
JT 617	:	The regulation of automobile transportation of dangerous goods
JT 618	:	Rules of transportation, loading and unloading of dangerous goods

Fig. 18 – Types of mobile hydrogen fuelling station (left) and standard protocols (right) (prepared from [219,225]).

significant infrastructure cost, which increases the levelised refuelling cost. On-site small-to medium-scale hydrogen production using a mature production technology, such as electrolysis, with renewable electricity (Fig. 20) can avoid transport from the production site to the refuelling station and thereby reduce the cost of hydrogen to the customer. Some companies have developed on-site electrolysis hydrogen generator units with a generation capacity of 20 kg/day to >1000 kg/day, and a few of them are already in the implementation stage in Europe, USA and Canada [17,233–238]. However, the renewable energy sources are uncertain and variable in nature with geographical location and season which can affect the overall hydrogen refuelling network. Therefore, country-specific feasibility studies of hybrid energy systems such as solar-wind-grid integrated on-site electrolysis, considering the uncertainty of renewables are required to understand the financial return of the investment in that specific location.

- Despite the higher cost of hydrogen production from water electrolysis compared to other production routes, it is considered as the cleanest technology to produce hydrogen from water with oxygen only as the waste from the process and has received interest due to environmental concerns associated with the use of natural gas and coal. However, the use of freshwater can cause water stress. Therefore, seawater can be an emerging alternative for producing green hydrogen via electrolysis. Seawater electrolysis is still in early development stage and numerous fundamental research is going on in laboratory scale investigations. For instance, the major challenges associated with the use of seawater in the electrolysis process include corrosion and chlorine production, which can be suppressed through pre-desalination, costing about \$0.7–2.5 per m^3 of water [239]. Additionally, researchers are currently focussing on direct electrolysis of seawater and working on the development of self-driven in-situ membrane purification process, new electrode or catalyst design to suppress chlorine oxidation/corrosion problems [240–243]. However, effects of dissolve impurities and other ions such as sulphate, carbonate calcium and magnesium on the competitive reactions and long-term performance stability is still unclear. The standardisation of electrolyte and optimisation of process conditions such as electrolyte composition, current density and electrolysis time with newly developed catalysts for direct electrolysis are critical for commercialisation of seawater electrolysis, as the composition of seawater significantly varies across the world. It is also necessary to understand how each of the approaches of seawater electrolysis is economically feasible, considering the transport of hydrogen from the production site (near to sea) to refuelling stations (far away from the sea).

Table 2 – Leading suppliers of mobile hydrogen refuelling station.

Company and product	Key specifications	Reference
 FUELINGAS	Dispensing pressure: 35–70 MPa; Pre-cooling temperature: 10 to −40 °C; Maximum discharge pressure: 45–87.5 MPa; Refuelling capacity: 50–1000 kg/12h	[223]
 SinoHy Energy	Dispensing pressure: 35 MPa; Pre-cooling mode: water cooling; Hydrogenation speed: 1–2 kg/h; Buffer hydrogen storage: 12–18 Nm^3/h	[231]
 NanoSUN	Dispensing pressure: 35–70 MPa; Maximum dispensing rate: 3.6 kg/min; Working temperature: 10 to +40 °C; Maximum storage pressure: 42.5 MPa; Maximum storage capacity: 420 kg; Storage vessel size: ~15,000 L water volume	[232]
 Wystrach	Dispensing pressure: 35 MPa; Storage capacity: 400–950 kg; Storage cylinder size: 20–45 ft; Buffer storage capacity: 190 kg; Dispensing capacity: 360 kg/day	[226]
 Hylium Industries	Capacity: 2500 L hydrogen; Storage pressure: 0.3 MPa; Pump pressure: 80 MPa; Dispensing pressure: 70 MPa	[222]

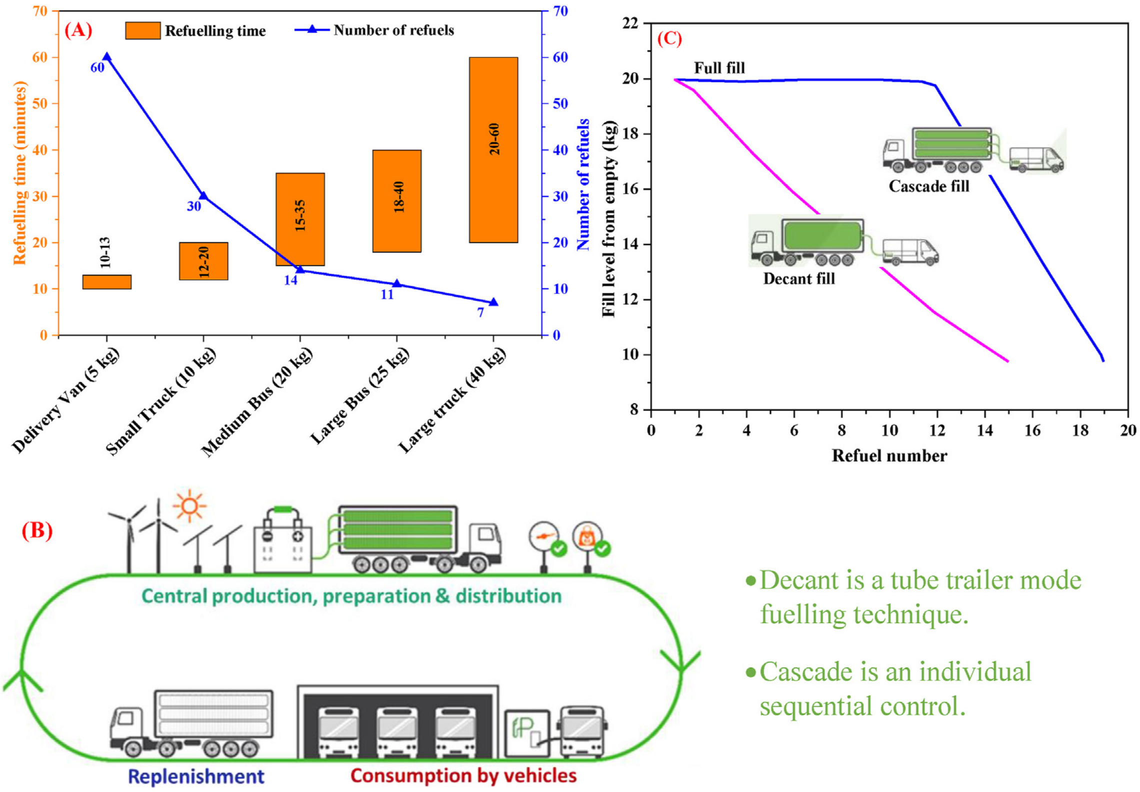


Fig. 19 – **(A) Refuelling capacity of NanoSUN developed mobile station, data in parentheses indicates on-board storge capacity (data from [232]), (B) basic value chain and deployment model (redrawn from [232]) and (C) performance of refuelling techniques for a typical bus (redrawn from [232]).**

- Hydrogen storage at ambient temperature using biomass-based carbon material is gaining interest over other materials, such as MOF, metal hydride, and liquid organic hydrogen carriers due to the renewability, low cost and ease of production. Biomass derived adsorbent-based hydrogen storage technology is in laboratory scale research stage. Therefore, a number of issues and fundamental knowledge gaps on the structure of feedstock for carbon material production, the role of inherent inorganics of biomass, storage capacity improvement, the adsorption-desorption cycle and an efficient hydrogen extraction strategy still need to be addressed for developing scale-up biomass-based efficient hydrogen storage systems and achieving the target of DOE.

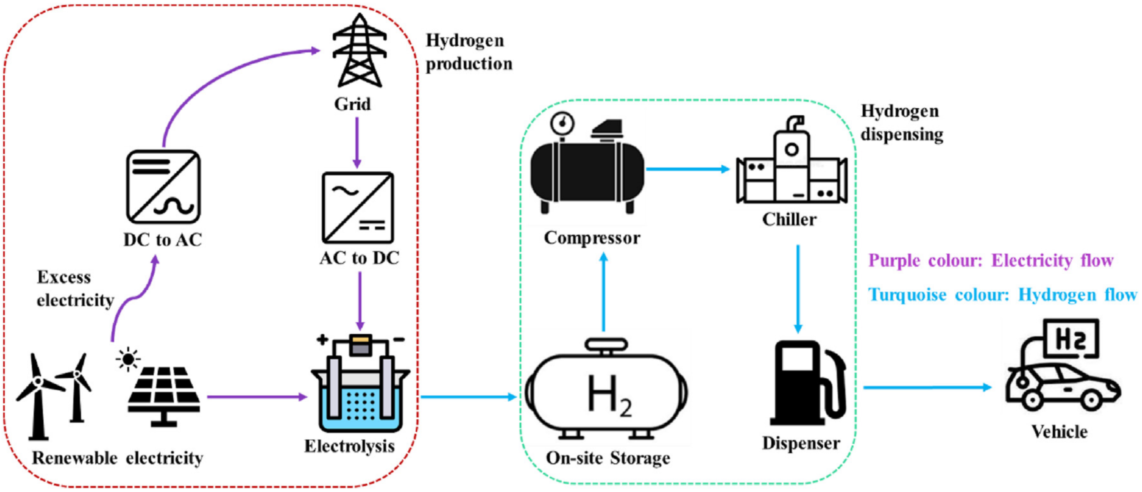


Fig. 20 – **Proposed on-site hydrogen production and refuelling with renewable energy integration.**

Table 3 – Summary of key hydrogen technologies.

Production technologies				Storage technologies		Transport technologies			Refuelling station capital and levelised refuelling cost[v]
Technology	Conversion efficiency (%)	Production cost (US$/kg)[x]	GHG emissions (kgCO$_2$eq/kg)	Technology	Storage cost (US$/kg)[y]	Technology	Cost (US$/kg/100 km)[z]	GHG emissions (gCO$_2$eq/kg/100 km)[u]	
Biomass gasification	4.9–14.0	1.21–7.48	2.67–6.20	Gaseous tube	0.15–25.5	Gaseous tube trailer	0.44–2.00	83–718	Capital cost: $0.84 –5.05 million for 150 –1000 kg/day. Levelised refuelling cost: $1.50–8.00/kg H$_2$
Coal gasification	3.0–21.0	0.90–1.50[a]	11.50–32.20[a]	Liquid tank	0.59–4.57	Gaseous pipeline	0.10–1.96	50–709	
		1.03–2.11[b]	2.20–10.9[b]	Metal hydride	0.40–4.00	Liquid road	0.12–0.88	255–3332	
SMR	4.0–36.4	0.75–2.33[a]	7.05–13.80[a]	Underground	0.14–3.28[f]	Metal hydride truck	0.38–2.49		
		1.22–3.49[b]	3.07–5.59[b]						
Electrolysis	1.5–11.1	4.01–13.11[c]	23.30–36.80[c]						
		0.98–23.27[d]	0.37–4.00[d]						
		2.27–9.37[e]	0.03–2.21[e]						

[a] – without CCS.
[b] – with CCS.
[c] – grid electricity.
[d] – solar electricity.
[e] – wind electricity.
[f] – varies with storage site types (salt caverns, depleted oil and gas deposits, saline aquifers and hard rock caverns).
[u] – includes emissions from compression/liquefaction for storage.
[v] – gaseous central hydrogen production plant.
[x] – varies with plant capacity, operating cost and other economic assumptions. Central gaseous hydrogen plant.
[y] – depends on capacity and storage time.
[z] – varies with transport capacity and distance.

- Temperature rise during fast refuelling of high-pressure hydrogen is a major technical challenge where a number of methods and strategies, such as control of refuelling rate, multi-stage refuelling and pre-cooling, have already been investigated. However, hydrogen mass flow rate control can increase refuelling time, pre-cooling can require high energy and higher number of stages can increase capital cost. The integration of refuelling rate control with the pre-cooling strategy can reduce the energy consumption while achieving the target of gas temperature and refuelling time. Therefore, experimental investigation of performance (energy consumption and ability to meet standard protocols) of coupling two or more strategies and their economic feasibility assessment would be interesting.
- Insufficient refuelling infrastructure is a major challenge for the commercialisation of hydrogen fuel cell vehicles, particularly for long-haul vehicles. An effective mapping and assessment are required prior to the development of refuelling infrastructure. A few studies have focussed on the planning and design of refuelling infrastructure for some countries [30,244,245]. However, more geographical studies are required to develop a GIS-based methodology considering the current vehicle's facility and future target. The GIS-based methodology, considering hydrogen delivery options and customers availability will help to determine the ideal location for setting up refuelling infrastructure and thereby develop policies for future expansion.
- Mobile hydrogen refuelling stations are promising alternatives to stationary refuelling stations for small fleet size, as the high capital costs of stationary refuelling stations is not economically attractive for a small fleet. Future research should focus on the supply chain and refuelling network design of portable stations for large fleet sizes and heavy-duty vehicles, considering the comparative economic feasibility assessment with stationary refuelling stations.

6. Conclusions

Hydrogen is a promising energy carrier which can store and deliver energy for the transition of future transport and heat sectors into sustainable options. This paper critically reviews the technological advancements, prospects and challenges of hydrogen production to refuelling into fuel cell vehicles to promote decarbonisation of the transport sector. The summary of key aspects, including efficiency, cost and associated emissions of major hydrogen technologies is highlighted in Table 3. Following a detailed literature review, this paper has drawn several conclusions as follows:

- The costs of hydrogen production by well-established fossil-based technologies such as SMR and CG are competitive to the DOE target; however, these technologies have shown significant GHG emissions, which contradicts net-zero goals. In this case, renewable electricity-based electrolysis can be promising in the long run.
- Carbon-based materials such as activated carbon, carbon nanotubes and nanofibers show promising hydrogen storage capacity at cryogenic conditions. On the other hand, metal hydrides exhibit superior hydrogen uptake capacity at ambient conditions.
- The temperature rise in the fuel tank/cylinder decreases with the increase in initial pressure and gas temperature; however, it increases with the increase in mass filling rate and ambient temperature. The temperature rise issue can be controlled by controlling the hydrogen flow rate, pre-cooling the inlet temperature and refuelling at a multi-stage initial pressure.
- The levelised cost of hydrogen at a refuelling station can be reduced by increasing the utilisation capacity of the station, fleet size and production volume of station equipment and reducing the hydrogen production cost. Portable refuelling stations show high potential for small fleet sizes.

The paper has identified several economic and technical challenges and suggested future research directions on cost-efficient sustainable hydrogen production routes and storage approaches, refuelling process optimisation and refuelling network design for improving the feasibility of hydrogen-powered transport in the long-run.

Declaration of competing interest

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

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Appendix A. Supplementary data

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