Chapter

Hydrogen Fuel Cell Implementation for the Transportation Sector

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Abstract

Global transportation possesses have compelling rationales for reducing the consumption of oil, emissions of carbon dioxide, and noise pollution. Transitions to alternative transportation technologies such as electric vehicles (EVs) have gained increased attention from the automotive industries. A fuel cell electric vehicle (FCEV) occupying a hydrogen engine is one of the most stupendous technologies, since it is suitable for a large-scale transportation. However, its performance limitations are in question due to voltage degradation in long term operations through steady conditions under constant load and dynamic working conditions. Other drawbacks of using fuel cells in EVs are energy balances and management issues necessary for vehicle power and energy requirements. An efficient solution to accommodate driving behavior like dynamic loads comprises of hybridizing PEMFCs with energy storage devices like supercapacitors and batteries. This opening chapter reviews the projected gist of FCEV status; considers the factors that are going to affect how FCEVs could enter commercialization, including the importance of fuel cells for EV technologies; the degradation diagnoses using accelerated stress test (AST) procedures; FCEV hybridization; and the contribution of an energy storage device for charging EVs. The article also addresses case studies relating to material degradation occurring from driving behavior. Information about material degradation can be compiled into a database for the improvement of cell component performance and durability, leading to the creation of new materials and new fuel cell hybridization designs. To support the growth of EV technologies, an energy storage is required for the integrated alternative electricity generations. A redox flow battery is considered as a promising candidate in terms of attractive charging station for EVs or HEVs.

Keywords: electric vehicle, fuel cell technology, energy storage, fuel cell hybridization, accelerated stress test, flow battery technology

1. Introduction

1.1 New transportation interface

Leading automotive and energy executives from industries all around the world are looking at implementing strategies for promoting the use of electric vehicles and helping reduce oil consumption and climate-related emissions by trying to become an industrial leadership position in advanced electric-drive and hybrid vehicle technologies. The environmental advantages of electric vehicles (EVs) compared to conventional internal combustion engine vehicle (ICEV) are overwhelming for the environment. The life cycle of a combustion engine relies heavily upon fossil fuels to generate the electricity it runs on, therefore emitting dangerous and harmful emissions during manufacturing. In comparison to ICEV, EVs have two superior technical features: better upstream energy source flexibility and higher vehicle efficiency [1]. The lower efficiency of ICEVs is due to friction losses, fuel pumping losses, transmission losses, and thermodynamic energy losses in the vehicle configuration. EVs should obtain highly efficient electric powertrains to evade these losses. In the scenario of utilizing alternative fuels, such as biofuels to reduce environmental problems, many countries may face supply limitations. EV deployment has been steadily rising over the past ten years, with the global stock of electric passenger cars achieving 5 million in 2018, an increase of 63% from 2017 [2]. According to the Global EV Outlook 2020, sales of EVs reached 2.1 million globally in 2019. The significant barriers for expected EV commercialization comprise of vehicle price, range, charge time, battery life uncertainty, electric generator durability, vehicle model availability, charging infrastructure, and awareness and understanding of the technology. EVs typically use one or more electric motors or traction motors for propulsion. They may be powered via a collector system by electricity from off-vehicle sources, or may be self-contained with an electric generator or energy storage device. Thus, they can be mainly categorized into two basic types of EVs all-electric vehicles (AEVs) and plug-in hybrid electric vehicles (PHEVs). If considered in details, EVs can be classified into five groups: Hybrid Electric Vehicle (HEV), Plug-in Hybrid Electric Vehicle (PHEV), Battery Electric Vehicle (BEV), Fuel Cell Electric Vehicle (FCEV), and Solar Electric Vehicle (SEV) [3].

1.2 Fuel cell electric vehicle

Among the different technologies of interest, the FCEVs containing hydrogen engines have been developed (**Figure 1**), mainly due to their high-power density, quick start-up and low operating temperatures. FCEVs use a fuel cell such as a proton exchange membrane fuel cell (PEMFC) or solid oxide fuel cells (SOFC) to convert the chemical energy in hydrogen and oxygen directly into electrical energy. Hydrogen is currently produced through many technologies either from non-fossil fuels or from fossil fuels. Examples of non-fossil fuel technologies are water electrolysis, thermolysis, thermochemical water splitting, and photonic process.

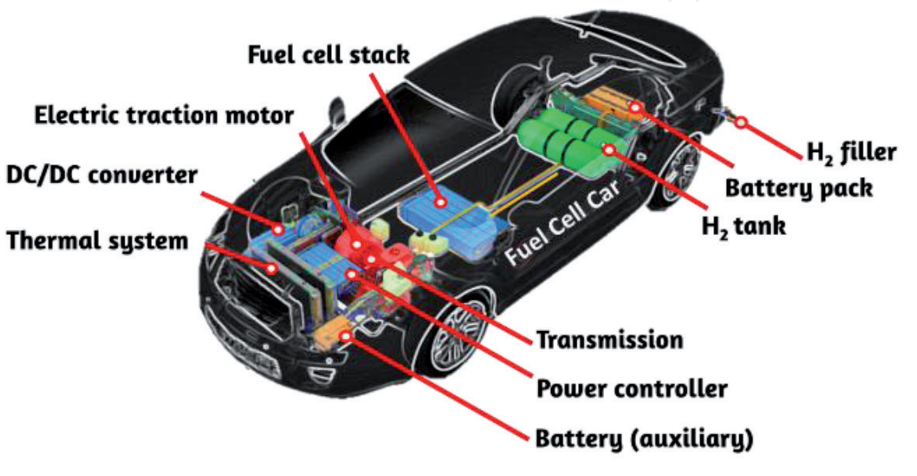

Figure 1.
Fuel cell electric vehicle configuration.

Fossil fuel technologies involve hydrocarbons reforming, these processes are carried out by methods such as steam reforming, auto-thermal, and partial oxidation [4]. FCEVs emphasized in this chapter are FCEVs that are using PEMFC as an electric generator. The dynamic response from driving behavior is one of the limitations for using PEMFC, especially if the system achieves a high load demand to acquire the desired speed ability. According to this situation the FC system would be unable to feed the fuel and oxidant in time, so "fuel and oxidant starvation" phenomenon would occur leading to materials degrading over time [5]. The auxiliary electrical source is one of solutions used to stabilize the electrical potential of the PEMFC system. Therefore, one part of this chapter discusses practical solutions to accommodate driving behavior via "Hybridization System". The missions of the hybridization system are relevant to the supply of traction power during PEMFC start-up, power assistance during driving cycles, regenerative braking energy recovery, the supply of electrical accessory loads, and PEMFC start-up and shutdown [6, 7]. An energy storage system such as a battery or a supercapacitor should be the preferred choice for the PEMFC hybridization system. A supercapacitor (SC) has interesting and effective functions such as its fast charge–discharge rate that can potentially support PEMFCs when they are operated under dynamic load demands [8]. Its fast responsiveness fulfills the power demand, rises the system power density and has to generate or absorb the power which either the PEMFC is not capable of generating or absorbing [9]. **Figure 2** indicates an example of PEMFC-SC hybridization. A supercapacitor offers transient power to PEMFC for attaining load demands in a short period. Moreover, the supercapacitor contributes advantages to PEMFC via capturing regenerative braking energy, enhancing fuel economy, providing a flexible operating strategy, overcoming PEMFC cold-start and transient shortfalls, and significantly lowering the cost per unit power [6].

Regarding transportation applications, the hybridization concept is not only applied for cars, but also for buses, trains, and tramways. The PEMFC-battery-supercapacitor hybridization was applied for electric trams [9] which possessed PEMFC for governing a stable operation. The battery offers a portion of the positive low frequency components of power demands which in turn decreases the responsibility of the PEMFC, and absorbs the slow-variation negative segments. The supercapacitor supplies the transient power demand effectively during sudden acceleration and braking.

The PEMFC performance investigations corresponding to driving behaviors will bring about information sustained to durability and lifetime of PEMFCs. Transportation applications require more than 5000 hours of PEMFC lifetime in order to be used under different circumstances [10]. This requirement results in

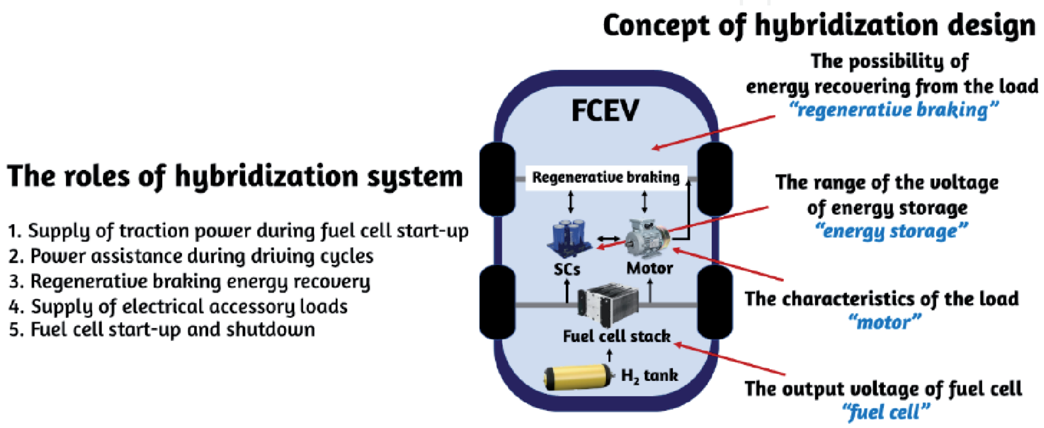

Figure 2.
Basic concept of PEMFC-SC hybridization.

time consuming and expensive methodology for experimental investigations. This chapter also provides information on the modification of Accelerated Stress Test (AST) created for exploring fuel cell degradation behavior and decreasing those restrictions [11].

Different driving behavior corresponds to various PEMFC operating conditions which can lead to numerous cases of PEMFC component degradation. For example, load cycling conditions such as voltage cycling, temperature cycling, and humidity cycling brings about membrane degradation; cracks, pinhole, and peroxide and hydrogen peroxide production [12]. These conditions also result in catalyst degradation such as Ostwald ripening of particles and sintering of particles [12]. If FCEV is driven at start-stop condition, PEMFCs will operate at high voltage, high temperature and low humidity [13, 14]. The detachment of catalyst particles from carbon support, Ostwald ripening of particles, sintering of particles, and dissolution of the catalyst can occur under start-stop circumstances [13, 14]. Evaluating and understanding material degradation and failure mechanisms through physical, chemical, and electrochemical analyses are important for material selection and fuel cell design. In terms of electrochemical analyses, electrochemical impedance spectroscopy (EIS) and cyclic voltammetry (CV) are preferable. EIS is applied for diagnosing behavior of PEMFC such as metal corrosion, electrode-electrolyte interface behavior, double-layer capacity, electrical properties of material and interfaces, and electrodeposition [15]. Morphological studies using Scanning Electron Microscopy (SEM) technique, Transmission Electron Microscopy (TEM), X-ray Diffraction (XRD) technique, and X-ray Photoelectron Spectroscopy (XPS) technique are proposed by scientists and engineers. This chapter presents case studies of material degradation occurring from driving behavior, since the information about the material degradation can be compiled into a database for the improvements of the cell component performance and durability, leading to the creation of new materials and new fuel cell hybridization designs.

1.3 EV charging station concept

Charging infrastructure and energy management are transforming in parallel with the growth in EV demand. The commercial success of the EV requires the development of an accessible charging infrastructure. Even though EVs do not produce the usual exhaust pipe emissions, main electricity utilized for EV charging systems is extensively generated from coal and natural gas that emits substantial CO_2 emissions. Even though electricity delivered for a charging system can be generated from renewable energy generators such as photovoltaic cells and wind turbines, these generators have weather-dependent issues. Weather and location play significant roles in how efficient a solution can be in regards to suppling generators. Integration of renewable energy sources, energy storage systems, and electrical vehicles with smart power distribution networks could be solution to this problem.

The type of battery mentioned in this article is a vanadium redox flow battery that captures energy generated from photovoltaic cells or wind turbines, this energy can be collect at one time and stored for use at a later time (**Figure 3**). Research works of literature focused on the abovementioned integrations include wind-PEM electrolyzer-hydrogen systems and solar-PEM electrolyzer-hydrogen systems [16]. In terms of FCEVs, hydrogen infrastructures are the most costly for many countries. Filling fuel of FCEV can be manipulated via charging stations and on-board PEMFC systems. This article would like to provide information of the stationary scenario. Hydrogen production using an integrated system between a redox flow battery and an electrolyzer is a remarkable new technology for FCEV fueling.

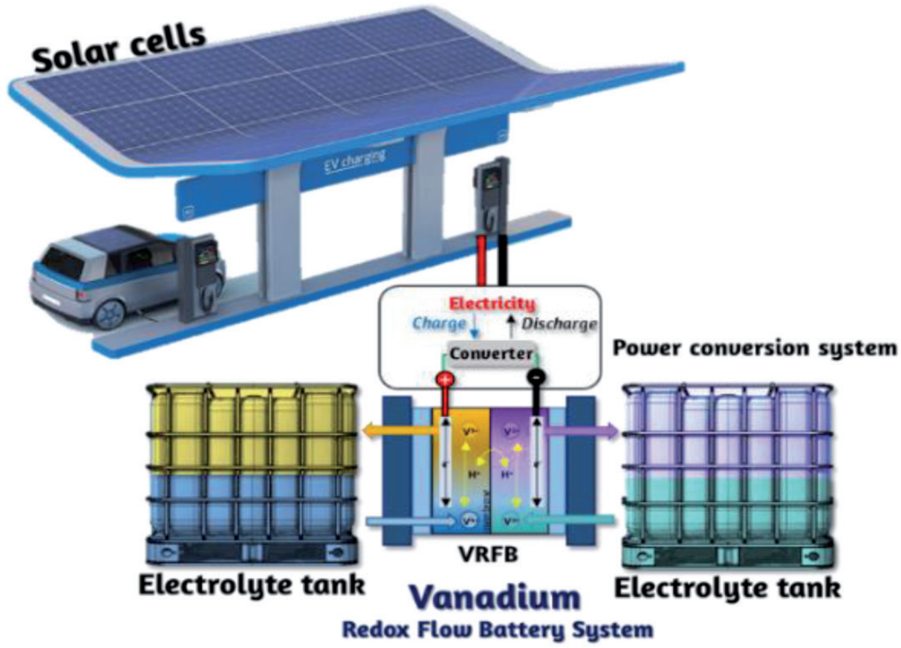

Figure 3.
Conceptual idea of using a redox flow battery for EV charging station.

It is possible to provide charging stations and hydrogen fueling stations in the same location. Prior to discussion about the contribution of an energy storage device for charging EVs, basic information of the redox flow battery is provided in the following statements. A redox flow battery technology is quite similar to PEMFC technology. The battery produces reduction and oxidation reactions between two active materials to capture and release energy. The redox flow battery system includes two external reservoirs for collecting soluble electroactive electrolytes, two electrodes, a membrane separator and a flow circulation system [17]. Flow batteries can be divided into three categories according to state of reactants such as all liquid phase, all solid phase, and hybrid redox flow batteries [17]. In a polymer electrolyte membrane electrolyzer is an electrolytic cell where water reacts at the anode to form oxygen and positively charge hydrogen ions. The applied electrons transfer through an external circuit, while the hydrogen ions selectively move across the PEM to the cathode. At the cathode side, hydrogen ions combine with electrons from the external circuit to form hydrogen gas [18]. Different types of electrolyzes consist of proton exchange membrane electrolyzer, alkaline electrolyzer, and solid oxide electrolyzer [18].

This chapter comprises of the following topics; the importance of fuel cells for EV technologies, the degradation diagnoses using accelerated stress test procedures, energy storage units integrated with fuel cells for the FCEV applications, and the contribution of an energy storage device for charging EVs. The authors expect to provide information related to hydrogen fuel cells for transportation prior to this technology becoming more frequent in daily life.

2. The importance of fuel cell for EV technologies

The EV market has witnessed rapid evolution with the ongoing developments in the transportation sector. The EVs market is projected to reach 35% of all globally vehicles by 2040 [19]. The factors impacting the market shares of this alternative advanced technology are retail price equivalent (RPE), energy cost per kilometer, range (kilometers between refuel/recharge events), maintenance cost (annual), fuel availability, range limitation for battery electric vehicles (BEVs), public recharging availability, risk aversion, and diversity of make and model options available [20]. The principal factors affecting EV technology development relate to reliability, durability, efficiency, voltage losses, the current generation, power generation, and energy generation [21]. Reliability is defined as the ability of a product to perform the required function under stated conditions for a certain period of time [22]. Durability is the lifetime within the repair rate and cost of planned repairs, overhaul and maintenance [22]. Efficiency refers to the ratio between the useful energy output, which is the electrical energy produced, and the variation and energy input, which is the enthalpy of hydrogen [23]. Voltage losses are described as the voltage drop between standard voltage and real voltage [24].

2.1 Types of electric vehicles

In this sector five sorts of electric vehicles; HEV, PHEV, BEV, FCEV, and SEV, are discussed as follows:

- hybrid electric vehicle (HEV) which combines an internal combustion engine system with a hybrid vehicle drivetrain.

- plug-in hybrid electric vehicle (PHEV) is an HEV whose battery is recharged by plugging it into an external electricity source, as well as by its on-board engine and generator.

- battery electric vehicles completely consume chemical energy stored in rechargeable battery packs, without a secondary source of drivetrain.

- solar electric vehicle (SEV) powered exclusively by direct solar energy.

- fuel cell electric vehicle (FCEV) uses a fuel cell, sometimes in combination with a small battery or supercapacitor, to power its on-board electric motor.

Powertrain configurations of EVs are illustrated in **Figure 4**, and it indicates that an energy storage device is necessary for specific functions, for instance, demand response, transmission, flexible generation, improve operational practices and providing high energy density [25, 26].

Even if electric vehicles have more than their share of advantages, it is worth noting that they still have their drawbacks. It is imperative to recognize that EVs are usually changing and their technologies are evolving if considering EVs' pros and cons. HEV has an advantage in component availability, but it takes higher initial

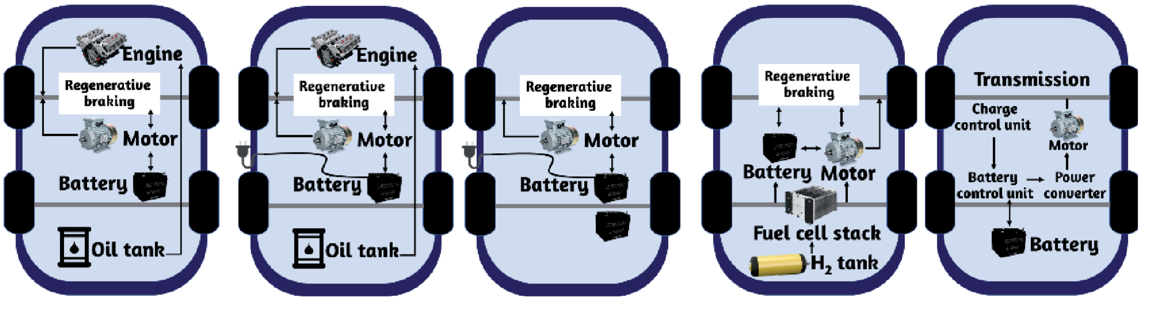

Figure 4.
The powertrain configurations of EVs.

cost. Furthermore, its two power trains build complexity of configurations and significant transmission energy loss. The characters of PHEV are similar to HEV, however, high cost of its batteries and battery replacement and added weight are taken into consideration. SEV does not have speed or power that regular cars have, and its operation is relevant to weather dependency. It seems likely that BEV is in the spotlight, nevertheless, there are some cons. It gives short distance range, while battery technology and public recharging infrastructure are needed to be improved these are the reasons that Toyota has long maintained that hydrogen fuel cell technology could be a zero-emission solution across a broad spectrum of vehicle types? Do FCEVs have a future?

Despite the development of hydrogen fuel cell cars started in 1966 with GM's Electrovan, they remain low in volume, expensive to produce, and restricted to sales in the few regions that have built hydrogen fueling stations. A big BEV manufacturer disagrees with this idea, and the CEO of the company described the fuel cell technology as a mind-bogglingly stupid technology. Why is Toyota still trying to make the fuel cell happen? Some scientists predicted that people would be able to drive FCEVs without any problems and to refuel 800 km of range in 2–3 minutes without any local emissions. BEVs maybe therefore only a temporary transitional technology.

Authors would like to provide readers with concise information that may help answer these questions. During PEMFC operations, hydrogen permeates through the anode via a bipolar plate and interacts with the catalyst for producing electrons and protons. The electrons are conducted via electrically conductive materials (catalyst, gas diffusion layers, bipolar plates, and current collectors) through an external circuit to the cathode, while the protons are simultaneously transferred via an ionic route through a polymer electrolyte membrane (Nafion membrane) to the cathode. At the cathode, oxygen permeates to the catalyst surface where it reacts with the protons and electrons with properly hydrated situations. Subsequently, the products of the fuel cell reactions are water, electricity and heat [27].

2.2 Filling fuel of FCEVs

FCEVs are charged using compressed hydrogen gas, and the hydrogen is drawn from an onboard tank and fuses it chemically with oxygen to create water. The BEV battery is recharged by connecting it to the electrical grid through a connector system. With 5 minutes for one tank filling, refuelling hydrogen is significantly faster than charging a BEV that is around 3 hours [28]. Additional 5 to 10 minutes are spent for a hydrogen pump to be ready for refuelling after a few refuelling operations until the fueling pressure is built up again. People have to drive FCEVs to a hydrogen gas station for refuelling, while BEV can be either charged at home or a station. As known. Hydrogen fuel stations are rare since construction costs of hydrogen stations are expensive. The costs are expected to be decreased via large-scale deployment and standardization. Moreover, a centralized control center for the hydrogen station is envisioned, accordingly dropping the operating costs.

Hydrogen production principally composes of 2 approaches; stream reforming and electrolysis [29]. Steam reforming is currently one of the most pervasive processes for hydrogen production. This technique gains advantages from high-efficiency production and low operational and production costs. Reactants used for the process are natural gas and lighter hydrocarbons, methanol, and other oxygenated hydrocarbons [30]. Electrolysis is a promising option for hydrogen production from a renewable resource such as water. The process uses electricity to split water into hydrogen and oxygen [30]. For charging the battery and fueling hydrogen via electrolytic cell, both come with energy and efficiency losses. In the case of BEV, an electrical grid provides AC currents, while the batteries discharge DC currents. Quick charging efficiency is around 92%. If BEV runs with an AC motor, the inverter efficiency would be 90%. Also, a lithium-ion battery can lose energy due to current leakage, so a good estimate for charging a lithium-ion battery is about 90%. All these factors combined lead to 75–80% of total efficiency for charging BEV. In the case of FCEV, rectifier requires AC current from the electrical grid to drive the electrolysis, thus conversion efficiency would be about 92%. We also need to convert DC produced from fuel cell to power the AC motor with inverter efficiency 90%. Finally, the efficiency of the motor must be considered for both fuel cell and battery, currently around 90–95%. However, storage hydrogen into cylinder and transportation must be included for fuel cell efficiency losses. Once the hydrogen is produced and compressed into liquid or gas, available hydrogen infrastructure requires as hydrogen be able to be delivered from where it is produced to the point of end-use. Since hydrogen is exceedingly low density as a gas and liquid, to achieve satisfactory energy density, actual density must be increased. There are two choices, compressing and liquifying hydrogen, for increasing the density. Hydrogen can be compressed to 790 times atm pressure, but that takes energy nearby 13% of the total energy content of the hydrogen [29]. Hydrogen can be turned into liquid cryogenically. Hydrogen is liquified by decreasing its temperature to −253°C with 40% of an efficiency loss. The benefit of hydrogen liquefaction is that a cryogenic hydrogen cylinder is much lighter than a cylinder holding pressurized hydrogen. In conclude, pressurization is the better option for efficiency losses. Focusing on hydrogen transportation, hydrogen is being transported by truck or pipeline, where know energy loss from 10% up to 40%. In the worst case, the total efficiency of FCEV may be approximately 20%, while the BEV efficiency could be 56% [29]. Another weakness of FCEVs is the price per kg of hydrogen. The FCEV named Honda Clarity gets about 589 km with 5.5 kg of hydrogen, so that would cost about $0.14/km. In contrast, Tesla Model 3 (BEV) employs $0.03/km or $0.20/ kWh of energy. Noted, the information relates to the US hydrogen price in 2018 that was $15/kg.

2.3 The interesting things about FCEVs

The above data imply that FCEV is inferior to BEV, but FCEV might have its place. FCEV has key benefits; charging duration, electric range, energy density, and vehicle weight. In terms of electric range, FCEV seems to come out on top of BEV as same as charging duration. The electric range is the driving range of a vehicle using only power from its electric electricity supply to traverse a given driving cycle. In the case of an EVs, it means the total range per charge [31]. Current FCEVs have the electric range from 312 to 380 miles, whereas most of BEVs possess the range under 259 miles. From this benefit, 78% of automotive executive believe that FCEVs will be breakthrough for electric mobility. Increasing electric range requires a lot of batteries that will add the weight of the vehicles. At a certain point (350 miles of range) the additional battery weight no longer yields the additional range. For FCEVs, this battery weight compounding is not an issue, because they can be refueled less than 5 minutes. This case will occur when hydrogen fueling has good availability. Energy density is another important benefit of FCEVs. Hydrogen gives the energy density 39 kWh/kg, in contrast with batteries of 75 kWh extended range Tesla Model 3 providing the energy density around 0.2 kWh/kg [32]. Gasoline stocks up 13 kWh/kg. Adding an electric range of FCEVs can be done by simply increasing size if the hydrogen tank. For BEVs, that would mean an additional 100 kg of weight and $1000 in cost. In terms of specific energy of the FCEVs using compressed hydrogen, the specific energy is near 40,000 Wh/kg that differs from lithium-ion batteries having just 278 Wh/kg of specific energy. BEVs are suitable for personal transportation, but current batteries can never replace gasoline applications as trucks, boats, airplanes, and trains. Considering energy density and specific energy, hydrogen very well could. In other words, FCEV is a potential solution for large scale transportation, and there are currently some interesting innovations and fuel cell products on the market worth taking [33]. For examples: Toshiba Energy Systems & Solutions Corporation announced that it has signed an agreement with New Energy and the Industrial Technology Development Organization (NEDO) for a multi-utility pure hydrogen fuel cell module for large modes of transport; the hydrogen train arrives in the Netherlands; hydrogen food retailer running in Norway; hydrogen truck in Japan [34]; Alstom (French manufacturer) plans to deliver 27 hydrogen fuel cell trains to subsidiary Fahma of regional public transport provider RMV in the central German state of Hesse by 2022, creating the world's largest fuel cell train fleet in passenger transport. Hydrogen fuel providers have invested in the expansion of hydrogen fuel production and distribution facilities to serve developing FCEV market, for instance, Germany aims to open hydrogen refueling stations by 2020, France is planning on opening renewable hydrogen station by 2020, and the UK wants to establish 100 hydrogen station by 2025 [34]. Is there adequate lithium to support the growth of BEV commercialization? BEVs and stationary storage revolutions are existing demand shooting up. If these revolutions occur, a hundred Gigafactory scenario may come true. The 13.5 million tons of reserves may be less than a 17-year supply.

As aforementioned, FCEVs should be developed for supporting an interface between the transport and the energy system. Therefore, basic knowledge and basic information of materials and applications of FCEVs should be comprehended. The U.S. Geological Survey produced a reserve estimate of lithium in early 2015, concluding that the world has enough known reserves for about 365 years of current global production of about 37,000 tons per year.

3. Degradation Diagnoses using Accelerated Stress Test (AST) Procedures

To analyze degradation data of the PEMFC components and system, the traditional observation of durability information involves the degradation data as a function of time. PEMFC typically show a continuing degradation in power output during their operations, and the cumulative influence of the gradual degradation is still acceptable. The degradation will be improper if the cumulative impacts of continuing degradation become too high. Regarding the characterizations, the modifications of accelerated stress test (AST) protocols have been created for diagnosing the PEMFC degradation behaviour to decrease those restrictions. ASTs are regularly created depending on a specific application, since the performed PEMFC degradations associate to the different cell components, the origins of the stresses, and their influences. Elevated temperature, reduced humidity, open-circuit voltage, and cycling conditions are the foremost accelerated factors ordinarily used for accelerated life diagnosing. The dynamic conditions consist of relative humidity (RH), temperature, potential, freeze/thaw, or start/stop [35]. The crucial conditions for transportation requirements are dynamic load cycling, startup-shutdown, and freeze–thaw [36].

The protocols were created to represent working behaviour in each application. The protocol can be divided into 2 sub-protocols such as static driving cycle and dynamic driving cycle. The Department of Energy (DOE) created the first protocol for the 2000-hour test. After that, the New European Drive Cycle (NEDC) was created, and it was created under the assumption of the European driving behaviour at the maximum speed as 50 km/h (20 A) [37, 38]. In our previous work [39] driving protocol which is the combination of load cycling and start-stop behaviors was designed as presented in **Figure 5**. The proposed load cycling approach intended to accelerate the influences caused by the real operating conditions correlated to a generic dynamic load. The primitive concept was to design the AST protocol right on the real working conditions, assuming the load profile as the major degradation source. Thus, working conditions were accelerated stressing the real load cycles magnitude and frequency. To assure the similar degradation mechanisms the progress of the real load value was evaluated and kept during the accelerated cycle. The methodology allowed accomplishing the AST profile steady with the actual load dynamics, but amplified in magnitude and scaled in the time domain. Finally, the scaled cycles were repeated in a loop [39]. The profile of start-stop cyclic represented starting and shutting down a vehicle in a short time. At the starting situation, electrochemical reactions were speedily fed into the fuel cell to generate the desired power. On the other hand, the reactions were terminated by stopping the reactant supply.

This dynamic behaviour severely involved the operating condition changing that would cause material degradation. This created protocol presented an overview of ordinary approaches adopted in hybrid FCEV for power management, being the starting point for the load profile set-up for PEMFC operations [39].

The results of voltage degradation indicated that the voltage drop produced by load cycling gradually increased that corresponded to slowly operating condition differentiation. In contrast, the voltage degradation rate was significantly increased. This occurrence corresponded to the voltage lost in driving the chemical reaction at both on anode and cathode. The reduction of oxygen is a much slower reaction than the oxidation of hydrogen, therefore the system requires higher activation polarization, anode side losses can be neglected. This ageing regarded to damaging electrochemical surface areas of catalysts. The resistance to an electron flow through the electrically conductive PEMFC components and to an ion flow through the membrane caused a voltage drop as well. This loss generally happens in main components; bipolar plates, gas diffusion layers, catalyst, and membrane. The consumption of reactant gases at the catalyst layers leads to concentration

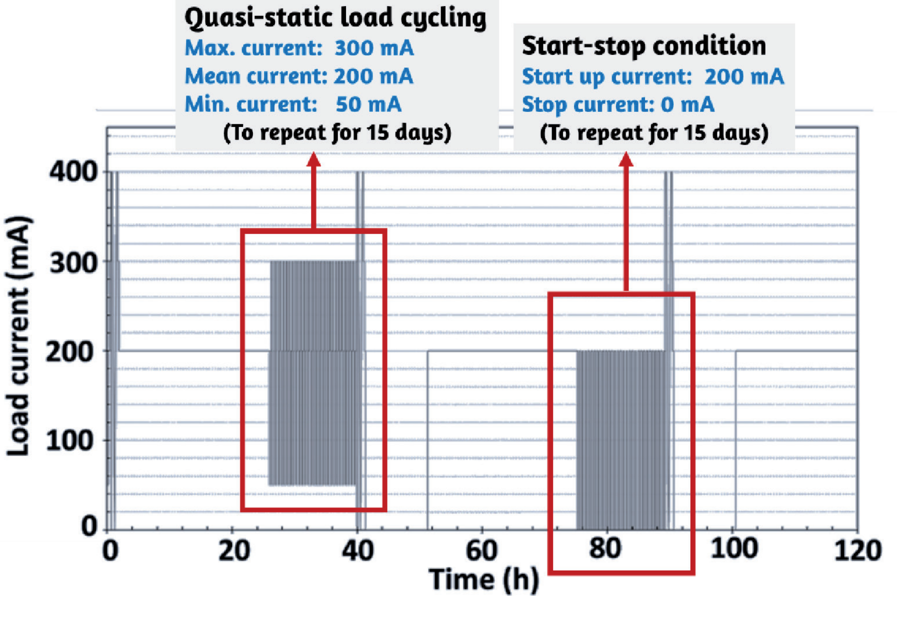

Figure 5.
The driving behaviors protocol [39].

gradients and the partial pressure of the reactants changing, affecting a decrease in fuel cell voltage [40]. **Figure 6** illustrates the case study of sudden load variation behaviour when the car speed varies from a low speed like driving to suddenly adding acceleration and overtaking cars in front. At high load demand, the system requires high power and voltage leading to high energy to drive electrochemical reactions thermodynamically. Under this situation system temperature increases significantly, while relative humidity decreases. This operating condition may result in membrane degradation; chemical and/or mechanical degradation [41]. In terms of chemical degradation, radicals such as peroxide or/and hydrogen peroxide radically react with the backbone of the membrane (Nafion: polytetrafluoroethylene). On the other hand, this reaction cannot occur if the backbone is not fluorinated [42–44].

The following mechanism, presented as Eqs. 1–3 [45], illustrates membrane damage where hydrofluoric acid was produced. **Figure 7** shows the morphological feature of a membrane electrode assembly (MEA) observed by scanning electron microscope and energy-dispersive X-ray (SEM–EDX) after 888 hours of operation duration. The investigated results found that the gas diffusion layer was dissolved by fluorine leaching, and the gas diffusion layer lost weight around 30.95%wt [46]. Small spots in **Figure 8** shows the catalyst removal from catalyst supports.

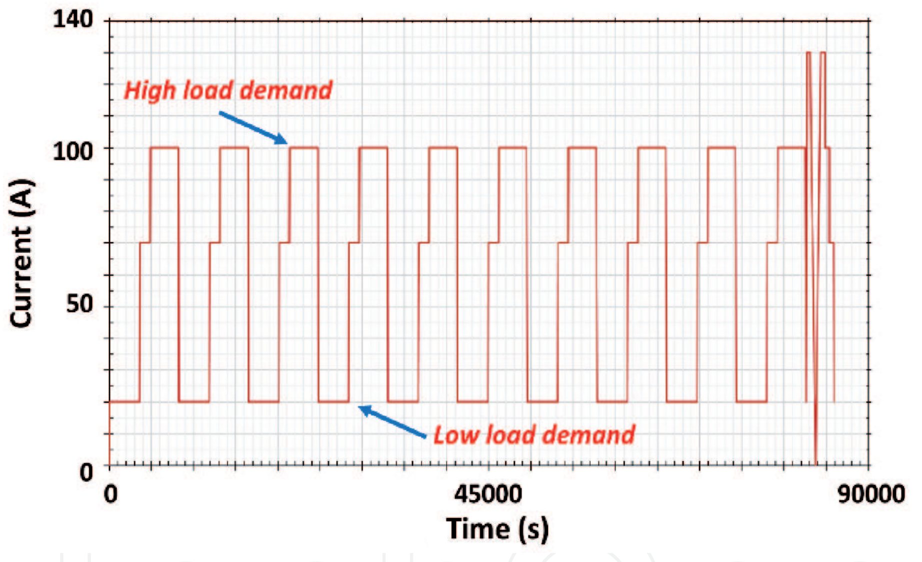

Figure 6.
The cyclic profile represents sudden load variation.

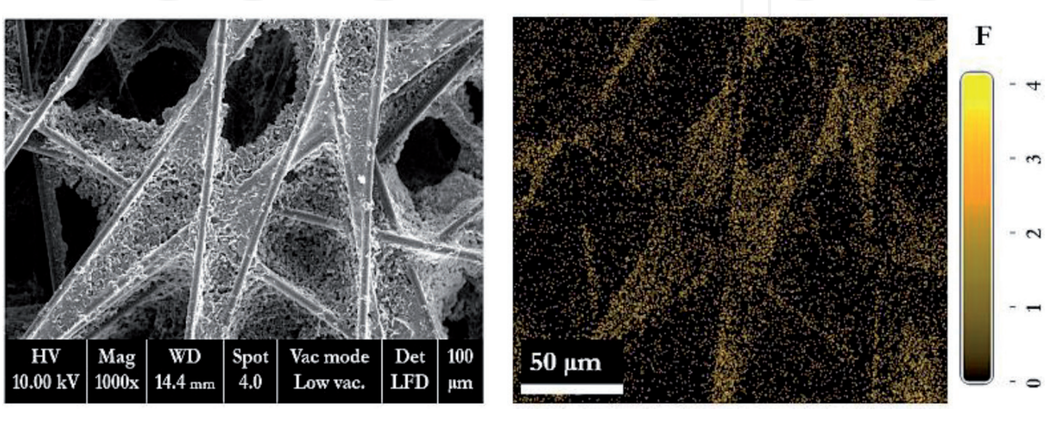

Figure 7.
SEM–EDX micrographs of gas diffusion layer damaged by hydrofluoric acid leaching.

Figure 8.
SEM–EDX micrograph of the catalyst layer on the PEMFC membrane.

Chemical degradation of the membrane [45]

$$R-CF_2COOH_2+HO^* \rightarrow R-CF_2+CO_2+H_2O \quad (1)$$

$$R-CF_2^*+HO^* \rightarrow R-CF_2OH \rightarrow R-COF+HF \quad (2)$$

$$R-COFH+H_2O \rightarrow R-COOH+HF \quad (3)$$

The relative humidity cycling related to load cyclic profile generated membrane swelling and shrinking. This phenomenon is associated with hydration state and operating temperature called mechanical degradation [27]. The postmortem analyses from the humidity cycling tests suggest that in-plane tensile membrane stresses result in cracks to initiate and propagate within the membranes in a subcritical fashion. Cyclic mechanical stresses cause if hydrophilic membranes are exposed to fluctuating hygrothermal conditions during the PEMFC operation [47]. Moreover, these phenomena may affect to catalyst degradation such as Ostwald ripening or sintering of catalyst particles. The Ostwald ripening appears from the thermodynamic driving force, while the sintering is caused by the reduction in surface energy with particle growth. The phenomenon leads to the dissolution of smaller particles and the growth of larger particles related to Eq. 4–6. In PEMFC, the platinum (Pt) transformation via a coupled process involving the transport of ions (Pt^{2+} and/or Pt^{4+}) through an ionomer/aqueous medium and a parallel (coupled) transport of electrons through the carbon support [48].

Pt dissolution reactions [49]

$$Pt \rightarrow Pt^{2+}+2e- \quad (1.188\ \mathrm{V\ vs.\ RHE}) \quad (4)$$

$$Pt+H_2O \rightarrow PtO+2H^++2e^- \quad (0.980\ \mathrm{V\ vs.\ RHE}) \quad (5)$$

$$PtO+2H^+ \rightarrow Pt^{2+}+H_2O+2e^- \quad (0.208\ \mathrm{V\ vs.\ RHE}) \quad (6)$$

The catalyst degradation can be diagnosed via several techniques; electrochemical impedance spectroscopy (EIS) technique, cyclic voltammetry (CV) technique, SEM–EDX technique, and X-ray photoelectron spectroscopy (XPS). The investigated results from EIS explain the impact of ionic resistance, activation loss (relate to the loss of the electrochemical surface area), and mass transport losses [50]. The CV can be used to diagnose the evaluation of catalyst activity. With is analyzing, the counter and reference electrodes generally act as an anode side because the kinetics of the oxidation reaction is relatively fast. Working electrode acts as a cathode side because the reduction reaction is a rate-determining step [51]. X-ray photoelectron spectroscopy (XPS) is a quantitative spectroscopic technique applied to characterize the surface chemistry of materials or to measure the elemental composition, empirical formula, chemical state, and electronic state of the elements existing in materials [51].

At low load demand in the sudden load variation, the system requires low voltage impacting slow reaction, so the system temperature decreases related to an increase in relative humidity. This operating condition causes a flooding phenomenon that can be studied via a hysteresis loop. The different voltage values from upward current testing and downward current testing in the hysteresis loop indicate accumulated water inside the PEMFC. The water flooding effects catalyst oxidation, reactant starvation, electro-osmosis, and back diffusion [52].

4. Why should energy storage unit be integrated with fuel cell for the FCEV application?

Although there are a lot of merits, PEMFCs are still not close to perfect. The major concern is relevant to performance and lifetime of the fuel cell. The performance of PEMFC is influenced by many internal and external factors, for instance fuel cell design and assembly, degradation of materials, operational conditions, and impurities [53]. Hybridization is one of solutions to reduce the problems. The hybridization in the energy sector can be categorized into four systems as follows:

- Hybridization between alternative energy and backup power unit, which is usually integrated with a high level to provide local energy security, is created to provide the intermittent availability of alternative energy sources.

- Hybridization between alternative primary sources, two or more alternative primary sources, is used to deliver complementary advantages.

- Hybridization between alternative energy and energy storage systems is the combination of energy storage with alternative energy. It is applied to ensure reliability and security of the distributed power generation system, while maximizing its benefit using alternative energy.

- Hybridization constructed with various types of energy storage devices is utilized for fast-dynamic storage devices and long-term storage devices [54].

In terms of transportation applications, PEMFC material suffers from closely random power load cycling such as the frequent start-up and shut-down [55]. An interesting approach to improve the efficiency and increase the lifetime of the PEMFC is to incorporate the new emerged energy storage, named Supercapacitor, in the system. A system that include several sources and/or energy storage devices is also known as hybrid system.

The concept of fuel cell-supercapacitor hybridization is quite new compared to the fuel cell-battery hybridization. The advantages of supercapacitors over the batteries are the higher number of charge/discharge cycles and the higher current rating. Once the supercapacitor is connected to the system, the stress due to the transient current were handled by the supercapacitor. There are many topologies of fuel cell-supercapacitor hybridization but most of them are connected together via various types of DC/DC converter. The very new concept of fuel cell/supercapacitor hybridization is to connect them directly together, this method is call direct-hybridization [56–59]. This concept is very interesting because it is able to increase efficiency, lifetime and reduce the system cost due to the absence of the DC/DC converter, which has significant impact on system design. **Table 1** presents the crucial characteristics of a supercapacitor applied to the energy storage hybridization. The supercapacitor generates higher power than a battery does, and either charging or discharging time is faster than the ability of a battery. The fast charging and discharging characters can diminish materials degradation and PEMFC lifespan. Also, a supercapacitor can offer transient power to meet load demand in a short time. According to this advantage feature chemical kinetic energy can be recovered during regenerative braking occurring while the automobile is slowing down or stopping. The supercapacitor can also save energy and protect materials components inside PEMFCs from deterioration [60].

Supercapacitors in a FCEV operate with two features; charging electricity by PEMFC and discharging electricity to PEMFC. Once electrical current is charged into a supercapacitor, the positive charges of electrolyte move to a negative electrode using electrostatic force, and the negative charges of electrolyte transfers to the positive electrode. Supercapacitors are normally composed of three main structures; electrolytes, electrodes, and separators. Typically, the electrode of the supercapacitor is made from carbon particles due to they have high surface areas and high porosity required for collecting the charge. During charging duration [61], the charges transfer through the pores, and then they are stacked layer by layer as shown in **Figure 9**. On the other hand, a discharging process is an inversion operation of the charging step. Supercapacitors discharge electricity to load based on charge volume and voltage change over time leading to speedy response to load.

The powertrain configuration of the FCEV is usually comprised of supercapacitors connected with PEMFC. The supercapacitor and convertor can be directly connected with PEMFC. The supercapacitor can also be connected in parallel with PEMFC through energy converters. A converter is an electromechanical device

Characteristics	**Supercapacitors**	**Batteries**
Specific energy (Wh/kg)	1–10	100–265
Specific energy is defined as energy per unit mass. It is used to quantify.		
Specific power (W/kg)	500–10,000	300–1500
Specific power is a measure of performance for the system. It is defined as the power output by it divided by its mass.		
Cell voltage (V)	1.2–3.3	2.5–4.2
A voltage or electromotive force, is a quantitative expression of the potential difference in charge between two points in an electrical field.		
Capacitance (F)	0.1–12,000	—
Capacitance is the ability to store electrical energy.		
Discharge time	s-min	min-h
Discharge time is time to release electrical energy from device by a discharge.		
Charge time	s-min	min-h
Charging time is time to accumulate electrical energy from device by a charge.		
Charge/discharge efficiency (%)	85–98	90
Charge/discharge efficiency is indications of how much energy is put into or drained from a cell relative to the amount of energy that is dissipated within the cell as a result of its internal impedance.		

Table 1.
Characteristics of battery and supercapacitor for hybridization.

transforming a source of direct current (DC) from one voltage level to another. The energy converter functions as an energy collector storing generated energy. If the system quickly requires energy, the generated energy will be supplied by the converter. This connection feature makes the system more complex and expensive. A directly connected structure that a supercapacitor directly connects in parallel with PEMFC plays a role in self-energy management, therefore, the system requires an energy management design. This scenario can directly protect against rapid power variations that can increase the dynamics of the hybridization system. It is worth noting that a directly connected structure between PEMFC and supercapacitor, a supercapacitor is a necessity to be pre-charged before utilization to limit inrush current [62]. A hybrid system requires PEMFC as the main power source and supercapacitors as an auxiliary energy source. The supercapacitors assist the system to reduce voltage fluctuations at an unstable demand. The supercapacitor also stores electrical energy from PEMFC when there is excess energy. In contrast to this, the supercapacitor will supply power to PEMFC once the load demand is high. This system acquires less equipment, less sophistication, and provides higher effectiveness [63].

The noticeable data from investigation of supercapacitor effect on PEMFC-supercapacitor direct hybridization performance related to a driving behavior

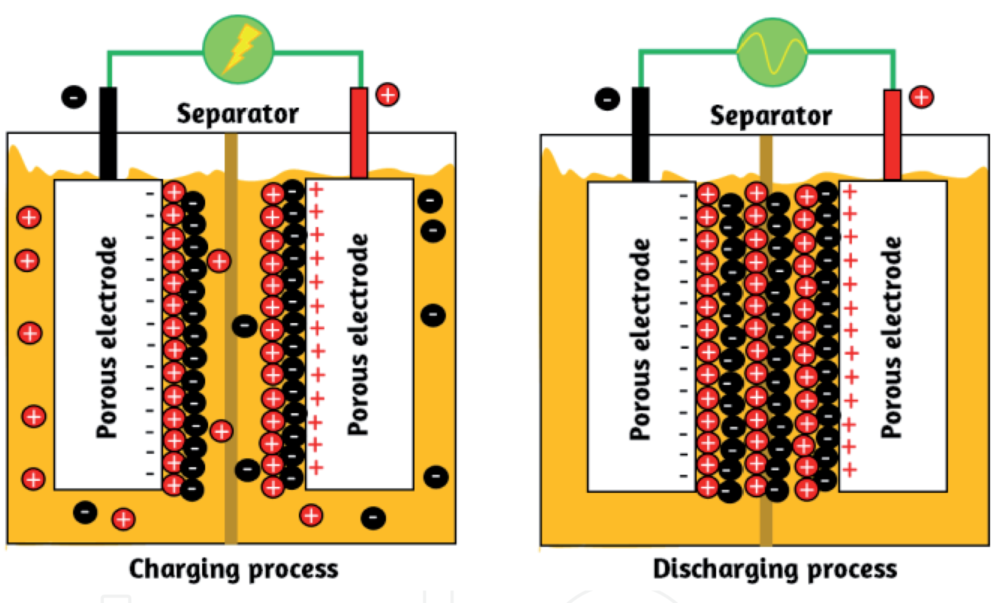

Figure 9.
The operations of a supercapacitor.

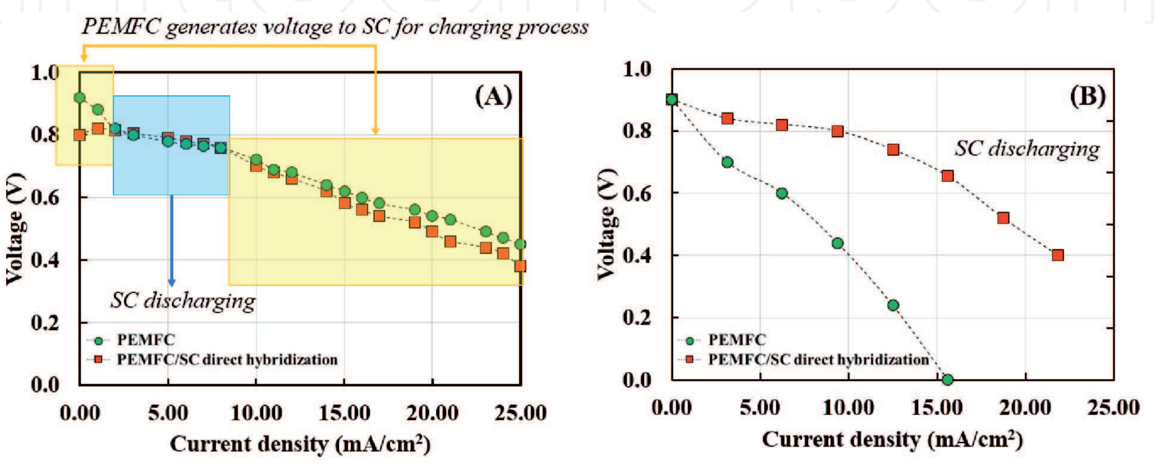

Figure 10.
The polarization curves of PEMFC and PEMFC-SC direct hybridization (A) non-charging supercapacitor (B) pre-charging supercapacitor at 0.90 V.

protocol are shown **Figure 10** [64]. Throughout the testing period, the PEMFC generates electricity to load demand and supercapacitor for charging process until the voltage level between PEMFC and supercapacitor are equivalent.

The curves in **Figure 10** can be separated into three transitions. In the first step of the test using a non-charging supercapacitor, the PEMFC charge electricity to load as indicated in low current density range (yellow area) until the electrical power of the PEMFC and supercapacitor is in the same level. Both of electrical providers supply electrical power to load demand in the second step. Due to supercapacitor properties, fast charging and discharging, the voltage of supercapacitor dramatically decreases observed in the third step. At this situation the voltage level of supercapacitor is lower than the one belongs to the PEMFC, thus, the PEMFC charges electricity to charge supercapacitor again. In pre-charging point of view, the voltage of PEMFC-SC direct hybridization is higher than PEMFC in all transition. It implies that the PEMFC and supercapacitor jointly supply electrical power to load demand. The supercapacitor assists the system to reduce voltage loss at high current density. The major voltage loss occurs from the mass transport [65].

5. The contribution of an energy storage device for charging EVs

It is estimated that over the next 15 years there will be more than a million electric vehicles in a small country as Thailand [66]. To reach this target both government and business sectors must provide infrastructure and technologies corresponding to technology for electric power supply within vehicles and electric vehicle charging station technology. Currently Thailand has 400 electric vehicle charging stations nationwide [67]. The acquisition of electric energy supplied for charging an electric vehicle is to connect a charging system through a meter to a transformer. If the EV demand grows significantly the share of electricity consumption from industrial and household sectors will be a serious issue. The EV driven 311 km requires approximately 40 kWh of energy which costs approximately 200 THB ($6.40) [68]. Thinking about 10,760,499 EVs in Bangkok, 430 million kWh, worth 2152 million baht per charge will be a requirement [69]. Using renewable energy sources to generate electricity for the charging system should give a positive effect on the country's energy security. Furthermore, an energy storage is needed for the integrated alternative electricity generation. The part aim to emphasize the information related to applying redox flow battery for the integrated charging system. Redox flow battery is considered as the most promising candidate in terms of its unlimited capacity, flexible security design and fast-response [70, 71]. In 2025, the forecast of rechargeable battery in the world expects to increase flow batteries using from 0.4% to 6.6%. Interesting properties of redox flow batteries are 85% of efficiency, 13,000 of cycle life, and 5–80 $/MWh•cycles of capital cost. The major costs of the redox flow battery are electrolyte solution at 37% and stack cell at 31% [72] (**Table 2**).

The priority performance of battery can be considered as following data.

Top power: Flow batteries > Lead acid > Li-ion > Electrolyzer.

The power of flow battery is defined by the size and number of cells.

Top energy: Flow batteries > Li-ion > Electrolyzer > Lead acid.

The energetic capacity is set by the amount of electrolyte stored in the reservoirs.

Discharge time: Flow battery > Electrolyzer > Lithium ion = Lead acid.

Round trip efficiency: Li-ion > Flow batteries > Lead acid > Electrolyzer.

Cycle life: Electrolyzer > Flow batteries > Li-ion > Lead acid.

Capital cost ($/MWh·cycles): Electrolyzer > Li-ion > Lead acid > Flow batteries.

Parameters	Technologies			
	Lead-acid	Li-ion	Electrolyzer/ Fuel cell	Redox flow battery
Top power (MW)	10–40	16	1	2–100
Top energy (MW h)	1–10	20	>10	6–120
Energy density (Wh/kg)	25–50	100–200	800–1300	10–50
Discharge time	1 h	1 h	> 1 h	1–10 h
Response time	ms	ms	ms	ms
Round-trip efficiency	75–85%	95%	35–45%	85%
Cycle life	3000	4000-8000	50,000	> > 13,000
Voltage	2	3.6	1.23	0.7–2.2
Energy cost (k$/kW)	200–400	500–2500	—	150–1000
Power cost ($/kWh)	300–600	175–400	—	600–1500
Capital cost ($/MWh·cycles)	150	150–200	200	<< 70

Table 2.
Performance of energy storage devices [72].

A conventionally single cell of the redox flow battery system consists of two external tanks storing electrolytes, electrodes both anode and cathode sides, a membrane separator, and pumps to generate circulation system. In a discharging process, the anolyte solution is fed in the anode side by the pump and electrolyte flow on bipolar. The electrolyte diffuses through the electrode and it occurs oxidation reaction creating electrons. The electrons move back from an electrode to a bipolar plate and take path around an electrical circuit from the anode side to cathode side. The charge-carrying species directly transfer from the anode to the cathode passing the ion-exchange membrane separating the anolyte and catholyte solution. The catholyte solution is fed in the cathode side, flows on the bipolar plate, and flows via a porous electrode occurring a reduction reaction where catholyte combines with the electron and proton. Once the flow battery is charged, a reduction takes place in the anolyte, while an oxidation is in the catholyte (**Figure 11**) [73, 74].

There are rational decisions for using redox flow battery as a generator in charging station as following examples. People mainly recharge vehicles between 7:00 to 9:00 am, and between 18:00 and 20:00 pm. Grid operators would not be capable to suppress peak power requirements without critically oversizing installed power. Supplement a battery to a fast charging station is a feasible approach to alleviate the oversizing installed power. This electric storage system acts as a buffer to decrease the peak power request on the network without increasing EV charging time [75]. In this situation, the redox flow battery charges installed power during low electricity demand periods to supply electricity for the fast charging. The redox flow battery possesses specific characteristics; ability to decouple rated power from rated capacity, good design flexibility, and nearly unrestricted life. Additionally, the liquid electrolyte contained in the redox flow battery system allowing their installation inside deactivated underground gas container located at gas stations. This installation method authorizes a transition of a conventional gas station to a commercial charging station [76, 77].

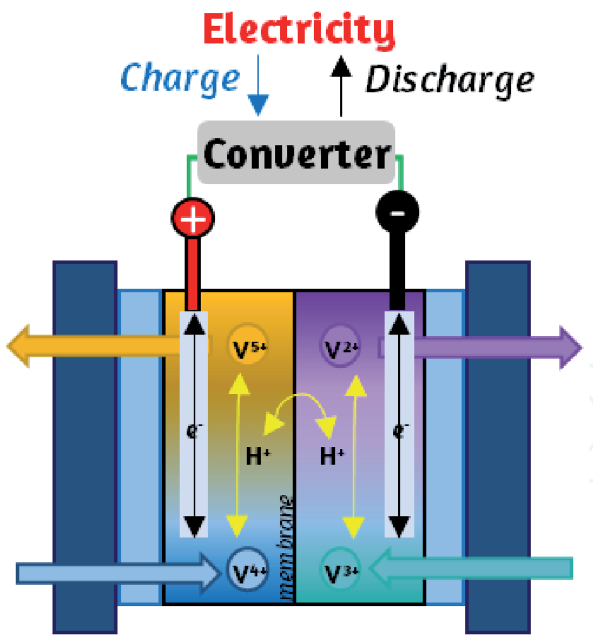

Figure 11.
The schematic diagram of a redox flow battery.

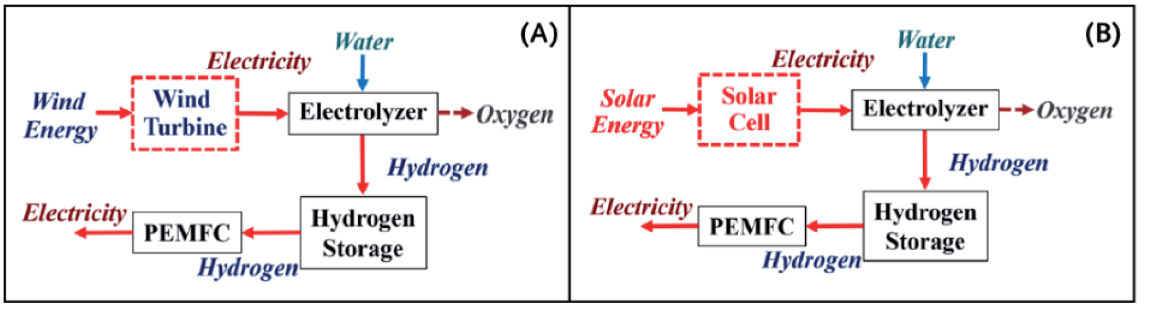

Figure 12.
(A) the overall process of electricity production initiated by a wind turbine. (B) the overall process of electricity generation initiated by a solar cell [79].

In terms of research related to a scaled-up vanadium redox flow battery for HEVs [78], it rapidly charged by electrolyte replacement making it attractive for charging EVs or HEVs. In the not so distant future, redox flow batteries will be useful for EVs, forklift truck, and golf-cart. EVs used for long journey distance may be conceivable if the infrastructure is in place. PEMFC can be another option for charging station when integrated system as primarily renewable source/electrolyzer/PEMFC is constructed. According to these two systems, the wind/PEM electrolyzer/ compressed hydrogen/PEMFC (**Figure 12 (A)**) generates 688.57 W of power. This generated power is significantly higher than the produced power of solar/PEM electrolyzer/ compressed hydrogen/PEMFC (262.37 W) (**Figure 12 (B)**).

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