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Development and application of fuel cells in the automobile industry

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ABSTRACT

The automotive industry consumes a large amount of fossil fuels consequently exacerbating the global environmental and energy crisis and fuel cell electric vehicles (FCEVs) are promising alternatives in the continuous transition to clean energy. This paper summarizes the recent development of fuel cell technologies from the perspectives of the automobile industry and discusses current bottlenecks hindering commercialization of FCEVs. Current status of the fuel cell technology, policies and market prospect of FCEVs, as well as recent progress of FCEVs are reviewed. Polymer electrolyte membrane fuel cells constitute the mainstream and most mature fuel cell technology for automobile applications. Hybridization with an auxiliary battery system will greatly boost the dynamic response of FCEVs. Hydrogen FCEVs have entered the preliminary commercialization stage since 2015 and the market share of FCEVs is expected to grow at a high rate. Challenges encountered by commercialization of FCEVs and future outlook are also discussed. Future efforts are expected to focus on solving problems such as the high cost of fuel cell stack production and maintenance, insufficient hydrogen supply facilities, insufficient reliability, slow cold start, safety concerns, and immature energy management systems of FCEVs. This review serves as a reference and guide for future technological development and commercialization of FCEVs.

1. Introduction

Climate change and energy crisis are two major problems facing humanity. Unfortunately, non-renewable fossil fuels remain the world's largest energy provider and contribute to climate change and environmental pollution [1]. One of the major products that use fossil fuel are automobiles and therefore, the transportation industry in many countries are gradually replacing fossil fuels with renewable energy in the effort to migrate into green vehicles consequently leading to the boom in the research and development of novel green energy strategies. A comparison of the major emerging energy devices including fuel cells (FCs), batteries, solar cells, and so on is presented in the Ragone Plot (Fig. 1) and Table 1. FCs have higher energy densities than other energy devices rendering them suitable for long-range vehicular applications and these benefits have consequently spurred the research and development of FC-powered vehicles.

Pure battery electric vehicles, gasoline hybrid electric vehicles, and fuel cell electric vehicles (FCEVs) are the main "green" vehicles. Pure battery electric vehicles have a typical driving range of less than 400 km per charge and the recharging time is as long as 1–3 h currently [4], although continuous improvements are being made by manufactors such as Tesla. Hybrid gasoline-electric vehicles are considered transition models since they still consume fossil fuels and exhaust pollutants [5]. Compared with the energy efficiency of traditional internal combustion engines (ICEs) of 30–40%, FCEVs have a high energy efficiency of 40–60% and the only by-product is water [6]. Hence, the market for FCEVs is enticing and broad [7]. In fact, as promising new and environmentally friendly vehicles, FCEVs have been demonstrated commercially. In addition to hydrogen FCs, some new types of FCs such as direct liquid FCs [8,9] and alkaline anion exchange membrane FCs [10] have been developed in recent years and may be applied in the electric vehicles. In this review, the energy issues and entailing

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environmental pollution are presented from the perspective of the transportation industry, followed by discussions of the operating principles of various types of FCs, FC market in the automobile industry, recent progress of FCs in the automobile industry, development of various types of FCs, and development of commercial FCEVs. Finally, the challenges and outlook for commercialization of FCEVs are discussed.

2. Energy concerns and environmental pollution

In 2019, energy consumption reached almost 14,000 million tons of oil equivalent and is expected to increase at a rate of 2.9% [11]. The main energy sources are oil, coal, natural gas, biomass, and electricity accounting for 32%, 26%, 23%, 10%, and 9%, respectively [12]. Electricity can be subdivided into hydroelectricity, nuclear power, and renewable energy accounting for 6.7%, 4.3% and 4%, respectively, in the total energy consumption [12]. At present, fossil fuels are still dominant [13] and the transportation industry accounts for about 30.8% of its use, as shown in Fig. 2. More than half of the fuel is currently consumed by passenger vehicles and the remaining by freights [14]. The transportation industry has made great progress in the past ten years. More than 80% of the increase in the energy demand is attributed to developing countries in Asia [15], in which the number of automobiles is rising rapidly [16]. Owing to worsening environmental issues, it is urgent to substitute fossil fuels with clean and renewable energy sources.

Combustion of fossil fuels produces a large quantity of pollutants and greenhouse gasses. In 2018, global carbon emissions reached 0.6 GT and continue to rise [12]. It has been reported that automobile exhausts account for about 54% of CO, 30% of nitrogen oxides ($NO + NO_2$), 10% of particulate matters (PM), 47% of non-methane volatile organic compounds, and 14% of CO_2 in global emissions [17]. CO is released when carbon in the fuel fails to burn completely. Transportation is also a major source of black carbon, or called "soot particles", accounting for about 35% of global emissions [18]. Vehicle emissions contribute to PM 2.5 at a proportion of 1.4% and the PM 2.5 problem is particularly acute in developing countries in Asia [19,20]. For example, some cities in China and India have reported an average PM 2.5 levels of over 100 μg m^{-3} [21-24] and internal combustion engines (ICEs) in vehicles are the major source of PM 2.5 [25]. Air pollution has become a severe problem plaguing some developing countries such as China and India [17,26] and PM2.5 emissions from vehicles stem from mainly flammable fuel

Table 1
Comparison of various emerging energy devices [2].

Device	Energy density	Life time	Advantage	Disadvantage
FC	Very high	20–25 years	• High efficiency; • Modular and compact; • Smooth power output; • Rapid H_2 refilling; • Eco-friendly	• High cost; • Slow cold start; • Dangers associated with H_2; • High price of H_2
Battery	High	4–6 years	• Portable and rechargeable; • Low cost; • Mature technology	• Slow recharging; • Short lifetime; • Pollution from battery preparation and recycling; • Flammable liquid electrolyte
Supercapacitor	Very low	10–20 years	• Rapid response and recharging	• Short duration energy storage; • High cost
Photovoltaic panel	Medium	15–20 years	• Eco-friendly	• Intermittency of power output; • Bulky for vehicles
Flywheels	High	5–10 years	• High power output and rating; • Eco-friendly	• Slow charging; • Heavy weight
Superconducting magnetic energy storage system	Low	25–30 years	• High power output and rating; • High efficiency; • Eco-friendly; • Rapid response	• Short duration energy storage; • High cost

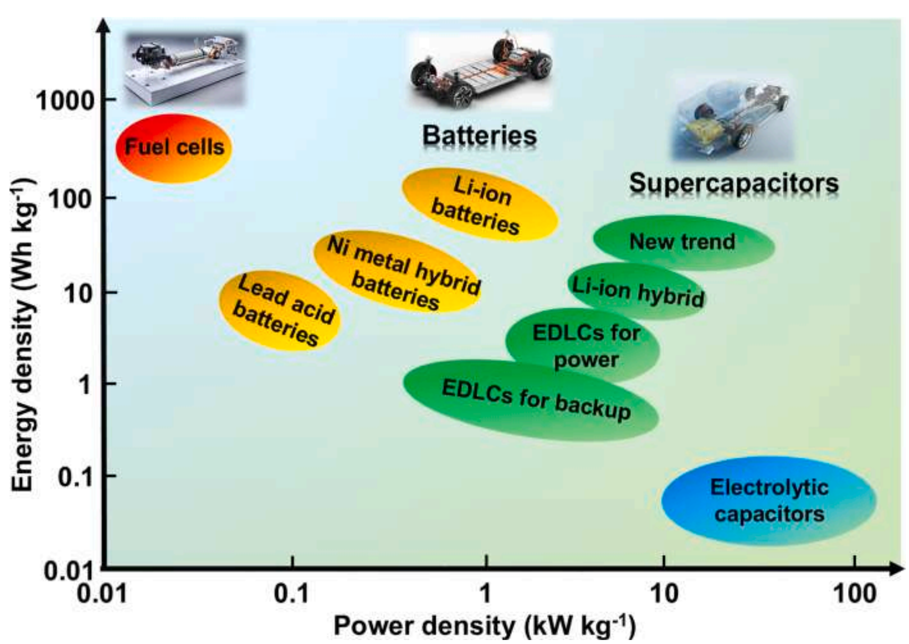

Fig. 1. Ragone plot of various emerging energy devices [3].

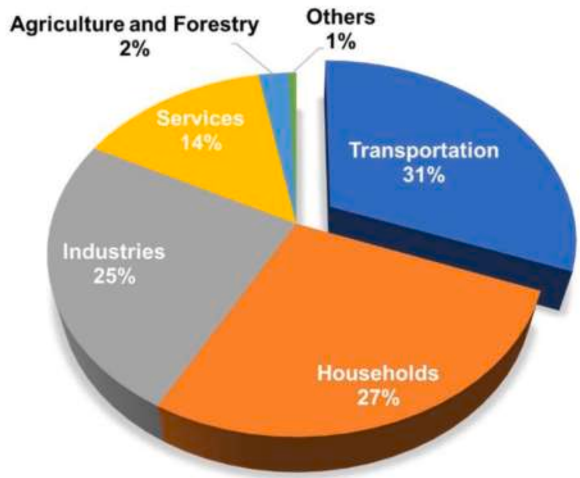

Fig. 2. Proportion of global energy consumption in 2017 [12].

components and lubricants [27].

3. Development of fuel cells

3.1. Proposition of fuel cells

In 1838, "the father of fuel cells" Sir William Robert Grove proposed the gas cell concept that the water splitting process can be reversed to generate electricity [28]. In 1889, Charles Langer and Ludwig Mond tried to design the power generation assembly using coal gas and air and defined the term "fuel cell" [29]. The first fully operational FC was developed in 1939 by Francis Thomas Bacon [30]. This prototype that comprises nickel electrodes and an alkaline catalyst is fed with pure H_2 and O_2. In the 1990s, as one of the important ways to mitigate environmental pollution and demand for oil, the FCEV technology received enormous attention. In 1995, Times magazine listed the FCEV as the first of ten high and new technologies in the 21st century [31,32]. The global major automobile manufacturers have invested a lot of manpower and resources in developing FCEVs and energy conversion devices that can convert chemical energy stored in fuels to electricity and heat electrochemically with high energy efficiency have witnessed tremendous development [33–35]. The timeline of the development of FC technologies is summarized in Fig. 3. Table 2 shows that there are six common types of FCs used in FCEVs depending on their chemical properties and operating characteristics: polymer electrolyte membrane FCs (PEMFCs), solid oxide FCs (SOFCs), direct methanol FCs (DMFCs), alkaline FCs (AFCs), molten carbonate FCs (MCFCs) and phosphoric acid FCs (PAFCs). The operating temperature and power range of different types of FCs are graphically displayed in. Fig. 4.

3.2. State-of-the-Art of fuel cells

Polymer electrolyte membrane FCs (PEMFCs) are also known as proton electrolyte membrane FCs [39]. A PEMFC is composed of two porous electrodes and a proton-conducting polymer electrolyte membrane (PEM) between them. The concept of a single PEMFC and its stack is shown in Fig. 5. The core of the PEMFC is a membrane electrode assembly (MEA) composed of the anode and cathode, which are supplied externally with H_2 and O_2, respectively. There are no environmental problems with acidic or lead runoff and tailpipe emissions. The solid polymer electrolyte that separates the electrodes provides ion conduction [40]. The theoretical potential of a single H_2/O_2 FC is 1.23 V at 25 °C and atmospheric pressure [41,42]. Assuming that all the Gibbs free energy (ΔG) can be converted into electrical energy, the theoretical efficiency of the FC is the ratio of ΔG to the higher calorific value (ΔH) of hydrogen, which is calculated to be 83% [43,44]. The actual efficiency of FCs is normally lower than the theoretical efficiency with a value of around 50% and inversely proportional to the current because of accompanying irreversible processes [45]. The energy efficiency of the FC can be calculated as the ratio of the actual voltage to theoretical reversible voltage at a given temperature and pressure [46]. FCs maintain a constant voltage, without any voltage drop at the end of the shift or in cold locations, but such voltage drop is inevitable in batteries.

The primary PEMFC components include the anode, cathode, PEM, and some catalysts. The internal operation process in the PEMFC is a catalytic electrochemical reaction, in which the voltage loss of cathodic ORR is about 80% of the total loss [48,49]. It has been shown that platinum is the most active catalyst for ORR in an acidic medium [50]. Pt

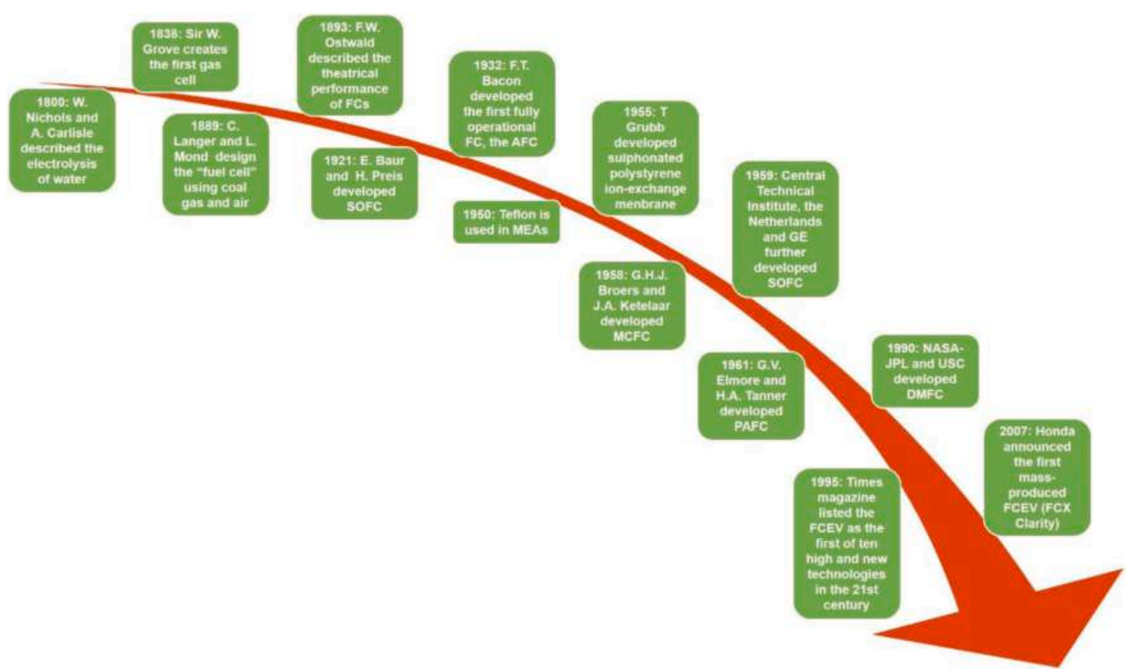

Fig. 3. Timeline of the development of FC technologies.

Table 2
Characteristics of different types of FCs [36-38].

Type	PEMFC	SOFC	DMFC	AFC	MCFC	PAFC
Representative electrolyte	Nafion membrane	Yttria stabilized zirconia	Nafion membrane	KOH solution	Lithium, sodium and/or potassium carbonate solution	Phosphoric acid
Fuel	Pure H_2	H_2, CO, CH_4, etc.	CH_3OH	Pure H_2	H_2, CO, CH_4, etc.	Pure H_2
Efficiency (%)	40–60	~60	~40	~60	~50	~40
Temperature range (°C)	<100	500–1000	60–200	90–100	600–700	150–200
Cell voltage (V)	~1.1	0.8–1	0.2–0.4	~1	0.7–1	~1.1
Stack power (kW)	<1–250	<1–3000	0.001–100	1–100	1–3000	50–1000
Advantage	• Lower operating temperature; • Fast start; • Fewer corrosion and electrolyte problems	• Combined heat and power; • Higher efficiency; • Fuel flexibility; • Hybrid gas turbine	• No need for fuel reformer; • Easy fuel storage	• Fast start and cathode reaction; • Lower material cost; • Lower operating temperature	• Combined heat and power; • Higher efficiency; • Fuel flexibility	• Higher efficiency; • Combined heat and power; • Better tolerance for fuel impurity
Disadvantage	• Expensive catalyst; • Sensitive to fuel impurity	• High operation temperature; • High corrosion; • Poor durability; • Slow start	• Easy to catalyst poisoning; • Low power	• Sensitive to CO_2 impurity	• High operation temperature; • Slow start; • Low power	• Expensive catalyst; • Slow start

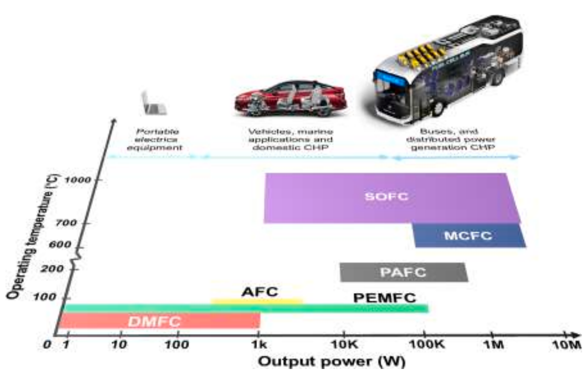

Fig. 4. Operating temperature and power range of different types of FCs [36].

alloys and some Pt-free catalysts have been studied extensively in the laboratory in the attempt to substitute for the expensive Pt-based catalysts [51,52]. However, success is still limited because these alternatives are typically unstable in the commercial fuel cell environment. The carbon-supported platinum (Pt/C) catalyst, in the form of highly dispersed Pt particles with a size of 2–6 nm supported on carbon particles with a size of 15–50 nm, is the most common commercial catalyst in PEMFCs [53,54]. In practice, the PEM must provide high proton conductivity, adequate barrier against mixing, as well as chemical and mechanical stability in the FC environment. The most well-known commercial membrane is Nafion® (perfluoro sulfonyl fluoride ethyl-propyl-vinyl ether, PSEPVE) produced by DuPont. Nafion has a backbone chain of fluorocarbon (CF_2—CF_2) and side chain containing sulfonic functional groups (SO^{3-}, H^+), rendering it efficient in transferring protons while preventing electron transportation [55,56]. The acidity of Nafion is approximately equivalent to that of 0.5 M H_2SO_4 [57]. Similar PEMs developed by other manufacturers include Flemion® from Asahi Glass [58], Aciplex® from Asahi Chemical [59], "C" membrane from Chlorine Engineers [60], Dow® from Dow Chemical [61], and Aquivion® from Solvay [62].

Although hydrogen-fueled PEMFCs have a high-power density, production, transportation, and storage of pure hydrogen are practical hurdles. Methanol with a theoretical energy density of 6.1 kWh kg^{-1} can be stored in the liquid form at ambient pressure and temperature [63]. The easy portability and storage of methanol makes methanol FCs (MFCs) attractive to the transportation industry. Direct MFCs have a similar structure and composition as hydrogen-fueled PEMFCs, as shown in Fig. 6**(a)**. The core of direct MFCs is also the MEA which consists the cathode, anode, and proton-conductive membrane. Both the anode and cathode are composed of a diffusion layer, microporous layer, and catalyst layer [64]. The most widely used membrane is also Nafion® from DuPont and platinum-based catalysts are also necessary in direct MFCs.

The direct MFCs can be operated at room temperature (~40 °C) or elevated temperature (> 100 °C) [68]. The high temperature accelerates the electrode reaction kinetics, resulting in a much higher power (above 100 mW cm^{-2}) compared to the room temperature one (~50–100 mW cm^{-2}) [69,70]. Nevertheless, hydrogen-powered PEMFCs provide higher power densities than direct MFCs at both room and high

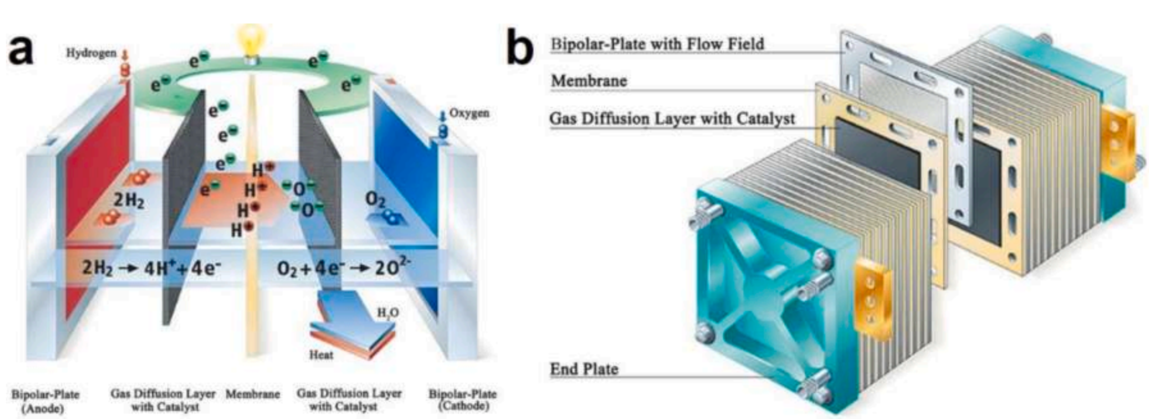

Fig. 5. Concept of a single PEMFC and PEMFC stack [47]. Copyright © 2011 Scientific Research Publishing.

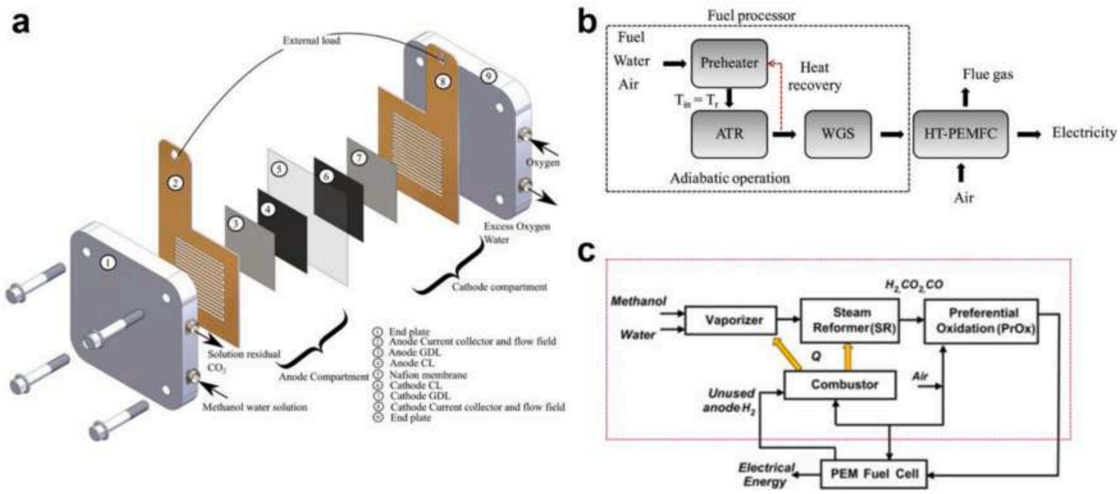

Fig. 6. (a) Schematic of the direct MFC components [65] (Copyright © 2019 John Wiley & Sons, Ltd.), (b) Auto-thermal reforming methanol FC system [66] (Copyright © 2015 Elsevier B.V.), and (c) Steam reforming methanol FC system [67] (Copyright © 2014 Elsevier B.V.).

temperature. Currently, the insufficient power density and energy density limits the utilization of direct MFCs in automobile industry. In this case, methanol is fed into a methanol reformer and the hydrogen-rich gas produced is fed into the PEMFC. This kind of FC is called the reformat methanol fuel cell (RMFC). The methanol reforming and feeding scheme in RMFCs can be classified into auto-thermal reforming (ATR, seeing Fig. 6**(b)**) and steam reforming (SR, seeing Fig. 6**(c)**), as shown below:

$$\text{ATR: } CH_3OH + 1/2O_2 \rightarrow CO_2 + 2H_2 \quad (1)$$

$$\text{SR: } CH_3OH + H_2O \rightarrow CO_2 + 3H_2 \quad (2)$$

The high operating temperature of solid oxide FCs (SOFCs) leads to slow start-up times, so SOFCs are generally not being considered for propulsion in automotive applications. However, SOFCs are being developed for auxiliary power units (APUs) since they can operate from reformed diesel fuel and thus do not require implementation of a hydrogen fuel infrastructure [71]. In particular, APUs for trucks are being developed and will allow truckers to power the sleeper cabins using SOFCs and thus eliminate the need for idling the diesel engine when the driver is resting.

3.3. Degradation of PEMFCs

The PEMFC stack, as well as accessories such as reactant storage, pumps, and other components, make up a PEMFC system. Table 3 outlines the deterioration pathways associated with the electrodes, PEM, gas diffusion layers (GDLs), and bipolar plates in the PEMFC stack. The

Table 3
Major failure modes of different components in PEMFCs [78-81].

Component	PEM	Electrodes	GDLs	Bipolar plates	Sealing gasket
Failure mode	• Mechanical degradation; • Thermal degradation; • Chemical degradation	• Catalyst loss; • Conductivity decrease; • Decreased transportation; • Reformate tolerance loss; • Deteriorated water management	• Mechanical degradation; • Conductivity decrease; • Deteriorated water management	• Mechanical degradation; • Conductivity decrease	• Mechanical degradation
Causes	• Uneven mechanical stress; • Inadequate humidification; • Corrosion of catalysts and seal materials; • Thermal stress and cycles; • Contamination; • Radical attack	• Catalyst sintering; • Corrosion of catalysts or supports; • Mechanical stress; • Contamination; • Change in hydrophobicity	• Degradation of backing materials; • Chemical corrosion; • Mechanical stress; • Change in the hydrophobicity; • Chemical corrosion	• Corrosion; • Oxidation; • Mechanical stress; • Thermal stress and cycles	• Corrosion; • Mechanical stress
Mitigation solution	• Use of radical scavengers such as CeO_2 nanopowder to cathode catalyst ink; • Thicker membrane to delay failure due to membrane thinning; • Use of composite membrane; • Larger radiator; • Wetter running with less RH variability	• More Pt loading; • Particle agglomeration management by novel catalyst geometries and formulations; • Use of inherently durable catalysts such as Pt alloy and Pt membrane with high surface area carbon supports; • Use of inherently durable catalysts such as Pt alloy and Pt membrane with high surface area carbon supports; • Cell-by cell voltage monitoring system; • Limit voltage slew rate; • Drier running with lower RH; • Shut-down to prevent voltage from going up	• Dummy cells on stack ends to prevent condensation; • Gas-purge of anode at shut-down	• New bipolar plate base materials such as Ti, graphite, surface modified or doped metals, and conductive polymer composites	• Limit leaching by Co in cathode catalyst ink

performance of a PEMFC stack is affected by several factors, such as FC design and assembly, degradation of the materials, operational conditions, and impurities or contaminants present in the feedstocks. Performance degradation is unavoidable, but it can be partially limited within time to increase its lifetime. DOE-suggested degradation targets usually require less than 10% loss in the efficiency of an FC system at the end of its lifetime. The degradation mechanisms can basically be subdivided into two families: FC component degradation (membrane, electrocatalysts and CL (catalyst loading), GDL-gas diffusion layer, gaskets, BPs-bipolar plates, sealing) and degradation effects due to operative conditions (air/fuel impurities, load cycle, startup/shutdown, environmental subfreezing conditions) [72]. Membranes can degrade mechanically, thermally, or chemically-electrochemically. Mechanical degradation usually causes early life failure, due to perforations, cracks, tears, or pinholes, which may result from inherent membrane defects, or from improper MEA manufacturing processes. The membrane protonic conductivity drops significantly with a decrease in the water content when the FC is operated at high temperature and under low humidity. Membrane thinning may be caused by a chemical attack from hydrogen peroxide formed by electrochemical reaction of oxygen and hydrogen that have crossed the membrane. Pt-based catalysts are usually employed in PEMFCs: Pt is in the form of highly dispersed metal particles at nanometer scale, usually in the 2–6 nm range. Considering the extremely harsh conditions in which PEMFCs operate, the corrosion of Pt/C catalysts (both catalytic metals and the support materials) is a problem. Nanoparticles inherently show a strong tendency to agglomerate, because of their high specific surface energy. Consequently, when Pt nanoparticles agglomerate to larger ones, the electrochemical surface area of the Pt-based catalysts decreases, and consequently the performance of the PEMFC degrades. The degradation of catalysts involves the two different features of the catalytic metal (Pt or Pt alloys) and the carbon support that influence each other: the catalytic metal, especially Pt, catalyzes the oxidation of carbon, and the oxidation of carbon accelerates Pt sintering. The PEMFC operating environment gradually changes the GDL from hydrophobic to hydrophilic. Likes all other PEMFC components, BPs also suffer from corrosion failure, mainly due to pinhole formation especially when metal plates are used.

Various data-driven and physical model-based methods are used to simulate the deterioration pathways of FEMFCs, analogous to battery deterioration simulations [73–75]. Data laws are used in data-driven methods, but the parameters must be modified if the working conditions are changed [76,77]. Chemical reactions that influence the FC degradation rate, such as carbon support loss, Pt surface area loss, and so on, are accounted for in physical models. Chemical reactions which affect the FC degradation rate such as carbon support loss, Pt surface area loss, etc. are taken into account in physical models. The solutions to enhance the durability of PEMFC stacks need to be developed from the aspects of material improvement and system control strategy optimization, as illustrated in Table 3.

In vehicle implementations, the degradation mechanism is much more complicated and difficult to describe precisely with a theoretical model [82,83]. FEMFC degradation in FCHEVs is inevitable and has a major impact on the system lifetime. However, degradation of the FC is roughly considered by a computational model in the architecture of a health-conscious EMS, which cannot be generalized to all real-driving conditions [84,85]. The FC's complex architectures and reactions as well as modeling challenges are the main reasons for this. Although some prognostic works have been done to assess FC degradation, the actions or control configurations are lacking. As a result, it is needed to design a health-conscious EMS based on state estimation and prognostics which can not only solve the modeling inaccuracy, but also implement automatic corrective control [86]. Presently, health-conscious EMSs are basically categorized into the rule-based EMSs and optimization-based EMSs, as shown in Fig. 7.

3.4. Comparison of fuel cells with batteries

FCs and batteries operate similarly. For example, they both convert chemical energy into electricity by separating the oxidation reaction from the reduction reaction with an electrolyte and conducting electrons from the anode to cathode through an external circuit. FCs are also stacked in a similar way as batteries and high voltage and power outputs can be achieved by connecting several devices in series. The main difference between FCs and batteries is the participation of electrode materials in the electrochemical reactions. During operation, the battery anode and cathode materials are involved in the chemical reactions themselves resulting in consumption or chemical change [88]. This kind of electrode materials change can be reversed by battery recharging or electrode replacement. In contrast, the electrode materials in FCs do not undergo chemical changes theoretically. The materials consumed in the electrochemical reactions are the externally fed fuel and oxidizer, meaning that the FC can be operated continuously with sufficient

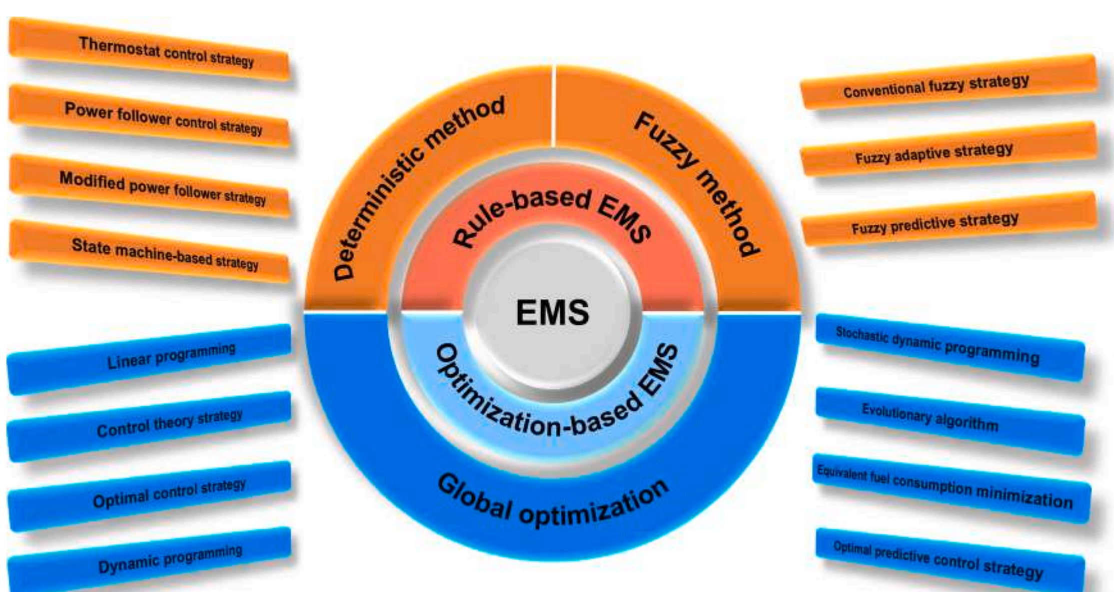

Fig. 7. Health-conscious EMSs for FCHEVs [87].

refueling [89]. Therefore, compared to batteries, FCs are easier to maintain and refuel as onboard power supplies.

The mileage standard of FCEVs is determined by the market and consumer demand. At present, various countries and enterprises have not yet formed a unified and clear standard for the mileage of FCEVs. Due to the higher energy density of FCs, the maximum mileage of FCEVs should be larger than that of pure electric vehicles. The current maximum mileage of mainstream pure electric vehicles can generally reach 400 to 500 km. From Table 3, it can be seen that the maximum mileage for commercial FCEV models can reach about 600 km. Therefore, the mileage is not one of the important factors restricting the commercialization of FCEVs. It is worth mentioning that the mileage of electric vehicles (including pure electric vehicles and FCEVs) is not the larger the better. The excessive mileage exceeding the demand is accompanied by the oversized batteries and hydrogen storage tanks, resulting in a lack of energy economy and cost performance.

After solving the cold start problem, the mileage of FCEVs in winter is the longest in the four seasons with the help of a comprehensive thermal management system. This is because FCs generate waste heat during operation like traditional diesel engines [90]. The waste heat must be specifically dissipated by the heat dissipation system in summer, but it can be used to heat the passenger compartment and batteries in winter. Therefore, in winter, the passenger compartment of a FCEV can maintain a comfortable temperature without consuming the hydrogen. However, pure electric vehicles must consume the battery electricity for heating in winter, which means that their millage in winter will be shorter.

4. Market and policies

4.1. Fuel cell market in the automobile industry

The market for hydrogen FCEVs was worth $651.9 million in 2018 and is expected to reach $42.0389 billion in 2026, corresponding to an annual compound growth rate of 66.9% [91,92], and more than 50% of new public and freight vehicles are expected to be powered by FCs and batteries [93]. Commercial FCEVs for passengers were firstly introduced by Toyota and Honda in Japan and Hyundai in South Korea and it was followed by other major car brands around the world. FC-powered trucks, forklifts, and other heavy vehicles have also been introduced [94]. However, at the current stage of technology maturity, the manufacturing cost of a medium-size FCEV is still about 50% higher than that of its ICE counterpart [95]. Compared to FCEVs, battery electric vehicles are growing faster in the passenger vehicle market, especially for short-distance urban transportation [96]. In contrast, FCEVs have a larger share in the long-range and heavy-duty transportation markets [97]. For example, FC electric buses are being promoted in Europe and China and in fact, have already been deployed in most major cities [98]. The H2Bus Europe consortium plans to build 1000 new FC electric buses, 600 of which will be completed by 2023 [99]. The passenger vehicle market is expected to be the largest source of growth for FCs in the coming decades. According to the 2019 report of Market Research Future (MRFR), the global automotive FC market is expected to grow at a compound annual growth rate of 25% until 2025 [100]. The Asia-Pacific region (APAC) is predicted to lead the global demand in this market followed by North America [101]. These two regions are currently the most economically dynamic regions in the world and major sources of greenhouse gasses. The relevant governments are actively taking measures to reduce the carbon footprint from the transportation sector, especially by vigorously promoting cleaner electric vehicles. FCs as a green alternative to traditional batteries are getting more attention. Modified modularization, higher efficiency, longer driving range, and construction of hydrogen supply for onboard FC systems are the future commercial development directions. Major original equipment manufacturers are also paying increasing attention to the market of FCEVs [102]. The onboard FC system can be divided into the fuel stack, fuel processor, and power regulator. The fuel stack is a key component that dominates the market. Based on the output power, the automotive FC market can be divided into two ranges: one located at 100–200 kW and the other one above 200 kW [103].

Japan and South Korea are in the leading position of FCEVs and occupy most of the market share. As shown in Fig. 8, from 2016 to 2019, the global sales and holdings of FCEVs showed exponential growth year by year. Among them, only the Toyota Mirai, Honda Clarity Fuel Cell, Hyundai ix35 FCEV, and Hyundai Nexo are truly mass-produced models. Currently, the Toyota Mirai tops the global sale of FCEVs. Japan, South Korea, USA, Europe, and China are expediting construction of hydrogen refueling stations to promote commercialization of FCEVs [104]. Furthermore, hydrogen refueling stations have been built in Denmark [105] and the world's vision and plans for the development of FCEVs are shown in Fig. 9.

4.2. Policies and subsidies for hydrogen FCEVs

It should also be noted that most of the FC development projects have been subsidized by governments [111,112]. Many countries in the world have strong support policies for the use of hydrogen energy. For example, Japan, the European Union, the US, China and South Korea have launched strong policies to support the development and popularization of hydrogen FCEVs. Policy subsidies are mainly concentrated in the consumption link, benefiting consumers by means of purchase tax exemptions or purchase subsidies [113,114]. Fiscal subsidy is only one aspect of industrial support, which should be supplemented with tax relief, government procurement and other guidance.

The US put forward the concept of hydrogen economy as early as 1970, and was the first country in the world to promote the development hydrogen energy and FCs as a national energy strategy. In the past ten years, the US government has spent more than $1.6 billion to provide continuous support for hydrogen energy and FCEVs, and has formulated industry standards and subsidies for hydrogen infrastructures and hydrogen utilization. The DOE is mainly responsible for the implementation of the hydrogen energy strategy in the US. The new bipartisan budget bill passed in 2018 decided to continually provide tax exemptions for FC applications in the transportation sector and stationary FC power generation. California, as the world's first market to realize the retail of FCEVs, has indispensable policy guidance and subsidies. It requires automakers to sell zero-emission vehicles and provides not only tax exemptions for the purchase of FCEVs, but also additional subsidies [115]. Specifically, California grants a subsidy of $5000 to consumers who purchase hydrogen FCEVs and also adopts a zero-emission vehicle points system for automobile manufacturers. The state government has increased public confidence in FCEVs through demonstration projects and coordinated with automobile manufacturers to set sales targets for FCEVs. In terms of hydrogen refueling station construction, incentives are provided through subsidies for construction, operation and maintenance subsidies. As of 2020, the number of FCEV-related patents in the US is second only to Japan, and it has the world's largest liquid hydrogen production capacity. The total number of FCEVs in operation has reached 8285 in the US.

Japan set an ambition to become the first country in the world to realize a hydrogen energy society. Over the years, the Japanese government has successively released the Japan Recovery Strategy, the Energy Strategic Plan, the Basic Hydrogen Energy Strategy, and the Strategic Roadmap for Hydrogen and Fuel Cells, planning the technological path for the realization of the hydrogen society strategy [116]. In the past 30 years, the Japanese government has invested hundreds of billions of yen in the development and promotion of hydrogen energy and FCEVs and the construction of hydrogen infrastructures. Japan specially established H2 Mobility (JHyM) to plan and deploy the construction of hydrogen supply stations. The Japanese government uses public funds to subsidize part of the cost of purchasing different types of clean energy vehicles such as battery electric vehicles, plug-in hybrid

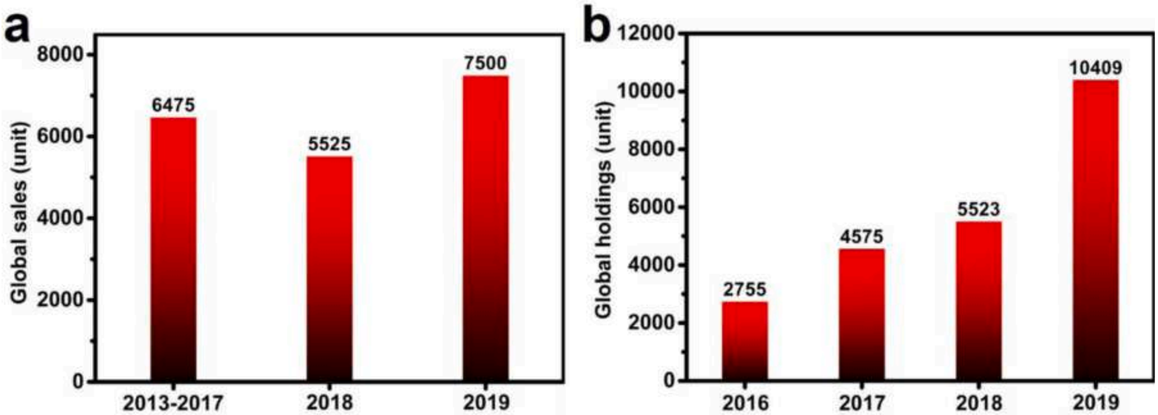

Fig. 8. (a) Global sales and (b) Global Holdings of FCEVs [106].

Fig. 9. World's vision and plans for the development of FCEVs (HRS: hydrogen refueling station) [104,107–110].

vehicles, FCEVs, and clean diesel vehicles. The specific subsidy amount is calculated based on the cleanliness level of vehicles. FCEVs are the most subsidized of all types. South Korea released its hydrogen and FCEV roadmap in January 2019. At present, the South Korean government subsidizes half of the price of FCEVs and half of the cost of hydrogen refueling station construction to rapidly expand the hydrogen electrical network [113].

The EU Joint Fuel Cell and Hydrogen Action Plan (FCH JU) has provided strong financial support and set a technical blueprint for the development and promotion of FCEVs in Europe [113]. The plan provided financial support of up to 665 million euros between 2014 and 2020. In March 2020, the European Clean Hydrogen Alliance was established. In July 2020, the EU issued the EU Energy System Integration Strategy and the EU Hydrogen Driven Economy Strategy to continue to support hydrogen energy and FCEVs. Germany is a leading practitioner of European hydrogen energy and FCEV programs. With the support of government finances, H2 Mobility GmbH, established in Germany, is responsible for the construction of a nationwide hydrogen refueling station network. A key feature of the German deployment plan is to try to make hydrogen price competitive with gasoline. At present, the price of hydrogen provided by Germany is about \$10 kg^{-1}, and the price of gasoline is \$5 gal^{-1}, which makes Germany a leading position in the construction of hydrogen supply infrastructures. Germany implements a series of subsidy policies for EV consumers such as vehicle purchase subsidies as well as tax reduction and exemptions. Consumers who purchase pure EVs before 2020 also enjoy a 10-year exemption from car taxes.

Since 2014, China has successively introduced a series of government policies and measures to support and encourage the commercialization of hydrogen energy and FCEVs, indicating China's determination in the development of FCEVs. In recent years, a number of preferential policies have been introduced in China to accelerate the popularity of FCEVs. The Chinese government provides subsidies to both EV consumers and manufacturers to stimulate the development of new-energy automobiles [117]. The FCEV subsidy standard issued by the China Ministry of Industry and Information Technology (MIIT) in 2018 stipulates that the Chinese government grants subsidies of up to 200,000 RMB, 300,000 RMB, and 500,000 RMB to FC passenger vehicles, medium-sized commercial vehicles, and large commercial vehicles, respectively. In December 2019, the MIIT issued the New Energy Vehicle Industry Development Plan to promote the construction of hydrogen fuel infrastructures. In April 2020, the Notice on Improving the Financial Subsidy Policy for the Promotion and Application of New Energy Vehicles made it clear that the Chinese government would continue to support and expand the financial subsidies for the promotion and application of new energy vehicles, and complete the transformation of "incentives instead of subsidies" for FCEV demonstration and purchase by 2022.

5. Recent progress of fuel cells in the automobile industry

5.1. Development of various types of fuel cells

Cars, buses, forklifts, scooters, airport vehicles, golf carts, locomotives, trams, aircrafts, ferries, and underwater vehicles are all terminal applications of PEMFC systems in the transportation field. PEMFC-powered electric vehicles have several competitive edges over battery-powered electric vehicles. First, increased operational efficiency is achieved since time-consuming battery recharging process is not required (refueling in less than 5 min). Secondly, hydrogen refueling stations can be distributed around a central storage tank requiring smaller room than a dispersed charging pile parking lot. Thirdly, a constant voltage can be maintained during the whole operation, unlike batteries in the late-term or frigid weather showing a significant voltage drop. Fourthly, there is no risk of electrolyte leakage. Price remains the most important factor hindering commercialization of FCEVs. At present, operation of a PEMFC costs \$45–60 kW^{-1} compared to \$25–35 kW^{-1} for an internal combustion engine. If the price per kilowatt of PEMFCs is not reduced, the public will still favor internal combustion engines. PEMFCs can also be promoted as auxiliary power units (APUs) for the non-propulsion power in refitted old vehicles [118].

The elevated operating temperature of SOFCs results in a long startup time and so SOFCs are generally not considered for propulsion in automotive applications [71]. However, SOFCs for APUs are being developed since reformed gasoline and diesel can be used in these systems without construction of hydrogen supply infrastructures [119]. For example, SOFC-based APUs are being developed to eliminate the need for idling diesel engines when the cars are not in motion.

Although the technology of PEMFCs-driven FCEVs is relatively mature, owing to the technical barriers of hydrogen storage and transportation, commercial promotion still depends on major hydrogen infrastructure construction. DMFCs are being developed as the power units for FCEVs by Ballard Power System Inc. in collaboration with Daimler-Chrysler, IRD Fuel Cell A/S, Siemens AG, Los Alamos National Laboratory, and others. Current gas stations can be easily converted into methanol fueling stations. Most onboard DMFCs are operated at a high temperature and pressure (at approximately 110 °C, 3 bar, vapor feed) to enhance the output power [120].

The global development and commercialization of FCEVs can be roughly divided into four stages. In the first stage (before year 2000), international famous automobile manufacturers launched their own FCEV prototypes. In the second stage (2000–2010), the key technologies of FCEVs were developed, demonstrated, tested, and validated. In the third stage (2010–2015), FCEVs were partially commercialized in some specific areas. In the fourth stage (after 2015), FCEVs manufactured by Toyota Mirai, Honda Clarity Fuel Cell, Hyundai Tucson ix35 FCEV and NEXO, are being sold in the world reflecting the early commercialization stage of FCEVs [95,121].

5.2. Fuel cell-battery hybrid systems

The dynamic response of pure FC power systems is not fast enough and so pure FC systems have difficulty in outputting power peaks during startup and acceleration, nor can they recover the energy generated during braking [122]. Therefore, an auxiliary battery system is helpful in the frequently changing driving scenario. The FC-battery hybrid systems have the advantages of (i) compensating the output power of FCs, (ii) recycling the feedback energy of the motors during decelerating and braking to increase the total efficiency, and (iii) reducing the voltage fluctuation of the power systems [123]. The FC-battery hybrid systems have a longer service life than single FC systems and battery systems [124]. Coordination and optimal operation of the FC and battery is managed by the energy management system (EMS). In 2014, FC-battery hybrid vehicles entered the market with the launching of the Mirai by Toyota [125,126]. The basic structure of PEMFC-battery hybrid systems is shown in Fig. 10 and a variety of deformation configurations enable PEMFC-battery hybrid systems to flexibly adapt to different vehicle types [127,128]. Currently, Japan and the US have used the PEMFC-battery hybrid systems in electric buses and trucks with the output power up to 120–320 kW, continuous mileage up to 250–480 km, and mean durability close to 10,000 h [129]. A SOFC-battery hybrid system is another option, in which the SOFC stack can either propel the vehicle or charge the battery, while the battery is responsible for the fast startup and recovery of energy through braking. The SOFC-battery hybrid systems have comparable driving range and acceleration time as PEMFCs, albeit a lower top speed [130].

5.3. Safety standards of FCEVs

Safety standards have been developed by transportation organizations in the world. SAE established by the United States is the most widely used vehicle standard. ISO from the international community, EC from the European Union, GB/T from China, CNS from Japan and Taiwan, KS from Korea, and CGA and CSA from the United States are the other standards. System design and testing of FCHEVs, safety concerns, efficiency, fuel systems, performance, and durability are regulated in these standards. These criteria apply to the FC stack as well. **Table S1** in Supplementary Materials summarizes the automotive standards and requirements.

5.4. Application examples of fuel cells electric vehicles

The landmark events and milestones in the development of FCEVs are illustrated in Fig. 11. Although the growth of passenger vehicles in the world has created a huge potential market for FC technology, commercialization of FCEVs still faces many challenges including the reliability of FC systems, construction of fuel infrastructures, broader market acceptance, and so on [93]. In particular, the lack of hydrogen refueling infrastructures is the biggest hurdle. There are many prototypes of configured methanol or gasoline reformers with the onboard FC systems, although this useful reformatting adds hardware complexity, maintenance difficulty, and extra costs [132,133]. Based on the current state of the market and technology, a number of leading automotive companies are playing critical roles in the near-term development of FCs. Fig. 12 shows the general scheme of a FCEV. The FCEV models manufactured by major car companies are listed in **Table S2** and some of them are highlighted in this paper.

5.4.1. Fuel cells for private transportation

5.4.1.1. GM concept (2001 and 2005). In 2001, GM showed off its non-commercial FCEV “HydroGen3” (Fig. 13**(a)**) at the Frankfurt Motor Show. The “HydroGen3” drive system works by generating electricity from a 472 × 251 × 496 mm battery pack of 200 individual FCs connected in series, outputting a voltage of 125–200 V, power of 94 kW and power density of 0.94 kW kg^{-1}. The electricity generated by the FC pack is fed into a 60 kW/82 hp three-phase asynchronous motor with almost no noise generation. The acceleration time from 0 to 100 km h^{-1} is about 16 s and the maximum speed is 160 km h^{-1}. The hydrogen storage systems are divided into two types. The first type of tank stores 68-L liquid hydrogen at a temperature of −253 °C and the other type stores 416 kg high-pressure hydrogen at a maximum pressure of 700 × 10^5 Pa. The driving distance for one fueling can reach 400 and 270 km, respectively.

The GM Sequel Hydrogen Fuel Cell (Fig. 13**(b)**) debuted at the 2005 Shanghai Auto Show has been considered a classic in the FCEV field. The Sequel is a breakthrough in drivability compared to its predecessor and its performance is comparable to that of ICE-powered vehicles. Its FC stack possesses a rated output power of 73 kW/98 hp supplemented by a lithium-ion battery system of 65 kW. Acceleration from start to 48 km h

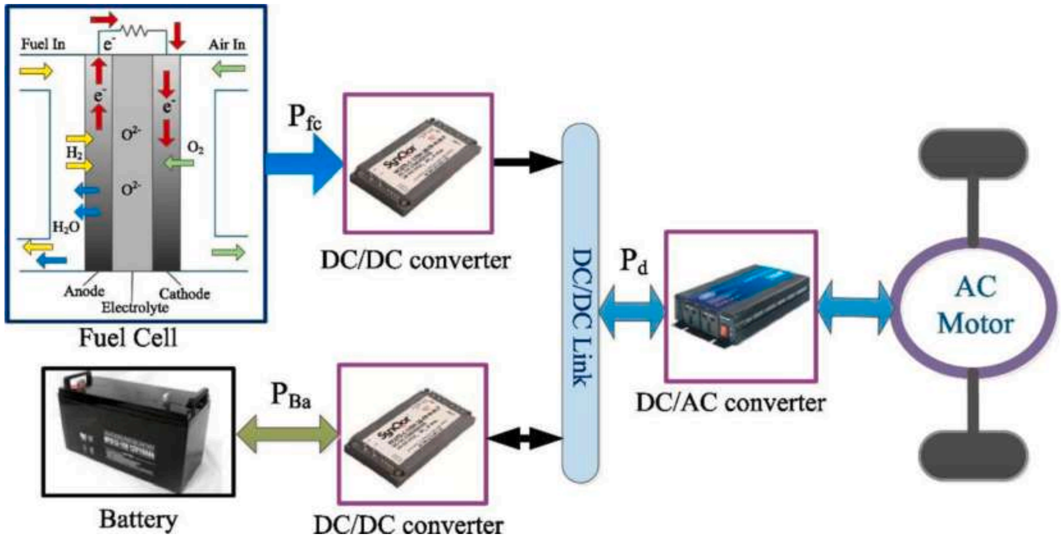

Fig. 10. Basic structure of the PEMFC-battery hybrid systems [131]. Copyright © 2018 Elsevier B.V.

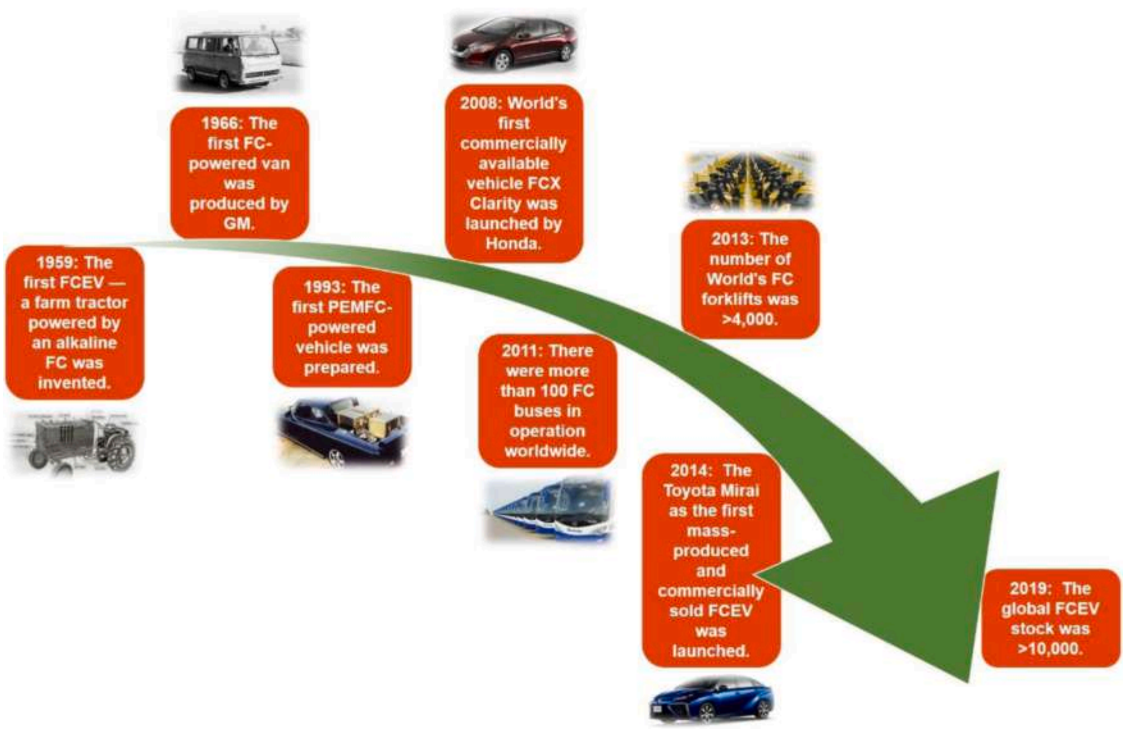

Fig. 11. Landmark events and milestones in the development of FCEVs [107].

$^{-1}$ takes only 3 s and to 96 km h $^{-1}$ takes less than 10 s, with the total torque provided by the hub motor reaching an astonishing torque of 3398 N.m. 8 kg of high-pressure (700 bar) hydrogen is stored in three carbon-composite tanks installed under the floor of the cabin leading to a road haul of more than 480 km.

5.4.1.2. Toyota Mirai (2014). The Mirai (Fig. 13**(c)**) is a mid-size FCEV launched at the 2014 Los Angeles Auto Show by Toyota, representing the first-generation FCEV to be mass-produced and sold commercially. The Mirai is very popular with the US, Japan and Europe markets. The powertrain of the Mirai called the Toyota FC Stack (TFSC) is different from that of a conventional gasoline car or pure electric vehicle. It is a hybrid system with the FC stack as the core component. The Mirai's TFSC has a maximum power output of 114 kW and specific power output of 3.1 kW L $^{-1}$. The maximum torque is 335 N.m, and it can reach 100 km h $^{-1}$ in 10 s, which can fully meet the daily driving needs. With a maximum range of 644 km and combined city/highway fuel efficiency of 0.8 kg/100 km, the Mirai is rated by the US Environmental Protection Agency (EPA) as the most fuel-efficient hydrogen FCEV as well as the vehicle with the longest range, according to a 2016 report. The Mirai uses much fewer platinum catalysts than other models and reducing the consumption of precious platinum pares the cost of FCEVs to improve the competitiveness. In summary, the Mirai delivers very competitive comprehensive performance among electric vehicles on the market, but the amount of hydrogen infrastructure to support the vehicles is not satisfactory thus limiting the potential market.

5.4.1.3. Honda clarity fuel cell (2016). The Honda Clarity Fuel Cell

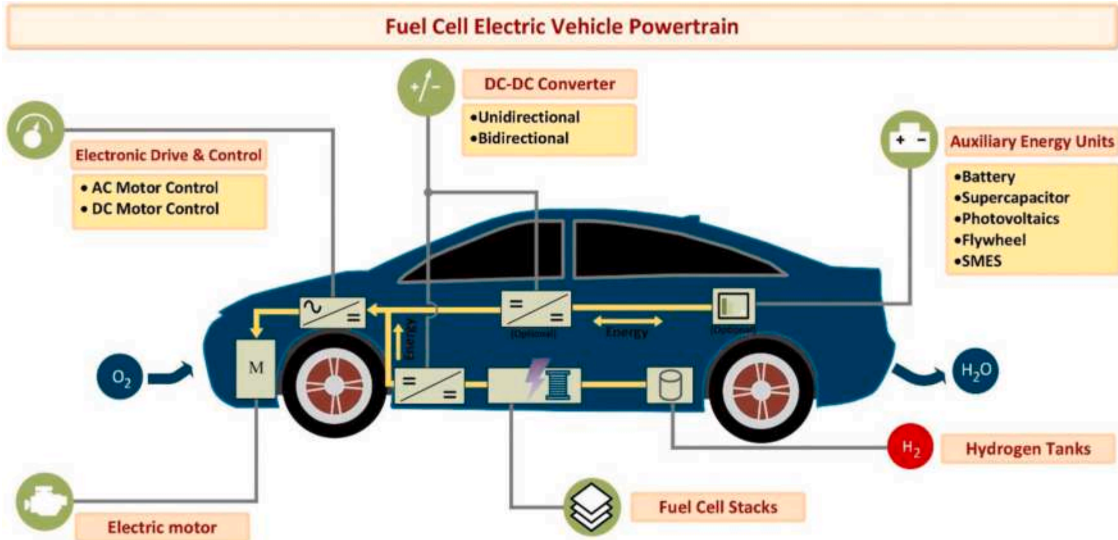

Fig. 12. General scheme of a FCEV [2]. Copyright © 2021 Elsevier B.V.

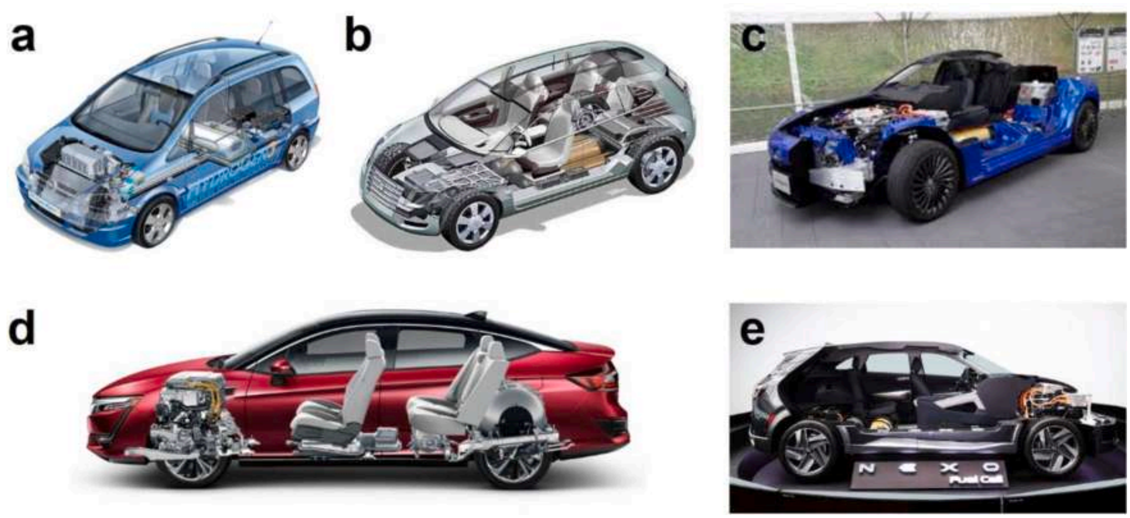

Fig. 13. Depictions of typical FCEVs: (a) GM HydroGen3, (b) GM Sequel Hydrogen Fuel Cell, (c) Toyota Mirai, (d) Honda Clarity Fuel Cell, and (e) Hyundai Nexo.

(Fig. 13**(d)**) is launched in 2016 and currently only available in Southern California. The Honda Clarity Fuel Cell has a maximum range of 589 km, wining the highest EPA range rating in the US among all the zero-emission automobiles including both FC and battery-powered electric vehicles, until the debut of Hyundai Nexo in 2018. Its FC-powered electric motor has an output power of 103 kW/138 hp. In the 2017 report of EPA, the Clarity Fuel Cell is rated as the highest fuel-economical model among all the FCEVs in both city driving (68 MPGe) and combined city/highway driving (67 MPGe).

5.4.1.4. Hyundai nexo (2018). The Hyundai Nexo (Fig. 13**(e)**) is a mid-size hydrogen FC-powered SUV unveiled in 2018 as the flagship model of Hyundai's Eco Series. Its electric motor is powered by a PEMFC with the only emission of water. The car model is designed based on the "sandwich" concept to provide maximum space for both passengers and propulsion components. The main attraction of Nexo is its reliable high-capacity hydrogen storage system which consists of three tanks with a total volume of 59 L and can store 6.4 kg of compressed hydrogen on-board. The more powerful drivetrain delivers a power output of 120 kW (163.2 PS) and large driving range of 611 km. The key indicators of the above typical FCEVs launched by major brands have been summarized in Table 4.

5.4.2. Fuel cells in public transportation

Buses are one of the major candidates for FCEVs due to advantages over private cars such as the higher power demand, fixed routes, more space availability, and easy access to refueling points [134,135]. Operation of FC-powered buses costs 30–40% less than buses powered by diesel or natural gas [93,136]. On account of the large size of buses, sufficient hydrogen can be easily stored on board, usually on the roof, making them much safer than private cars. In addition, buses are centrally parked at the same station, meaning that one central hydrogen infrastructure can meet the refueling needs. The diagram of the main structure of a FC-powered bus is shown in Fig. 14. The most common fuel for FC-powered buses is compressed hydrogen, although methanol is another viable option. The PEMFC stack can generate an average power output of 200 kW and is usually installed on buses with length of 9–12 m [137]. The first FC-powered bus was built in 1990 and major automobile manufacturers including Toyota, Honda, Volvo, Daimler-Chrysler, BYD and Yutong have launched FC bus prototypes in the last three decades. The FC-powered buses and trucks launched by global major brands are listed in **Table S3**. The ZeEUS eBus Report

Table 4
Key indicators of typical FCEVs launched by major brands.

Model	Power output	Power density	FC durability	Maximum mileage	Fuel efficiency (H_2)	Sales	Selling price
GM HydroGen3	Hydrogen fuel cell of 94 kW; NiMH battery of 35 kW	1.6 kW/L	>4000 h	400 km	1.15 kg/100 km	Only concept	Concept
GM Sequel Hydrogen Fuel Cell	Hydrogen fuel cell of 73 kW; Li-ion battery of 65 kW	3.1 W/L	–	480 km	1.64 kg/100 km	Only concept	Concept
Toyota Mirai	114 kW	3.1 kW/L	>5000 h	644 km	0.8 kg/100 km	12,015 (from 2014 to 2020)	$50,495 in 2021
Honda Clarity Fuel Cell	103 kW	3.1 kW/L	>5000 h	589 km	0.77 kg/100 km	1900 (from 2016 to 2020)	$59,445 in 2021
Hyundai Nexo	120 kW	3.1 kW/L	>5500 h	611 km	0.84 kg/100km	12,717 (from 2018 to 2020)	$57,343 in 2021

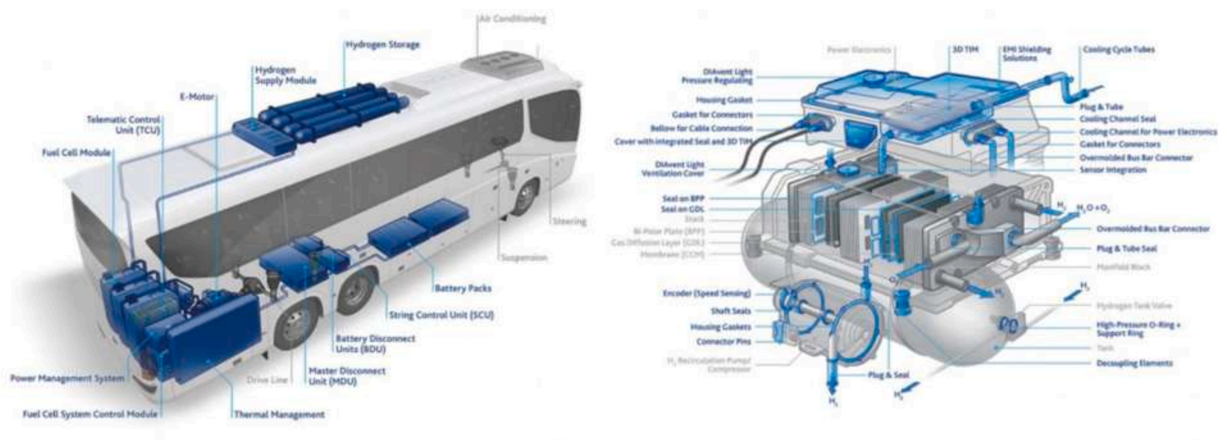

Fig. 14. Diagram of the main structure of a FC-powered bus [139].

Number 2 of the International Association of Public Transport (UITP) points out that the FC urban bus market in Europe is expected to be 10% in 2030 [138].

6. Challenges for commercialization of FCEVs

6.1. High cost of FC stack production and maintenance

Hydrogen FCs not only require complex manufacturing processes, but also high-cost platinum catalysts and electrolyte membranes. Platinum is currently used as a catalyst in hydrogen FCs but the high price remains a major hurdle plaguing the development of FCs until alternatives are found. In commercial production, the catalyst accounts for about 40% of the total cost of the onboard FC stack, as shown in Fig. 15. The US Department of Energy (DOE) has set a target to reduce the usage of platinum catalysts to 0.125 mg cm^{-2} by 2020 [140]. Since the electrolyte membranes in hydrogen FCs must have strong oxidation and reduction stability, the price of the electrolyte membrane remains high. Currently, the PEM used in hydrogen FCs costs about \$600 m^{-2}. The bipolar plates in the hydrogen FCs use graphite materials and manufacturing of graphite bipolar plates needs to be performed at a temperature above 2500 °C [140]. The graphite bipolar plate is required to meet the strict impermeability requirements resulting in excessively high processing costs [55]. Taking the Toyota Mirai as an example, when the annual production capacity reaches 500,000 units, the cost of catalysts, membranes, and bipolar plates will account for about 80% of the total cost of the FC system [141]. According to the US Department of Energy, the current operating cost of FCEVs is \$45–60 kW^{-1}. To provide more affordable FCEVs, it is also necessary to recover valuable FC materials and components from obsolete ones [142].

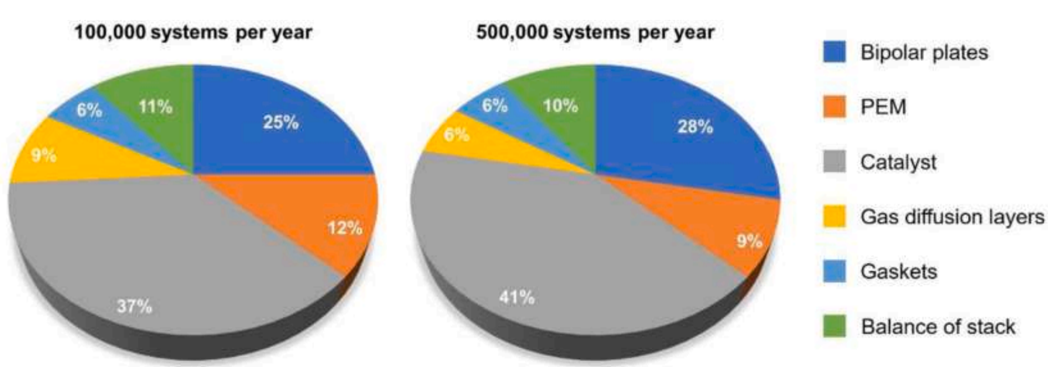

Fig. 15. Cost breakdown of the Toyota Mirai FC system [141].

6.2. Insufficient hydrogen supply facilities

Hydrogen is not a natural source and must be produced by coal gasification, natural gas reforming, or hydrolysis, all of which consume electricity or other form of energy. Because the purity of hydrogen is crucial to the stable operation of FCs, the high-purity hydrogen for fueling FCs is mainly produced by electrolysis of water, which requires a large amount of energy and increases the cost [143]. Moreover, owing to the small density of hydrogen, high pressure vessels are needed to ensure the driving range of FCEVs [144]. The compression process of hydrogen also consumes energy and the cost of hydrogen tank manufacturing is high due to the need to make the tanks as light as possible while meeting the pressure standard [145]. Therefore, the cost control of the whole process of hydrogen production, purification, compression, and storage impacts the economics and attractiveness of hydrogen FCEVs. Progress of FCEVs is anticipated with installation of more hydrogen refueling stations. However, the high cost of constructing hydrogen refueling stations is also one of the factors restricting the development of FCEVs [111]. Construction of a medium-sized hydrogen refueling station costs $ 2.8 to 3.5 million in the US, $ 5 to 5.5 million in Japan, and $ 2 to 2.5 million in China [146]. In recent years, the number of hydrogen refueling stations has been increasing and the global data from 2014 to 2019 are presented in Fig. 16**(a)**. At the end of 2019, the number of hydrogen refueling stations worldwide reached 432, 36% in Europe, 48% in Asia, and 15% in North America, as shown in Fig. 16**(b)**. On a whole, the number of hydrogen refueling stations in the world is still extremely small thereby hindering commercialization and adoption of FCEVs [95].

6.3. Insufficient reliability

The reliability of FCs is critical to commercialization and end-user satisfaction. Compared to ICEs, FCs can generate power at a higher energy efficiency without noise and polluting exhausts [147]. The main limitations for FCs are coupled with the reliability and extra cost for repair [148]. The lifetime of FCs is influenced by environmental factors including variable start-stop, acceleration-deceleration, partial pressure of gasses, humidity, contact load, and temperature variation [149-154]. By calculation, 10% stack element failure can increase the cost of the FC system by 60%, especially in the integrated automotive FC system. The service life of PEMFCs is highly dependent on the degradation rate of the PEM [155] and distortion and formation of pinholes are common problems affecting the operation of FC stacks [156,157]. The feasible proton conductivity of the PEM should be $\geq$ 0.1 S cm^{-1} and the gas permeability needs to be $\leq 10^{-11}$ mol.O_2 cm^{-1} s $^{-1}$ kPa^{-1} and $\leq 10^{-12}$ mol.H_2 cm^{-1} s $^{-1}$ kPa^{-1} [158]. Besides, the Pt catalyst may lose electrochemical active surface area and be poisoned after long-term operation [155]. The resistance of bipolar plates requires to be $\leq$ 50 mΩ cm^2 and they should have a tight seal to prevent leakage during operation [159,160]. Therefore, the repeatability and durability should be improved [161]. Maintaining the FC cell channel flow distribution in a constant state can prolong its life and reduce maintenance costs, although it is still challenging based on the existing technology [162]. An optimized geometry design of the bipolar plates can be used to curb the uneven flow distribution in FCs to increase the durability [163,164]. The V-I curve and H_2 consumption-power curve in Fig. 17 is commonly monitored to estimate the operation characteristics of the FC. In addition, for a more precise lifetime forecast, the attenuation mechanism, failure mechanism, and life prediction methods must be thoroughly understood and developed.

6.4. Slow cold start

Cold start can be problematic for PEMFC stacks when internal water freezes, as ice can block the flow channels of the electrodes and damage the MEA resulting in performance degradation [165]. Therefore, subfreezing cold start is essential for PEMFC commercialization and the critical components, cold start techniques, and auxiliary powers should be optimized to improve the low-temperature performance [166]. The membrane conductivity is reduced at low temperature and the voltage suffers as well. Therefore, the primary optimization focus is to enhance the water uptake ability and conductivity at subfreezing temperature. The SPEEK/PVA/PA composite membrane showed the high and stable proton conductivity of 5.30 × 10–2 S/cm at −30 °C after 7 temperature cycles from −30 °C to 30 °C, and retained 3.33 × 10–2 S/cm at −30 °C after 980 h continuous testing at −30 °C [167]. The POM-based acid--base adduct in SBA-15 called HPW-HSN@SBA-15 has been reported to have a high proton conductivity of 6.8 × 10–5 S cm-1 at −40 °C [168]. Cold start may be affected by the ionomer content and thickness of the catalyst layers. A high Nafion ionomer content can inhibit gas transfer to the catalytic active sites, whereas a low content can result in weak catalyst particle-to-electrolyte connection. It has been shown that a Nafion content of 25 wt% is optimal [169]. Effects of ionomer/carbon (I/C) ratio on the cold start of PEMFCs was systematically investigated by experiments and theoretical calculations [170]. When the ionomer/carbon (I/C) ratio is between 0.7 and 1.7, both the increase of I/C ratio and the decrease of temperature will lead to the deterioration of cold start performance. However, at temperatures below −5 °C, the total product water is nearly the same regardless of the I/C ratio due to rapid freezing before the water adsorption of membranes. The operation time of PEMFCs is extended at a temperature range of −4.2 °C to −10.0 °C using a new form of microporous layer with plane-distributed wettability, in which hydrophilic and hydrophobic rows are arrayed alternately in the in-plane direction [171]. Ice melting can be accelerated during cold start by reducing the porosity of the gas diffusion layer

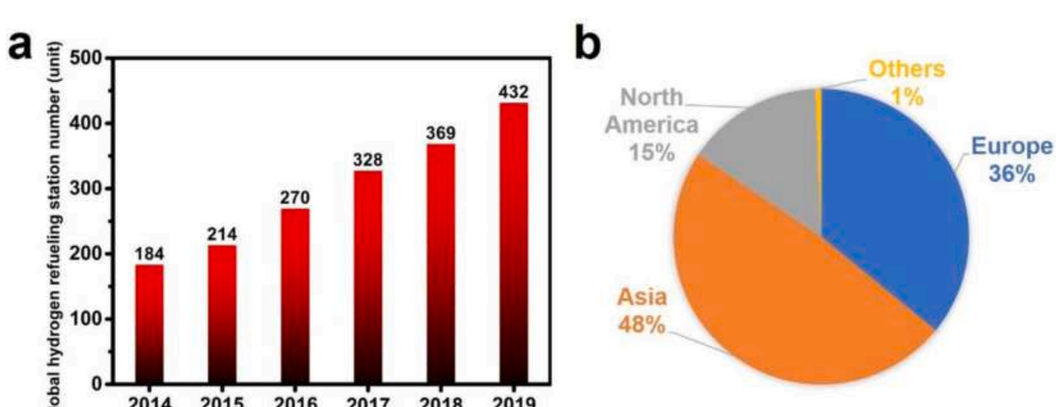

Fig. 16. (a) Statistics on the number of global hydrogen refueling stations from 2014 to 2019 and (b) Global regional distribution of hydrogen refueling stations in 2019 [106].

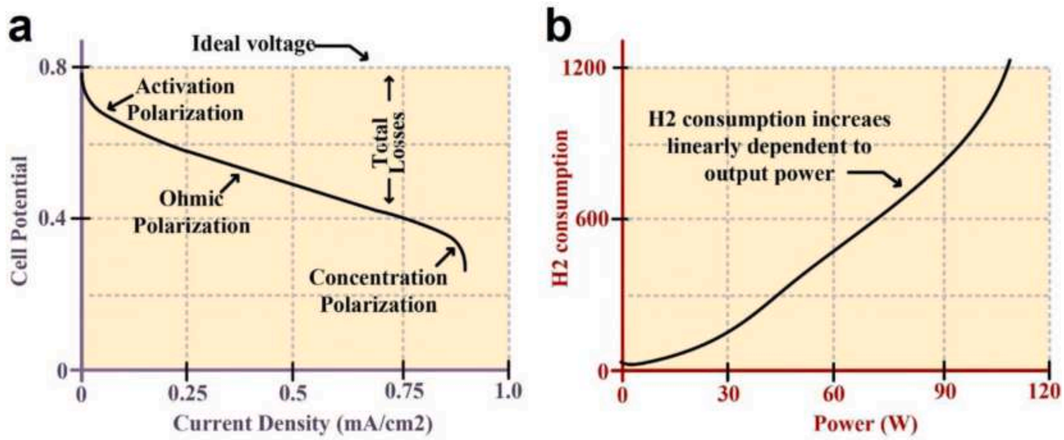

Fig. 17. Typical (a) V-I cure and (b) H_2 consumption-power curve of a PEMFC [111]. Copyright © 2021 Elsevier B.V.

[172]. When the porosity is high, the number of carbon fiber layers has a greater impact on the ice melting than the fiber length, while when the porosity is low, the effect is opposite. The cold start ability may also be improved by designing the materials and structures of the bipolar plates. Vanadium oxide thin films fabricated onto metallic bipolar plates such as 316 L and 446 M stainless steel can deliver competitive cold start output at −20 °C [173]. Optimized power control strategies provide effective cold start methods [174]. With regard to the heating time, the constant voltage cold start strategy outperforms the constant current strategy [175]. Real-time control of the operating current is possible thanks to an internal-based adaptive strategy that maximizes the produced heat flux [176]. The performance of maximum power mode based real-time adaptive cold start strategies in a PEMFC stack highly depends on the selection of a precise model and suitable model selection will enhance the cold start-up performance [177]. Power off purging and heating are two effective ways to the cold start of PEMFC systems, and according to different heating sources, the heating schemes can be divided into self-heating strategy and auxiliary heating strategy [178]. The aim of purging the solution is to reduce the amount of residual water in the PEMFC before the next start. In order to improve the temperature uniformity during the cold start process, a reverse air supply device (RFU) was designed to start the PEMFC reactor within 79 s with a uniform temperature distribution of −32 °C [179]. The transport phenomenon as well as spatial distribution of water and temperature can be analyzed by multi-dimensional cold-start models. An online self cold startup methodology consisting of a water extraction step after the PEMFC shutdown and a self-heating step during the cold startup was invented, which maximizes the internal heat of PEMFC according to the variation of operating parameters, attempts to maintain a high current density, and improves the performance by increasing the hydration and temperature of the PEM [180]. The cold start processes of PEMFCs with zigzag-channeled flow field (ZZFF) has been numerically studied and it was found that ZZFF can better distribute the reactants/products and current density within PEMFCs because ZZFF enhances the flow direction transport between under-land and under-channel areas [181]. It has been reported that it is easier to realize the success of cold start by current ramping mode than by constant voltage or constant current strategies [182]. Hot air blowing, batteries, and catalytic furnaces can be used to achieve fast heating both internally and externally [178].

6.5. *Safety concerns*

According to market investigation and survey, potential consumers have doubts about the safety of hydrogen FCEVs [183,184] due to the flammable and explosive characteristics of compressed hydrogen. Once leakage or severe collision occurs, it must be ensured that they will not experience violent combustion and even explosion [185]. To improve the safety of FCEVs, automobile manufacturers have designed and installed the hydrogen tank and FC stack in the safer part of the vehicle. Taking the Toyota Mirai as an example, the FC stack and hydrogen storage tank are situated on the bottom of the seat and trunk, as shown in Fig. 18 and in case of an accident, the FC system and hydrogen storage tank can be protected as much as possible. In addition, automobile manufacturers have carried out a series of safety tests before introducing FCEVs to the market. However, these theoretical data are not convincing enough for consumers due to the small market share of FCEVs and lack of actual support. Therefore, commercialization of FCEVs is still limited.

6.6. *Immature EMS*

The goal of the EMS is to control multiple power sources. In the hybrid system, controlling the flow of power is vital to the market demand [186]. The EMS ensures that energy is distributed efficiently among the FC, auxiliary power units and electric motor in order to avoid FC power irregularities and extend the life of the FC and auxiliary power units. Owing to the proper integration of systems in the vehicle, one of the challenges is to optimize the setup and design of the appropriate controller [187]. The goal of EMS is to reduce hydrogen consumption and improve the coordination between multiple power sources [188]. During regenerative braking, maximum energy recycling is expected via auxiliary energy units such as battery and supercapacitor. Except simulation, experiments for EMS are also necessary [189]. Conditions such as the FC oxygen pressure, FC support during high loads, maximum energy recovery during regenerative braking, state of charge (SOC) and discharge depth of batteries as well as life of the battery and supercapacitor should be investigated [188]. The current EMS still needs significant improvement in a complex FC + battery + supercapacitor hybrid topology to balance the cost and performance. Additionally, the EMS has a direct impact on vehicle maintenance and operating costs [190,191].

7. Future outlook

In the context of global decarbonization after the Paris Agreement, popularization of FCEVs is an inevitable trend and historical development. The development route of FCEVs will be similar to that of petrol vehicles requiring continuous improvement and innovation. In fact, more than 100 years ago, petrol vehicles were introduced in spite of immature technology, insufficient safety, and difficulty in refueling. However, on the heels of continuous research and development, these obstacles have been overcome one by one and petrol vehicles are widely accepted by consumers. Nowadays, in the era of rapid development of science and technology, FCEVs will be faster than that of petrol vehicles to bring the technology to fruition in the hope of replacing petrol

Fig. 18. Schematic of FC and hydrogen storage tank in the Toyota Mirai.

vehicles. After decades of efforts, hydrogen FCEVs have passed the most difficult stage of technological development and entered the initial stage of market expansion and commercialization in 2015. According to the prediction of the International Energy Agency (IEA), the global market share of hydrogen FCEVs is expected to reach 2–3% in 2030 and about 15% in 2050 [192].

As a global leader in FCEVs, Japan's hydrogen social concept is worthy of global learning. The Basic Hydrogen Strategy released in 2017 lists the basic vision of Japan's hydrogen society, key areas of R&D and market development. Since then, this strategy has been incorporated into Japan's most authoritative energy policy—the National Basic Energy Strategy. It can be seen that the development of FCEVs has become Japan's national strategy, and its ambitions will not change in the next few decades [116]. Japan's efforts to popularize FCEVs are mainly focused on passenger cars in the mass market. This approach places FCEVs in direct competition with pure battery electric vehicles, rather than in niche applications where FCs have advantages in shorter charging times and longer mileage. Japan's choice is not without benefits. Targeting the mass market can minimize FCEV production costs through economies of scale, which will bring spillover effects to countries wishing to import Japanese FCEVs. However, Japan's current FCEV production and sales still rely on Toyota's Mirai, and its plan to achieve 40,000 on-road FCEVs by 2020 was ultimately not achieved. The FCEV production scale of Honda is limited, and the corresponding market sales are also not satisfactory. Honda announced that it would cease sales of its FCEV models in August 2021, which has aroused great repercussions from the international community. However, Honda's move does not mean abandoning its FCEV strategy. At present, Honda is pushing forward the reorganization of domestic and foreign factories by stopping the production and sales of unsalable models. It will still maintain cooperation with GM on the R&D of FCEVs and introduce new FC models in the future. It must be admitted that the global FCEV market will indeed be influenced. This situation will reduce Japan's ability to export a large number of FCEVs overseas, which in turn will hinder the promotion of FCEVs in regions such as California and Europe that rely on imported vehicles from Japan [116]. However, the stakeholders in Japan have made it clear that the suspension of Honda's production is just an episode in the development of hydrogen FCEVs and will not affect Japan's original intention and belief in the development of hydrogen FCEVs.

The fundamental research carried out by universities and institutes have mainly focused on catalyst development and materials science, which help to bring down the cost of FC systems [193,194]. It should be noted that the challenges of catalysts and materials are more research biased while some scaling-up engineering issues may be ignored. The inability to directly scale up laboratory or pilot-scale technologies to industrial production is a key obstacle to this novel energy conversion technology. Systematic integration is highly suggested to accelerate commercialization of new technologies and reasonable technology maturity evaluation, scaling-up simulation, system optimization, energy management, and life cycle assessment should be actively explored by researchers, investors, enterprises, and governments [195-198]. In order to obtain the optimization of electric powertrain systems and energy systems, the genetic algorithm-based simulations under the Worldwide Harmonized Light Vehicle Test Procedure (WLTP) driving cycle are recommended to be conducted in the design stage to make sure the simulation results can be representative to the real conditions [87,199].

In the face of the challenges mentioned in Section 6 for the commercialization of FCEVs, further fundamental and practical research should consider the following suggestions:

- Reducing the cost of FC stacks will be the most important means to improve the economic competitiveness of FCEVs. The high price of FCs is mainly contributed by the Pt catalytic layer. Therefore, reducing the loading of Pt, improving the utilization rate of Pt, and developing non-Pt catalysts are the focuses. By developing novel catalyst supports, such as carbon nanotubes, graphene, and titanium oxynitride, the dispersibility and utilization rate of Pt can be significantly improved. The development of Nafion alternative materials with simple preparation process and good chemical stability can lower the cost of PEMs. For bipolar plates, the thermal/electrical conductivity, corrosion resistance, mechanical properties and machinability should be improved at the lowest possible cost.
- FCs should be further modularized. Through the assembly of modules, the vehicles' requirements for different FC power levels can be easily realized, thereby reducing vehicle production costs. The power system should also be further hybridized. The hybrid of FC and battery systems can effectively extend the life of the FC, reduce the cost of the electric vehicle, realize the recovery of braking energy, and improve the cold start performance.
- The hydrogen infrastructure construction should focus on the four links of production, storage, transportation and distribution. The "blue hydrogen", which is transformed from natural gas and industrial by-products, has become the first choice due to its low energy intensity and high technological maturity. However, its purification cost needs to be further reduced. The "green hydrogen" produced by water splitting driven by renewable energy and nuclear energy is the most promising hydrogen supply in the future. There are many ways for hydrogen storage and transportation, such as high-pressure hydrogen cylinder storage and transportation, metal hydride storage and transportation, vehicle transportation, and pipeline transportation. A reasonable hydrogen storage and transportation method should consider both economy and energy consumption. Governments of various countries should gradually increase investment in hydrogen infrastructures, accelerate the deployment and construction of hydrogen refilling stations, and promote the formation of a hydrogen supply system for FCEVs.
- In order to improve the durability of FCEVs, in terms of material design, high-stability electrocatalysts resistant to impurities such as CO, corrosion-resistant catalyst carriers, and corrosion-resistant high-conductivity bipolar plates should be further developed. From the aspect of control strategy, appropriate start-stop strategy and operation control strategy should be designed according to the FC degradation laws. In addition, in order to make life prediction more accurate, it is also necessary to understand the attenuation

mechanism and develop failure simulation methods and life prediction methods.

- By optimizing key components, cold start strategies and auxiliary means, the low-temperature performance of FCEVs can be improved. Improving the water absorption and conductivity below the freezing point is the optimization direction of the PEM. Optimizing the content and thickness of the ionomer, the thickness of the gas diffusion layer, and the internal flow field and geometric design of the FC can accelerate the ice melting during the cold start process. The optimized current, voltage, and power control strategies also make significant contribution to the cold start performance. Other auxiliary methods include gas purging and rapid heating.
- The material, size, and location of hydrogen storage tanks should be optimized, and hydrogen usage specifications for FCs should be established. More laboratory crash tests should be carried out and more actual driving data should be collected to ensure the safety of FCEVs.
- Developing advanced EMSs and optimizing algorithms to improve fuel efficiency and realize the management and distribution of power sources such as FC, battery, and supercapacitor.

8. Conclusion

FCEVs are helpful in solving the environmental and energy crisis. This review summarizes the recent development of FCs in the automobile industry and current bottlenecks that hamper commercialization of FCEVs. The energy issue and environmental pollution are discussed from the perspective of the transportation industry and it is crucial to substitute fossil fuels with green energy sources such as hydrogen. The development and state-of-the-art of FCs such as PEMFCs as well as degradation mechanisms of PEMFCs are discussed. The PEMFC is the mainstream and most mature FC technology for vehicles. FC-battery hybrid systems and safety standards of FCEVs are introduced. Hybridization with an auxiliary battery system will greatly boost the dynamic response of FCEVs. The automobile FC market is analyzed and the global automotive FC share is expected to grow at a high speed for passenger vehicles and buses. Policies and subsidies for hydrogen FCEVs in the US, Japan, South Korea, the EU, and China are interpreted. Development of different types of FCs and application examples of FCEVs are summarized, indicating that hydrogen FCEVs have entered the preliminary commercialization stage. However, further adoption of FCEVs by consumers continues to face challenges on account of the high cost of FC stack production and maintenance, insufficient hydrogen supply facilities, insufficient reliability, slow cold start, safety concerns, and immature EMS. In response to these challenges, some important suggestions are provided and future development of FCEVs requires joint efforts by academia, industry and governments. It is projected that FCEVs will undergo rapid development and be well accepted by customers after these shortcomings are addressed.

CRediT authorship contribution statement

Yang Luo: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing. **Yinghong Wu:** Formal analysis, Investigation, Validation. **Bo Li:** Data curation. **Tiande Mo:** Project administration, Funding acquisition. **Yu Li:** Resources. **Shien-Ping Feng:** Project administration, Funding acquisition. **Jingkui Qu:** Supervision. **Paul K. Chu:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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Supplementary materials

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