IEEE Access
Multidisciplinary : Rapid Review : Open Access Journal

Received November 4, 2020, accepted December 2, 2020, date of publication December 14, 2020, date of current version December 29, 2020.

Digital Object Identifier 10.1109/ACCESS.2020.3044362

Hydrogen Leakage Diagnosis for Proton Exchange Membrane Fuel Cell Systems: Methods and Suggestions on Its Application in Fuel Cell Vehicles

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This work was supported by the National Key Research and Development Program of China under Grant 2018YFB0105401.

ABSTRACT Proton exchange membrane fuel cell vehicles using hydrogen as fuel has become one of the key development directions in the field of new energy vehicles. However, hydrogen is flammable, it is easy to trigger serious safety accidents once hydrogen leak occurs. Therefore, the application of hydrogen leakage diagnosis methods is essential for the safe operation of proton exchange membrane fuel cell system. This paper reviews the existing methods of diagnosing hydrogen leakage for proton exchange membrane fuel cell system. The principles, research status and application scopes of different methods are elaborated, and these methods are analyzed based on three aspects of on-line diagnosis, real-time performance and calculation complexity. Furthermore, suggestions are provided for the selection of on-line hydrogen leakage diagnosis methods for fuel cell vehicles. Based on this review, readers can easily understand the current status of hydrogen leakage diagnosis methods for proton exchange membrane fuel cell system, and it can provide references for the application selection of the hydrogen leakage diagnosis methods for fuel cell vehicles.

INDEX TERMS Fuel cell vehicles, hydrogen leakage, PEMFC Systems, diagnosis methods.

NOMENCLATURE

FCVs	Fuel cell vehicles
PEM	Proton exchange membrane
PEMFC	Proton exchange membrane fuel cell
$a1 \ldots a12$	Coefficients
$PH2$	Hydrogen pressure at anode inlet
t	Time
$Ucell$	Output voltage of the cell

I. INTRODUCTION

With the increasingly serious problems of energy and environment, the research on renewable and sustainable energy has arouse wide attention in the world. Because of the impact of the Carnot cycle, just 30% of the energy generated by fuel combustion in internal combustion engine vehicles is used to drive the wheels, while the rest is dissipated by heat, and a large amount of CO, NO, sulfide and CO_2 are discharged in fuel combustion process. With the increasing number of cars in the world, energy shortage, global warming, air quality deterioration and other energy or environmental problems are becoming apparent. For solving the problem of environment pollution aroused by automobile fuel consumption, many countries' governments and world-famous automobile enterprises have invested large amounts of funds in R & D on electric vehicles and fuel cell vehicles (FCVs).

Compared to conventional internal combustion engines, proton exchange membrane fuel cell (PEMFC) powered by hydrogen have the characteristics of higher efficient (up to 60% [1]), lower noise, low operation temperature and less pollutant discharge [2]–[4]. Therefore, FCVs are expected to be the next generation of new energy vehicles [5]. In the past few years, FCVs have been transformed gradually from laboratory validation to commercial applications. At the beginning of 2016, Hyundai, Toyota and Honda launched FCVs into the market, and other automobile manufacturers, like Nikola, also plan to launch their FCVs [6], [7].

The associate editor coordinating the review of this manuscript and approving it for publication was Lei Wang.

Although hydrogen is a clean and renewable energy, it is easy to leak because of its strong permeability. Moreover, hydrogen is colorless and odorless. It has a wider range of combustibility and requires less ignition energy than other flammable gases (e.g., methane). Once hydrogen leakage occurs, it is prone to causing combustion and explosion [8]. In many countries, such as the United States, China, South Korea *et al.*, have occurred safety accidents due to hydrogen leakage. In order to avoid the occurrence of the accidents, the research and application of hydrogen leakage diagnosis technologies have aroused much attention in academic and engineering fields. These technologies make it possible that some emergency measures can be taken in time to ensure the safety of people's life and property. With the rapid development of FCVs, it has become urgent that how to diagnose hydrogen leakage quickly and accurately.

In the existing research, there are many diagnosis methods, either for the hydrogen leakage from the hydrogen supply system or for the hydrogen leakage in the stack. As shown in Figure 1, these methods used different strategies to diagnose hydrogen leakage, mainly including environmental hydrogen concentration diagnosis [24]–[30], hydrogen pressure decay diagnosis [9], [10], [32]–[34], [100], [112]–[114], gas leakage diagnosis by model-based methods or data-driven methods [36]–[50], [115], [123], cross-current diagnosis [10], [100]–[103], and output voltage diagnosis [10], [100], [104]–[111]. The principles of these diagnostic strategies are different, and as such, a comprehensive review of hydrogen leakage diagnosis methods based on these strategies is essential. In addition, with the increasing popularity of FCVs, the selection of FCVs' on-line hydrogen leakage diagnosis methods is of great significance for ensuring safe running of FCVs. Therefore, while reviewing the hydrogen leakage diagnosis methods of PEMFC system, it is necessary to analyze and discuss on-line hydrogen leakage diagnosis methods of FCVs.

This paper aims to provide a comprehensive review of hydrogen leakage diagnosis methods for PEMFC system, where the basic principles, research status and application scopes of the hydrogen leakage diagnosis methods for PEMFC systems are presented. Moreover, they are compared from three aspects of on-line diagnosis, real-time performance and computational complexity. Furthermore, some suggestions are provided on the selection of on-line hydrogen leakage diagnosis methods for FCVs.

Following parts of the paper are organized as follows. The possible causes of hydrogen leakage in PEMFC system are discussed in Section II. The existing hydrogen leakage diagnosis methods for PEMFC are summarized and classified in Section III. In Section IV, different types of hydrogen leakage diagnosis methods are compared, and some suggestions on selection of on-line hydrogen leakage diagnosis methods of FCVs are provided, followed by the conclusions discussed in Section V.

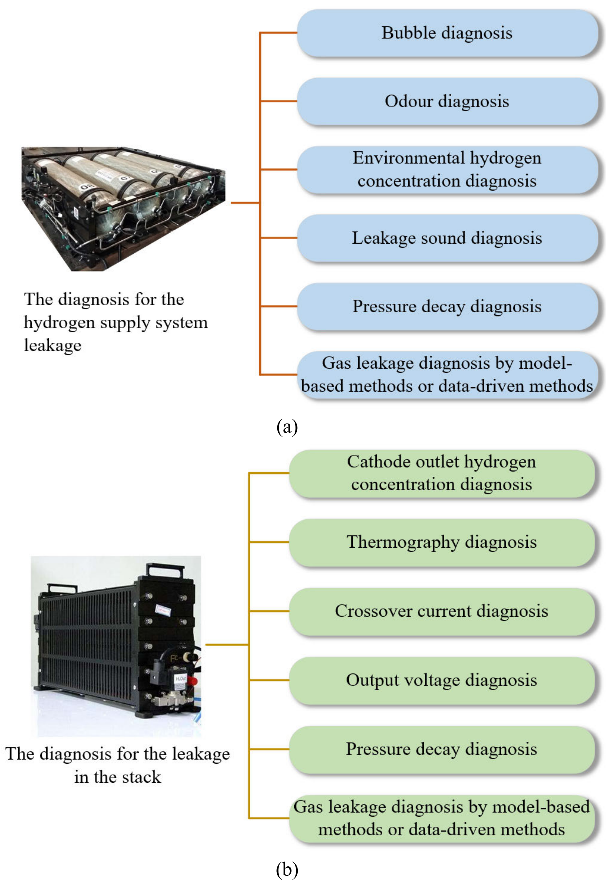

FIGURE 1. Various hydrogen leakage diagnosis methods: (a) The diagnosis for hydrogen supply system leakage; (b) The diagnosis for leakage in the stack.

II. POSSIBLE CAUSES OF PEMFC SYSTEM HYDROGEN LEAKAGE

A. HYDROGEN SUPPLY SYSTEM AND STACK IN FCVs

Figure 2 shows the structure of PEMFC system in FCVs, which includes a hydrogen supply system, air supply system, cooling system and stack, etc. The hydrogen supply

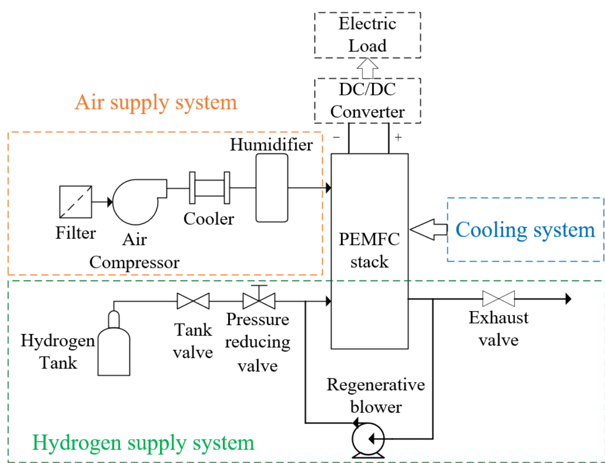

FIGURE 2. The structure diagram of PEMFC system [16], [95].

system is responsible for supplying hydrogen to the stack. The hydrogen tank is used to store high-pressure hydrogen, which can be opened or closed through the tank valve. The pressure reducing valve can reduces the hydrogen pressure to an appropriate range. After hydrogen enters the stack anode, some of it participates in the electrochemical reaction generate water, and the remaining hydrogen flows out from the stack anode. After which, it is sent to the anode inlet by a regenerative blower to improve the utilization rate of hydrogen. In some hydrogen supply systems, instead of the regenerative blower, an ejector or electrochemical pump is used to achieve hydrogen reuse [80]. In order to remove impurities (e.g., liquid water, nitrogen permeated through the membrane to the anode, etc.) in the stack anode, the exhaust valve is opened at intervals [51].

The PEMFC in the stack consists of an anode plate, a cathode plate, and a membrane electrode assembly which includes diffusion layers, catalytic layers and a proton exchange membrane (PEM). The hydrogen gas on the anode side resolves into protons and electrons. The protons pass through the PEM to the cathode, and the electrons pass through the external load circuits to the cathode and generate electric power in the process. Oxygen, electron, and hydrogen ions combine in the cathode catalytic layer to form water. The electrochemical reactions in the PEMFC stacks can be described as follows [9], [84]:

In the anode:

$$H_2 \rightarrow 2H^+ + 2e^- \tag{1}$$

In the cathode:

$$2H^+ + 2e^- + 1/2O_2 \rightarrow H_2O \tag{2}$$

B. POSSIBLE CAUSES OF THE PEMFC HYDROGEN SUPPLY SYSTEM LEAKAGE

In order to extend the range of FCVs, the gas pressure in the hydrogen supply system reaches up to 70MPa [11], [12], which means that hydrogen leakage is prone to occur. The leakage of the hydrogen supply system can mainly be attributed to the degradation of the seals used to connect the pipelines and components as well as the degradation of the sealing structure in the system components (e.g., valves, pumps). At present, the sealing methods used in the hydrogen supply system generally include two types: metal-to-metal seals (e.g., flared or compression joints) and nonmetal seals (e.g., gaskets, packings and pipe thread compounds). Possible reasons for the sealing performance deterioration of the metal-to-metal seals are thermal expansion/contraction and vibration, both of which can increase the seal gap among sealing structures and thereby result in gas leakage [8]. For nonmetal seals, the aging of the sealing material [8], [121] and the bad operating environment (e.g., overly temperature or pressure hydrogen in seals or components) can lead to gas leakage. In addition, strong external extrusion or collision are also important reasons for hydrogen leakage from hydrogen supply system [8], [9]. The most dangerous situation is vehicle collision caused by traffic accidents. Vehicle collision may lead to severe distortion of the sealing structure in the hydrogen supply system or rupture of the hydrogen tank, pipeline and the shell of components, which will result in hydrogen leaks with large flow rates. It is estimated the conditional probability that a vehicle accident would result in sufficient rear damage such that hydrogen storage or delivery components could be breached to be 0.76% [14], [15]. This shows that, for FCVs, one out of every 132 accidents will result in hydrogen leakage.

C. POSSIBLE CAUSES OF THE PEMFC STACK HYDROGEN LEAKAGE

In general, there are two situations for the hydrogen leakage of the stack: hydrogen leakage caused by reduced sealing performance between various components in the stack and hydrogen crossover leakage due to PEM rupturing (or holes) in the stack. In some circumstances, sealing performance is negatively affected by deformation or damage to the stack's internal structure caused by external extrusion or collision [9], aging of sealing materials in the stack [121], mechanical vibration, and a bad operating environment. In terms of hydrogen crossover leakage, PEM rupturing or holes are usually the result of the following: hydrogen starvation [104], a large pressure difference between the cathode and anode in the stack [9], [45], too high temperature inside the stack [9], and strong external extrusion or collision [9].

When hydrogen starvation occurs, the stack will suffer from a reverse potential fault, which can lead to anode catalyst degradation and holes in the membrane due to local heat generation [104]. Hydrogen starvation can be caused by insufficient hydrogen due to a hydrogen supply system failure or a sudden increase in the external connective load. In addition, when flooding occurs in the stack, liquid water can block the gas flow channel, which causes starvation. The pressure difference between the cathode and anode in the stack will lead to unequal forces on the PEM sides. When the pressure difference between the membrane is too large, it will lead to membrane rupture and, as a result, hydrogen crossover leakage occurs. The large pressure difference between the cathode and anode can be caused by an insufficient or excessive fuel supply due to a failure of the hydrogen supply system, failure of the air supply system, or failure of pressure sensors in the stack. When the temperature inside the stack is too high, the PEM will dehydrate, shrink, or even rupture, which will lead to hydrogen crossover leakage. Too high temperature inside the stack may be caused by a failure of the stack cooling system, failure of the temperature sensor in the stack, or serious long-term overload. When the stack is subjected to strong external extrusion or collision, the PEM will rupture due to deformation or penetration by other objects, which will lead to the occurrence of hydrogen crossover leakage in the stack. In addition, an increase of clamping pressure will also lead to an increase in the flow rate of hydrogen crossover leakage in the stack [17]. The hydrogen crossover leakage in the stack will decrease the

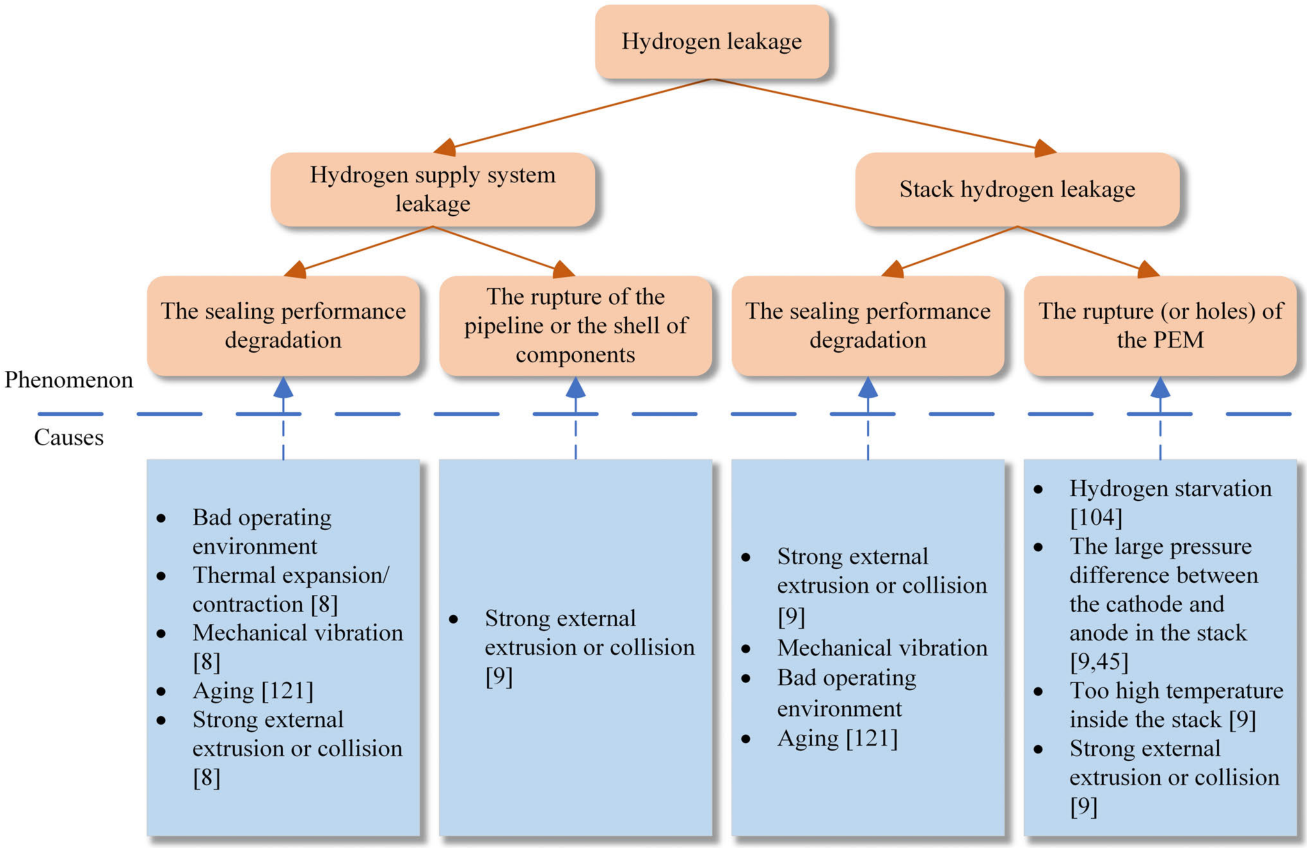

FIGURE 3. Summary of possible causes of the PEMFC system hydrogen leakage.

hydrogen utilization rate; more importantly, it can result in dangerous accident (e.g., combustion or explosion) once the hydrogen in the anode mixes with the air in the stack cathode. The possible causes of the PEMFC system hydrogen leakage are summarized in Figure 3.

III. HYDROGEN LEAKAGE DIAGNOSIS METHODS FOR PEMFC SYSTEM

A. LEAKAGE DIAGNOSIS METHODS FOR THE HYDROGEN SUPPLY SYSTEM

In the existing research, there have been some methods that are proposed for leakage diagnosis in the PEMFC hydrogen supply system, including the bubble diagnosis, the odour diagnosis, the environmental hydrogen concentration diagnosis, the leakage sound diagnosis, the pressure decay diagnosis and the gas leakage diagnosis by model-based methods or data-driven methods. The detailed introduction of the leakage diagnosis methods for the hydrogen supply system are presented as follows.

1) BUBBLE DIAGNOSIS

Bubble diagnosis is to judge the leakage from hydrogen supply system by observing whether bubbles occur after the gas leakage detection agent is applied to the part of the fuel cell hydrogen supply system where the hydrogen leakage may occur. The bubble diagnosis is effective for the diagnosis of hydrogen leakage at the pipes, valves, connections between pipes and valves, and connections between pipes and sensors in the hydrogen supply system of the FCVs. Its operation is simple, and it is widely used for hydrogen leakage diagnosis in the process of vehicle fuel cell system integration tests and related Test-beds test [9].

2) ODOUR DIAGNOSIS

In the odour diagnosis, odorants which have special odor and can be recognized by humans are added to hydrogen. In case of the leakage occurs, drivers, passengers or other related personnel can recognize the hydrogen leakage from its special odor. Under the premise that the relevant personnel are on site, it can be used to recognize hydrogen leakage in the process of vehicle fuel cell system integration tests and related Test-beds test, as well as to recognize hydrogen leakage during FCVs parking or running.

Imamura *et al.* [18] compared the effects of various sulfur compounds and sulfur-free compounds odorants on the performance of fuel cells and their condensation performance under high pressure conditions. Hibino *et al.* [19] suggested organic acids (e.g., acetic acid and butyric acid) be used as odorants for recognizing hydrogen leakage. Kopasz *et al.* [20] discussed the feasibility of using chemicals containing sulfur, nitrogen-based compounds, 5-ethylidene-2-norbornene, ethyl isobutyrate and 2,3-butanedioneas as hydrogen odorants. However, the results show that these compounds will affect the performance of the fuel cell system. In their study, it is considered that hydrogen concentration diagnosis method is more suitable for hydrogen leakage diagnosis than odorants [20]. Therefore, the odour diagnosis has much to be desired for its practicability when applied to diagnose the hydrogen leakage from the PEMFC hydrogen supply system.

3) ENVIRONMENTAL HYDROGEN CONCENTRATION DIAGNOSIS

In the environmental hydrogen concentration diagnosis, hydrogen concentration sensors are installed in the workshop, laboratory, inside or around the vehicle to measure the concentration of hydrogen in the air. Trocin *et al.* [21] introduced nanocomposites based on Pt-doped TiO_2/multiwalled carbon nanotubes as sensitive materials for hydrogen at low temperatures. Pt-TiO_2/multiwalled carbon nanotubes based sensors are sensitive to hydrogen at volume concentrations between 0.5% and 3% in air, which can meet the requirements for practical applications. Yu *et al.* [22] proposed a tilted fiber Bragg grating hydrogen sensor coated with a palladium membrane by the electroless plating method, it is able to identify the hydrogen in 5 minute at a 1% hydrogen volume concentration. Chauhan and Bhattacharya [23] reviewed different sensing technologies used in existing hydrogen concentration sensors and various materials used in hydrogen sensing. In addition, Sun *et al.* [122] proposed a data-driven diagnosis method based on the convolutional neural network and the random forest for different failures of hydrogen concentration sensors (such as impact fault, stuck fault, etc.)

The methods based on environmental hydrogen concentration diagnosis are to diagnose hydrogen leakage by measuring hydrogen concentration in the air. Therefore, no matter the flow rate of hydrogen leakage is large or small, the methods can realize the diagnosis effectively as long as the leaked hydrogen concentration in the air is greater than the threshold (e.g., volume concentration 0.1%). Researchers [13], [24]–[30] used the hydrogen concentration sensors to diagnose the PEMFC system hydrogen leakage. Generally, the installation positions of hydrogen concentration sensors inside fuel cell car are as shown in Figure 4. The method based on environmental hydrogen concentration diagnosis is currently the most widely used hydrogen leakage diagnosis method in workshops, laboratories, hydrogen refueling stations and FCVs.

4) LEAKAGE SOUND DIAGNOSIS

In the leakage sound diagnosis, hydrogen leakage is diagnosed by recognizing the special sound generated when the hydrogen leakage occurs. However, there are few related studies. Maeda *et al.* [31] proposed a method to diagnose hydrogen leakage of FCVs by recognizing hydrogen leakage sound. The schematic of leakage sound measurement is shown in Figure 5. In addition, the influences of different directions, heights and distances on the recognition effects of hydrogen leakage sound are discussed. Finally, in a 74dB traffic noise environment, the threshold value of the hydrogen leakage flow rate that the leakage sound can be perceived by humans at 5-10m away from the leakage position is determined.

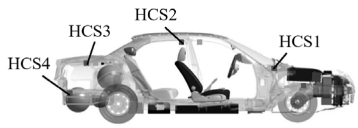

FIGURE 4. The installation positions of hydrogen concentration sensors inside fuel cell car [28].

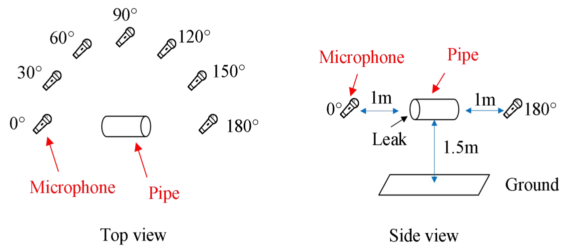

FIGURE 5. The schematic of leakage sound measurement [31].

5) PRESSURE DECAY DIAGNOSIS

In the pressure decay diagnosis, pressure sensors are used to diagnose hydrogen leakage by measuring the gas pressure decay in the closed areas of the system after a period of time [9]. Tachibana [32] proposed a method to diagnose hydrogen leakage by measuring the gas pressure decay in the pipeline. At the start of the PEMFC stack, hydrogen is filled into the low-pressure pipeline of the hydrogen supply system through an injector in the hydrogen supply system. After the hydrogen in low pressure pipelines reaches a certain pressure, the pipeline is sealed off. The hydrogen pressure decay in the pipeline is measured to determine whether there is hydrogen leakage from the low-pressure pipeline. Yoshida [33] proposed a method for hydrogen leakage diagnosis. In their study, hydrogen leakage is diagnosed by measuring the hydrogen pressure decay during the fuel cell system activation, intermittent operation and stoppage. By closing the tank valve, the hydrogen supply valve, the stack inlet valve, the stack outlet valve, etc., the high-pressure area, the low-pressure area, the hydrogen circulation area of the hydrogen supply system and the stack anode are sealed in sequence, and then the hydrogen leakage is diagnosed by measuring the hydrogen pressure decay in each area after a period of time. Imanishi and Toida [34] proposed a hydrogen leakage diagnosis method. In their proposed method, when the operation of fuel cell system in the vehicle is stopped, hydrogen is filled into the low pressure pipes and the circulation circuit of hydrogen supply system until the internal pressure of the pipes reaches a certain value, and then the relevant valves are closed to seal the low pressure pipes and the circulation circuit. When the fuel cell system is started again, the gas pressure in the closed pipe is measured, the gas leakage is recognized by the decay of the gas pressure in the sealed pipe. It can be seen from [32]–[34] that the methods based on pressure decay diagnosis can identify the hydrogen leakage only through the gas pressure in the system. It can diagnose hydrogen leaks with small flow rates by extending the diagnosis time and locate the leakage area. It can be used to diagnose hydrogen leakage during the PEMFC system activation, intermittent operation and stoppage.

6) GAS LEAKAGE DIAGNOSIS BY MODEL-BASED METHODS OR DATA-DRIVEN METHODS

In the gas leakage diagnosis by model-based methods or data-driven methods, relevant pattern recognition algorithms or other diagnostic strategies are used to diagnose hydrogen leakage from fuel cell hydrogen supply system by system state data collected by relevant sensors (e.g., temperature sensors and pressure sensors) which were installed in the PEMFC system.

The model-based diagnosis methods are to diagnose the hydrogen leakage of the system based on a comparison of the system model's output and the actual system's output [35] (as shown in Figure 6a). Lira *et al.* [36] established a fuel cell linear parameter varying model and a Luenberger observer for PEMFC system. The residuals, between the measured outputs and the outputs estimated by an observer of the faultless model, were used to calculate the fault sensitivities and the Euclidean distances corresponding to different faults, so as to realize the diagnosis of hydrogen leakage from the PEMFC hydrogen supply system. Li *et al.* [37] proposed a hierarchical fault diagnostic method for PEMFC system. Firstly, faults were diagnosed at the subsystem level by the residuals between the model outputs and the measured outputs. Secondly, artificial neural networks, support vector

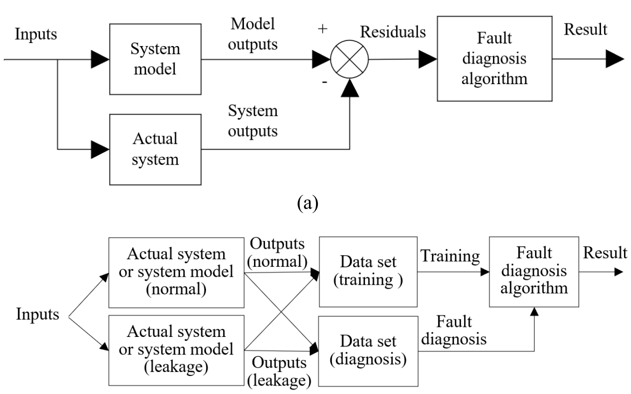

FIGURE 6. Schematic diagram of the gas leakage diagnosis by model-based methods or data-driven methods: (a) The model-based methods (b) The data-driven methods.

machine, discriminant analysis, the Naive Bayes model and k-nearest neighbor were used to isolated fault (e.g., hydrogen leakage) in the corresponding subsystem, and the diagnostic effects of 5 different diagnostic algorithms are compared. Vijay *et al.* [38] designed an adaptive observer based on the solid oxide fuel cell dynamic model, and realized the diagnosis of hydrogen leakage based on the residuals between the system state variables collected by the sensors and the outputs of the adaptive observer. Pellaco *et al.* [123] proposed a solid oxide fuel cell fault diagnosis method based on a quantitative model of the solid oxide fuel cell plant and support vector machine. Faults were simulated by modifying model parameters. Support vector machine was used to diagnose a leak between the exit of the reformer and the entrance of the solid oxide fuel cell stack by the residuals.

In addition, many scholars used the model-based diagnosis methods to diagnose the gas leakage from fuel cell air supply system [39]–[43], which provide important references for the design of hydrogen leakage fault diagnosis methods. Kamal *et al.* [39] established an independent radial basis function network model. Parameters at the different historical time steps was used for inputs: stack current and voltage, compressor control voltage, the partial pressure ratio of used oxygen and remaining oxygen in the stack cathode, and stack net power output. For the outputs, they used the stack voltage of the current moment, the partial pressure ratio of used oxygen and remaining oxygen of the current moment in the stack cathode, and the stack net power output of the current moment. The residuals between the model outputs and the actual outputs were used as the inputs of radial basis function network classifier to diagnose air supply manifold leakage and other faults. Escobet *et al.* [40] calculated the relative fault characteristic sensitivities using the residuals between the model outputs in normal operation and the model outputs under the circumstance of faults, and combined them into a fault characteristic matrix. The diagnosis and isolation of air supply manifold leakage and other faults of PEMFC system were realized by calculating the Euclidean distance between the observed fault characteristics and the theoretical characteristics of each fault. Rosich *et al.* [41], [42] proposed a method of extracting residuals from system model based on causality. The calculation formulas and redundancy formulas of variables of air supply system were selected from the air supply system model, and the fault simulation was realized by changing the parameters of the system model. The residuals of the calculation results based on the calculation formulas and the redundant formulas of variables were used to diagnose the air supply manifold leakage and other faults. Escobet *et al.* [43] proposed a leakage diagnosis method based on the fuzzy inductive reasoning methodology. In their study, the fuzzy inductive reasoning qualitative models of the PEMFC system in normal operation and failure mode were established. Air supply manifold leakage was diagnosed by checking the consistencies between measured system outputs and model outputs. Laghrouche *et al.* [124] proposed a fault reconstruction method base on the adaptive second-order sliding mode observer, which is helpful for the diagnosis and tolerance of air leak in the air supply manifold.

In recent years, with the rapid development of data mining technologies, the data-driven gas leakage diagnosis methods have been applied widely [44]. The data-driven methods realize the diagnosis of hydrogen leakage from hydrogen supply system by fault features, which are extracted from the state data of system in case of normal and hydrogen leakage (as shown in Figure 6b). The procedure of the data-driven diagnosis methods usually proceeds in two steps. First, an classifier is established from history data. This is considered as the training process. Then, by using the trained classifier, the state data are classified into certain classes that correspond to the health states (normal state or leakage states) [125]. Liu *et al.* [45] used the state data of the system under the normal and hydrogen leakage states of the PEMFC hydrogen supply system to establish a discrete hidden Markov model, and then used the maximum value of the model output probability to diagnose the hydrogen leakage. Zhang *et al.* [46] achieved the accurate diagnosis of single-point hydrogen or air leakage and multi-point hydrogen and air simultaneous leakage from solid oxide fuel cell hydrogen and air supply system by stack sparse automatic encoder which was trained with the system state data. By using a multi-label pattern recognition method based on the multi-label support vector machine trained by the system state data, Li *et al.* [47] realized the diagnosis of multi-point hydrogen simultaneous leakage from the solid oxide fuel cell hydrogen supply system. Costamagna *et al.* [48] realized the diagnosis of hydrogen leakage from the solid oxide fuel cell hydrogen supply system based on support vector machine and random forest. The diagnosis effects using support vector machine and random forest are further compared in their study. In addition, the differences between data-driven fault diagnosis and isolation system and hybrid fault diagnosis and isolation system are also analyzed in [48].

It can be seen from [35]–[48] that the model-based methods and data-driven methods can diagnose the gas leakage through the operating state data of the fuel cell system collected by the sensors. Therefore, they can be used to diagnose hydrogen leakage during the fuel cell system testing or FCVs running. However, from the perspective of method design, the diagnosis effect of the model-based gas leakage diagnosis methods has a strong correlation with the accuracy of the system model. Therefore, the establishment of an accurate system model is a premise for the model-based gas leakage diagnosis methods to achieve an accurate diagnosis of the gas leakage. Although the data-driven gas leakage diagnosis just require the state data of the hydrogen supply system under the states of normal and hydrogen leakage to realize the diagnosis of the hydrogen leakage, it is dangerous to collect the training and test data which are required for the development of fault diagnosis algorithms by hydrogen leakage experiments. Therefore, in [46], [47], [49], [50], the training and test data were collected by simulating hydrogen leakage fault in the model. It can be known that, either for the model-based gas leakage diagnosis or the data-driven gas leakage diagnosis, it is of great significance to establish accurate simulation models of fuel cell hydrogen supply systems with different structures for the research of hydrogen leakage diagnosis of fuel cell system. At present, many scholars have established mechanism models of hydrogen supply systems with different structures [51]–[60] and key components in hydrogen supply system [61]–[83]. These models provided important references for the design of the model-based methods or data-driven methods. For some uncertain parameters in the modeling process, the parameter identification methods described in [84] and the experimental data can be used to certain the relevant parameters of the system model. In addition, in the process of the design of the data-driven diagnosis methods, the state data of the system in case of normal and hydrogen leakage are required to train and test the diagnosis method. Therefore, it is necessary to use a high-accuracy fault simulation method to simulate the hydrogen leakage fault. At present, many scholars have used different methods to establish hydrogen leakage models [85]–[92], these models can be combined with the hydrogen supply system models to simulate the hydrogen leakage, so that the state data of system in case of hydrogen leakage can be obtained by the model.

B. DIAGNOSIS METHODS FOR THE HYDROGEN LEAKAGE IN THE PEMFC STACK

In the existing research, there have been some methods that are proposed for hydrogen leakage diagnosis in the PEMFC stack, including the cathode outlet hydrogen concentration diagnosis, the thermography diagnosis, the crossover current diagnosis, the output voltage diagnosis, the gas pressure decay diagnosis, and the gas leakage diagnosis by model-based methods or data-driven methods. The detailed introduction of the hydrogen leakage diagnosis methods for the stack are presented as follows.

1) CATHODE OUTLET HYDROGEN CONCENTRATION DIAGNOSIS

When hydrogen crossover leaks with small flow rates in the stack occurs, the hydrogen leaking to the cathode of the stack will react with the oxygen in the cathode. Therefore, it is difficult to detect the presence of hydrogen at the cathode outlet of the stack. However, when hydrogen crossover leaks with large flow rates in the stack occurs, the hydrogen leaking to the cathode of the stack will react with oxygen, and then resulting in oxygen starvation. Some unreacted hydrogen will be discharged from the cathode outlet of the stack [93]. Therefore, the diagnosis of the hydrogen crossover leakage can be achieved by measuring the hydrogen concentration using the mass spectrometer or hydrogen concentration sensor at the cathode outlet (as shown in Figure 7).

Jung *et al.* [17] and Baik *et al.* [94] studied the effects of different clamping pressures, different temperatures in the stack, different humidity in the stack, and different hydrogen pressures in the stack anode on the hydrogen crossover leakage in the stack. In [17], [94], the mass spectrometer is

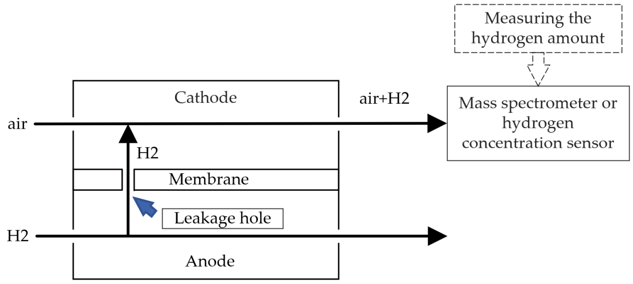

FIGURE 7. Schematic diagram of the cathode outlet hydrogen concentration diagnosis.

used to measure the hydrogen concentration at the cathode outlet of the stack that the flow rate of the hydrogen crossover leakage in the stack can be estimated. Narimani *et al.* [93] used the hydrogen concentration measurements to measure the hydrogen concentration at the cathode outlet of the stack (as shown in Figure 8), and studied the relationship between the hydrogen crossover leakage flow rate in the stack and the hydrogen concentration at the cathode outlet of the stack. It can be seen from [17], [93], [94] that the cathode outlet hydrogen concentration diagnosis is simple, and it can be used to diagnose the hydrogen crossover leakage in the stack during the relevant fuel cell experiments.

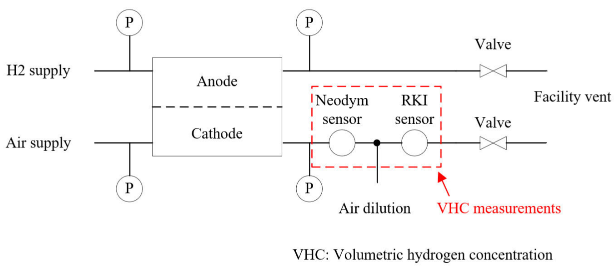

FIGURE 8. A schematic diagram of experimental setup [93], [119].

2) THERMOGRAPHY DIAGNOSIS

In the thermography diagnosis, the hydrogen crossover leakage in the stack can be diagnosed by detecting the heat which is generated by the reaction of hydrogen leaking across the membrane and oxygen at the cathode (as shown in Figure 9).

By using thermal imaging camera to detect the local temperature rise in the stack caused by hydrogen crossover leakage, Asghari *et al.* [10] realized the diagnosis of hydrogen crossover leakage in the PEMFC stack. Phillips *et al.* [96] and ulsh *et al.* [97] both proposed a method which can locate the pinholes on the PEM by detecting the heat released from the reaction process of hydrogen and oxygen with infrared thermal detector. Bender *et al.* [98] realized the location of pinholes and failure points in PEM after accelerated stress test using IR thermography. It can be seen from [10], [96]–[98] that the methods based on the thermography diagnosis can diagnose hydrogen crossover leaks with the small flow rates

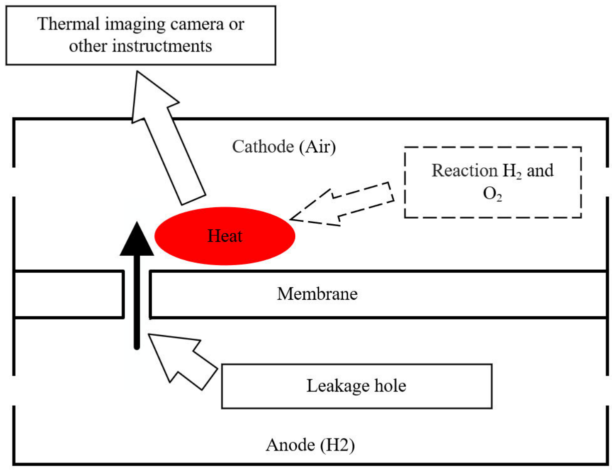

FIGURE 9. Schematic diagram of the thermography diagnosis.

(the hydrogen transported through the a membrane pinhole can be identified when the area of the pinhole is only 0.0071471mm2 [97]), which is suitable for off-line diagnosis of hydrogen crossover leakage in the fuel cell stack and the diagnosis of membrane defects during production or test of the PEM.

3) CROSSOVER CURRENT DIAGNOSIS

The crossover current diagnosis is to diagnose the hydrogen crossover leakage in the stack by measuring the crossover current. Nitrogen and hydrogen are delivered respectively to two different compartments (e.g. cathode and anode) in the stack, and then the voltage is applied across each individual cell in the stack. In the cells with crossover leakage, the hydrogen leaking to the nitrogen side is oxidized under the action of external applied voltage, thus generating crossover current (as shown in Figure 10).

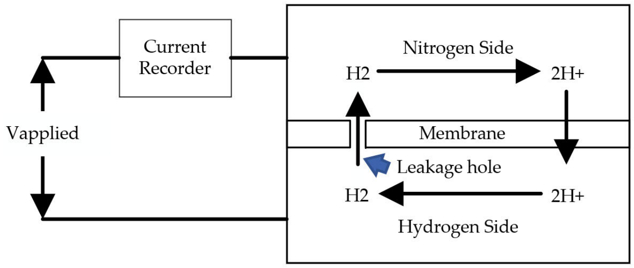

FIGURE 10. Schematic diagram of the crossover current diagnosis [10].

Under the high potential, the hydrogen leaking to the stack cathode should be oxidized, which can instantaneously generate a limiting hydrogen oxidation current density. Since nitrogen is the only substance introduced into the stack cathode, any current generated in the given potential range is solely attributable to the electrochemical oxidation of hydrogen into the stack cathode. Thus, the hydrogen crossover leakage in the stack can be diagnosed by measuring the crossover current. The reaction in the anode and cathode of fuel cell stack during the hydrogen crossover leakage diagnosis can be expressed as [99]:

$$\text{Cathode}: H_2 \rightarrow 2H^+ + 2e^- \tag{3}$$

$$\text{Anode}: 2H^+ + 2e^- \rightarrow H_2 \tag{4}$$

Asghari *et al.* [10] proposed a method for identifying the cells with crossover leakage by crossover currents generated by different cells in the stack, and the types of leakage in the cells were analyzed by a comparison between crossover currents in the direct and reverse tests. Chen and Kaye [100] realized the location of the cells with crossover leakage in the stack by measuring the crossover current of each single cell in the stack, and the effects of nitrogen flow rate and applied voltage on the crossover current were studied. Pei *et al.* [101] studied the influence of temperature and humidity in the stack on the hydrogen crossover leakage flow rate and crossover current. In fully humidified gases, with the increase of the cell temperature, the micro-channels become larger due to the expansion and the hydrogen molecule movement become severer, so the flow rate of hydrogen crossover leakage and crossover current increase linearly. With the humidity in the stack increases, the volume of impregnated polymer increases, which leads to the decrease of porosity of PEMs and prevents the hydrogen penetration. However, this conclusion is not consistent with the conclusion provided in [17], which may be resulted by different operating conditions of fuel cells. Therefore, further research is required in the future. In addition, Pei *et al.* and Gunji *et al.* [102], [103] proposed several improved methods for measuring crossover current. It can be found from [10], [100]–[103] that the methods based on the crossover current diagnosis can accurately locate cells with hydrogen crossover leakage in the stack. It is applicable to the diagnosis of hydrogen crossover leakage in fuel cell stacks during the manufacturing and testing of fuel cell stacks [100].

4) OUTPUT VOLTAGE DIAGNOSIS

In the output voltage diagnosis, hydrogen leakage in the stack (or single cell) is diagnosed by detecting the abnormality of the stack (or single cell) output voltage caused by the hydrogen leakage in the stack (or single cell). In [10], [100], [104]–[110], the different diagnosis methods based on the output voltage diagnosis for diagnosing hydrogen crossover leakage in the stack (or single cell) have been proposed respectively.

Under the condition of normal and hydrogen crossover leakage, Mousa *et al.* [104], [105] analyzed the influence of different oxygen concentration and the pressure difference between the cathode and anode on the output impedance. In their studies, the impedance signature model of the fuel cell stack was established based on the Randles model, and the output voltage of the stack was estimated by the impedance signature. Finally, the fuzzy rule was established to judge the hydrogen leakage by taking the difference of the stack output voltage measured by the actual system under different pressure differences between the cathode and anode, and the difference of the stack output voltage estimated by impedance signature model under different pressure differences between the cathode and anode.

Tian *et al.* [106], [107] injected hydrogen and air into the stack anode and cathode, and then cut off the reaction gas supply. When crossover leakage occurred in cells, the gas in the anode of the cells was consumed rapidly, and the open-loop voltage of the cells with hydrogen crossover leakage decayed faster than that of normal cells [106]. Therefore, the hydrogen crossover leakage in the stack can be diagnosed by measuring the open-loop voltage decay of each cell.

Asghari *et al.* [10] and Chen *et al.* [100] injected hydrogen and air into the anode and cathode of the stack and pressurized the hydrogen in the anode. The pressure difference between the cathode and the anode caused hydrogen to leak into the cathode, which reacted with oxygen. This resulted in the rapid consumption of oxygen in the cathode of cells with hydrogen crossover leakage [10]. Therefore, after the hydrogen in the anode was pressurized, the hydrogen crossover leakage in the stack can be diagnosed by measuring the decay in the open-loop voltage of every cell. In addition, Chen *et al.* [100] proposed another method that can diagnose hydrogen crossover leakage in the cells by measuring the recovery speed of the cells voltage after interrupting the output current of the stack. However, this method has low sensitivity in diagnosing hydrogen crossover leakage in the stack [100].

Niroumand *et al.* [108] proposed a diagnosis method for hydrogen crossover leakage in the stack during the development of the fuel cell stack. This method can estimate the hydrogen crossover leakage flow rate based on parameters, including the cell open-loop voltage, the humidity in the stack anode and cathode, the stack temperature, and the gas pressure in the stack anode and cathode. Based on the phenomenon that the cells output voltage drops rapidly after lowering the air flow rate at the stack cathode, Niroumand *et al.* [109] proposed the formulas for calculating the flow rate of the hydrogen crossover leakage in the stack and the number of cells with crossover leakage in the stack. In addition, Niroumand *et al.* [111] proposed a method of diagnosing hydrogen crossover leakage, which can be used in the process of stack manufacturing. This method uses the proposed formulas to estimate the flow rate of the hydrogen crossover leakage through the open loop output voltage of a single cell in the stack and other system parameters. In addition, the calculation formula of anode overpressure was proposed in [111]. It can be used to calculate anode overpressure which can result in the open loop voltage drop of cells caused by hydrogen crossover leakage reaching a certain fixed value.

Okui [110] analyzed the cause of hydrogen crossover leakage in the fuel cell stack. Based on the phenomenon that an increase in the membrane electrode water content can increase the stack output voltage (when the water content of membrane electrode increases and the stack output current remains unchanged, the liquid water will block the holes in the membrane electrode, thereby resulting in a decrease in hydrogen crossover leakage and an increase in stack output voltage), the hydrogen crossover leakage can be diagnosed by measuring the stack output voltage after the humidity of the membrane electrode increased.

In addition, Tian *et al.* [106] also proposed three diagnosis methods for the leakage between the anode and the cooling compartment in the stack. Method 1: After turning on the hydrogen and air supply of the stack separately, the water level in the expansion vessel of the cooling circuit should be observed with respect to whether it rises. If it rises slightly when the hydrogen supply is turned on separately, then leakage has occurred between the anode and the cooling compartment. Then the cells with leakage can be located by measuring the voltage decay of each cell after turning off the hydrogen supply and the voltage rise of each cell after turning on the hydrogen supply. Method 2: After filling the cooling compartment of the stack with hydrogen, the anode and cathode of the stack are sealed. When the hydrogen in the cooling compartment leaks to the anode, the stack output voltage increases. Therefore, the hydrogen leakage can be diagnosed by measuring the change of the stack output voltage. Method 3: The cubic polynomial model based on the relationship among the output voltage of the each cell, the time, and the hydrogen pressure at the anode inlet can be established by fitting the cell output voltage data and the hydrogen pressure data (see (5)).

$$\begin{aligned} Ucell = {} & a1 + a2 \cdot t + a3 \cdot t^2 + a4 \cdot t^3 + a5 \cdot PH2 \\ & + a6 \cdot PH2^2 + a7 \cdot PH2^3 + a8 \cdot t \times PH2 + a9 \cdot t \\ & \times PH2^2 + a10 \cdot t^2 \times PH2 + a11 \cdot \ln(PH2) \\ & + a12 \cdot t \times \ln(PH2) \end{aligned} \tag{5}$$

where $Ucell$ is the output voltage of the cell, t is time, $PH2$ is the hydrogen pressure at the anode inlet, and $a1 \ldots a12$ are the coefficients. The hydrogen leakage in cells can be diagnosed by comparing the differences among the polynomials coefficients, which are fitted by the data of cells with leakage and the data of normal cells.

It can be seen from [10], [100], [104]–[110] that the methods based on the output voltage diagnosis can realize the diagnosis of hydrogen leakage in the stack only by comparing the output voltages of the normal stack (or single cell) and the stack (or single cell) with leakage. The cells with leakage in the stack can be located, and some methods proposed in [106], [109], [110] can realize on-line hydrogen leakage diagnosis in the running process of FCVs. Therefore, the output voltage diagnosis has a wide application range. It can be used for the off-line diagnosis of hydrogen leakage in the stack (or single cell) during the manufacturing and testing of the fuel cell stack, and it can also be used for the on-line diagnosis of the hydrogen leakage in the stack (or single cell) in the running process of the FCVs.

5) OTHER DIAGNOSIS METHODS

In addition to the above methods, the pressure decay diagnosis and the gas leakage diagnosis by model-based methods or data-driven methods can also be used to diagnose hydrogen leakage in the stack. Methods based on the pressure decay diagnosis for diagnosing hydrogen leakage in the stack were proposed in [10], [112]–[114]. Salvador [112] introduced a method of diagnosing hydrogen leakage in the stack at the PEMFC system shutdown. Firstly, relevant operations were performed to exhaust the oxygen in the stack cathode, and then the relevant valves were closed to seal the anode after the stack anode was pressurized to a certain value. The hydrogen leakage in the stack can be judged by monitoring the decay of gas pressure in the stack anode in a period of time. Harris [113] also described a method to diagnose the hydrogen leakage in the stack when the PEMFC system in idle-stop mode (little or no power is being drawn from the stack). The diagnosis of hydrogen leakage in the stack was realized by comparing the preset value and the actual value of the hydrogen supply flow rate required to maintain the anode pressure of the stack during the idle-stop mode. Song [114] used the gas tightness detection device to fill nitrogen into the anode or cathode compartment of the stack and maintain the nitrogen pressure in the compartment. Then, the flow rate of gas leakage in the stack was estimated by measuring the flow rate of the nitrogen from the rest compartment in the stack. Asghari *et al.* [10] pressurized one of compartments in the stack to the set value and then seal other compartments under the environmental pressure. The gas leakage in the stack was diagnosed by measuring the increase of gas pressure in the sealed compartment.

In [49], [50], [115]–[117], various model-based methods and data-driven methods were proposed to diagnose the hydrogen leakage in the stack. Ingimundarson *et al.* [115] diagnosed the hydrogen leakage in the stack with the difference between the mass flow rate of hydrogen supplied to the stack, the mass rate of hydrogen consumed by the reaction, and the mass flow rate of hydrogen due to the natural leakage. Riascos *et al.* [49] used the Bayesian network to diagnose hydrogen crossover leakage in the stack by qualifying and quantifying the cause–effect relationship among the variables. Based on the fuel cell stack transient model, Shao *et al.* [50] established four neural networks which used different state variables as input after analyzing the changes of various state variables of the system under the stack normal and the hydrogen crossover leakage occurs in the stack. Then the hydrogen crossover leakage in the stack was diagnosed by integrating the output of each neural network. Polverino *et al.* [116] proposed a fault diagnosis method based on the isolated system component sub-models, which can realize the identification of the hydrogen leakage from solid oxide fuel cell stack. Compared with a complete model, the use of isolated sub-models can increase the generation of residuals that diagnostic information extraction can be improved. Lazar *et al.* [117] provide an open-source, dynamic, 1D, PEMFC model, it can provide help for the design of model-based methods or data-driven methods for the diagnosis of hydrogen crossover leakage in the stack.

IV. SUGGESTIONS ON THE SELECTION OF ON-LINE HYDROGEN LEAKAGE DIAGNOSIS METHODS FOR FCVs

In reality, for FCVs application, a fault diagnosis method should be operated real-time, on-line and can work in the dynamic operating conditions of FCVs [118]. Compared to researchers or engineers related to fuel cell system, the danger awareness and protection awareness of FCVs' drivers and passengers is poorer. If a hydrogen safety accident occurs in a FCV, it is possibly to cause injury to the driver, passengers in the accident vehicle and other people, vehicles nearby the accident vehicle. Therefore, in order to prevent hydrogen safety accidents of FCVs as much as possible, a desirable diagnosis method for FCVs hydrogen leakage is required to satisfy the following requirements:

- The method can realize on-line diagnosis that it can work in a parked or moving FCV.
- The method has good real-time performance that it can diagnose hydrogen leakage at the earliest possible moment.
- The method has low calculation complexity that it can run quickly in embedded systems [120], 125].

In terms of on-line diagnosis, based on the relevant literatures and the principles of various hydrogen leakage diagnosis methods. It can be found that the existing methods of the bubble diagnosis, the thermography diagnosis and the crossover current diagnosis are only applicable for the off-line hydrogen leakage diagnosis of fuel cell systems. Although the cathode outlet hydrogen concentration diagnosis has not been used for on-line diagnosis hydrogen leakage in the stack of FCVs in literatures, it seems feasible to measure the hydrogen concentration of the exhaust gas by installing a hydrogen concentration sensor at the end of the stack cathode outlet line in FCVs. Therefore, the feasibility of using the methods base on the cathode outlet hydrogen concentration diagnosis for on-line diagnosis of hydrogen leakage in the stack of FCVs is required to be further validation.

In terms of real-time performance, during the processes of the pressure decay diagnosis and the output voltage diagnosis, a series of operations are required to be performed to ensure that the specific areas of the fuel cell system are sealed or change the operation state of fuel cell system before diagnosing the hydrogen leakage. By comparison, the methods based on the environmental hydrogen concentration diagnosis, the odour diagnosis, the cathode outlet hydrogen concentration diagnosis, the leakage sound diagnosis, and the model-based methods or data-driven methods can diagnose hydrogen leakage merely through the data collected from sensors, special odour or leakage sound. They are exempt from related operations before diagnosing hydrogen leakage. Therefore, their real-time performance are superior to the pressure decay diagnosis and the output voltage diagnosis. However, the odour diagnosis and the leakage sound diagnosis require human olfaction and hearing to recognize hydrogen leakage. They can not be used to diagnose hydrogen leakage when people are far away from the FCVs. In addition, existing odorants would have negative impacts on the performance of fuel cell or hydrogen storage. Therefore, the odour diagnosis and the leakage sound diagnosis have much to be desired for its practicability when applied to diagnose the hydrogen leakage of FCVs.

In terms of the calculation complexity, some fault diagnosis algorithms used to diagnose hydrogen leakage (e.g. stack sparse auto encoder, etc.) are complex. Therefore, they are difficult to run smoothly in normal embedded systems. In addition, for the method of fitting polynomials [106], it requires to fit the cubic polynomials for each cell with the data of hydrogen pressure at the anode inlet and the output voltage of each cell in the stack continuously, and each polynomial contains 12 parameters. Therefore, it may also be difficult to run smoothly in the normal embedded system. Other diagnosis methods do not involve complex calculations. The various PEMFC system hydrogen leakage diagnosis methods are summarized in Table 1.

Based on the review of the existing diagnosis methods for hydrogen leakage in PEMFC systems, the suggestions below are proposed on the selection of on-line hydrogen leakage diagnosis methods for FCVs at the present stage:

1. It is suggested that the methods based on environmental hydrogen concentration diagnosis and pressure decay diagnosis, and model-based methods or data-driven methods are used together for diagnosing the hydrogen leakage from FCVs hydrogen supply system.

The environmental hydrogen concentration diagnosis refers to diagnosing hydrogen leakage by measuring the hydrogen concentration in the environment. It can diagnose hydrogen leakage in the running process of FCVs, and it can diagnose hydrogen leaks with a small flow rate. However, methods based on the environmental hydrogen concentration diagnosis have two major shortcomings. First, they can only diagnose hydrogen leakage when the hydrogen volume concentration in the environment is greater than the minimum volume concentration threshold (e.g., 0.1%). If the sensors' installation positions are far from the leakage location, diagnosis may take a long time; if the hydrogen concentration sensors are deployed in a well-ventilated environment, hydrogen leakage (especially small flow rate) cannot be diagnosed. Second, they cannot locate the leakage area of the hydrogen supply system [9].

In order to make up the shortcoming that the methods based on the environmental hydrogen concentration diagnosis may take a long time for the diagnosis, the model-based methods or data-driven methods can be used as a supplement to the environmental hydrogen concentration diagnosis. The model-based methods or data-driven methods can diagnose hydrogen leakage with system state data collected by relevant sensors and the diagnosis time is constant. If the data acquisition period is short and the calculation complexity of the used fault diagnosis algorithm or strategy is low, the diagnosis time

TABLE 1. Various hydrogen leakage diagnosis methods for the fuel cell system.

Method	On-line diagnosis	Real-time performance	Calculation complexity	Applicable scopes	References
Bubble diagnosis	Not applicable	Not involve (because only applicable for the off-line diagnosis)	Low	Vehicle fuel cell system integration tests and related Test-beds test	[9]
Odour diagnosis	Applicable	Superior to the pressure decay diagnosis and the output voltage diagnosis. (under the premise that the relevant personnel are on site)	Low	Under the premise that the relevant personnel are on site, it can be used to diagnose hydrogen leakage in the process of vehicle fuel cell system integration tests and related Test-beds test, as well as to diagnose hydrogen leakage during FCVs parking or running.	[18-20]
Environmental hydrogen concentration diagnosis	Applicable	Superior to the pressure decay diagnosis and the output voltage diagnosis.	Low	It is widely used for hydrogen leakage diagnosis in workshops, laboratories, hydrogen refueling stations and FCVs.	[24-30]
Leakage sound diagnosis	Applicable	Superior to the pressure decay diagnosis and the output voltage diagnosis. (under the premise that the relevant personnel are on site)	Low	Under the premise that the relevant personnel are on site, it can be used to diagnose hydrogen leakage during FCVs parking or running.	[31]
Cathode outlet hydrogen concentration diagnosis	Need to be further verified	Superior to the pressure decay diagnosis and the output voltage diagnosis.	Low	It can be used to diagnose the hydrogen crossover leakage in the stack during the relevant fuel cell experiments.	[17,93,94]
Thermography diagnosis	Not applicable	Not involve (because only applicable for the off-line diagnosis)	Low	It is suitable for off-line diagnosis of hydrogen crossover leakage in the fuel cell stack and the detection of membrane defects during production or testing of PEMs.	[10,96-98]
Crossover current diagnosis	Not applicable	Not involve (because only applicable for the off-line diagnosis)	Low	It is applicable to the diagnosis of hydrogen crossover leakage in the fuel cell during the manufacturing and testing of fuel cell stacks.	[10,100-103]
Output voltage diagnosis	Applicable	Relatively poorer (except the method of fitting polynomials in [106])	Low (except the method of fitting polynomials in [106])	It can be used for the off-line diagnosis of the hydrogen leakage in the stack (or single cell) during the manufacturing and testing of the fuel cell stack, and it can also be used for the on-line diagnosis of the hydrogen leakage in the stack (or single cell) in the running process of FCVs.	[10,100,104-111]
Pressure decay diagnosis	Applicable	Relatively poorer	Low	It can be used to diagnose hydrogen leakage from hydrogen supply system during the PEMFC system activation, intermittent operation and stoppage in FCVs. In addition, it can also be used to diagnose hydrogen crossover leakage when the PEMFC system in idle-stop mode or during off-line testing of the stack.	[9,10,32-34] [100,112-114]
Gas leakage diagnosis by model-based methods or data-driven methods	Applicable	Superior to the pressure decay diagnosis and the output voltage diagnosis.	Relatively higher	It can be used to diagnose hydrogen leakage during the fuel cell system testing or FCVs running.	[36-50,115,123]

is short. However, due to the presence of sensors measurement noise, the model-based methods or data-driven methods are difficult to diagnose hydrogen leaks with small flow rates. Therefore, it can only be used as a supplement to the methods based on the environmental hydrogen concentration diagnosis.

In addition, in order to make up for the shortcoming that the methods based on the environmental hydrogen concentration diagnosis can not locate the area where the leakage occurs and diagnose hydrogen leakage in a well-ventilated environment, the methods based on pressure decay diagnosis can be used to diagnose hydrogen leakage during the PEMFC system activation, intermittent operation and stoppage in FCVs, because they can locate the leakage area. In addition, the methods based on pressure decay diagnosis can diagnose hydrogen leaks with small flow rates in a well-ventilated or closed environment, but they will take a long diagnosis time. Therefore, the methods based on the pressure decay diagnosis can also only be used as a supplement to the methods based on the environmental hydrogen concentration diagnosis.

2. It is suggested that the model-based methods or data-driven methods and the method based on the output voltage diagnosis proposed in [109] or [110] are used together for the diagnosis of hydrogen crossover leakage in the stack in the running process of FCVs. In addition, at the PEMFC system of FCVs shutdown, the pressure decay diagnosis proposed in [112] is recommended to be applied to diagnose the hydrogen leakage in the stack.

The model-based methods or data-driven methods do not require to perform relevant operations before the diagnosis of the hydrogen leakage. However, due to the presence of sensors measurement noise, these methods are difficult to diagnose the hydrogen leaks with small flow rates in the stack.

The method based on the output voltage diagnosis proposed in [109] or [110] can realize the diagnosis of hydrogen crossover leakage in the stack by detecting the abnormal stack output voltage after performing relevant operations (e.g. reducing the air supply flow rate of the stack cathode or increasing the humidity of the membrane electrode) in the running process of FCVs. To a certain extent, the performed relevant operations can enlarge the effect of hydrogen crossover leakage on the output voltage of the stack. Therefore, compared to the model-based methods or data-driven methods, it is easier to diagnose hydrogen crossover leaks with small flow rates. In addition, the method based on the pressure decay diagnosis described in [112] can be used to diagnose the hydrogen crossover leakage in the stack at the PEMFC system of FCVs shutdown. It can diagnose the hydrogen crossover leaks with small flow rates in the stack by extending the diagnosis time. Therefore, these methods can be used as supplements to the model-based methods or data-driven methods. The suggestions on the selection of on-line hydrogen leakage diagnosis methods for FCVs are summarized in Figure 11.

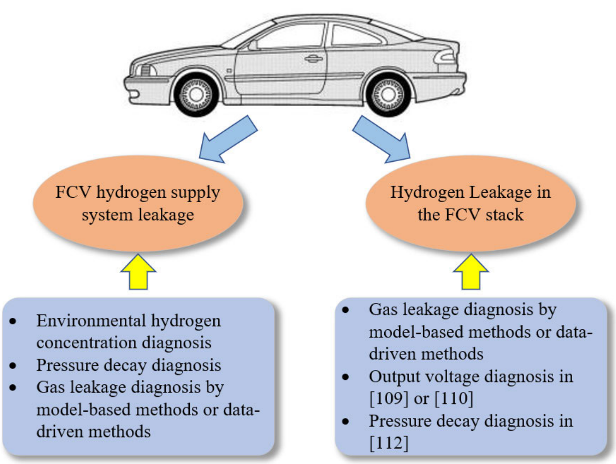

FIGURE 11. Suggestions on the selection of on-line hydrogen leakage diagnosis methods for FCVs.

V. CONCLUSION

In this paper, the existing hydrogen leakage diagnosis methods for the hydrogen supply system and the stack in PEMFC system are reviewed. The basic principles and research status of different hydrogen leakage diagnosis methods are elaborated, and their applicable scopes are summarized. In addition, two suggestions are provided on the selection of on-line hydrogen leakage diagnosis methods for FCVs at the present stage by comparing various methods from three aspects of on-line diagnosis, real-time performance and calculation complexity. One suggestion is that the methods based on environmental hydrogen concentration diagnosis, the pressure decay diagnosis, and the model-based methods or data-driven methods are used together for the diagnosis of hydrogen leakage of FCVs' hydrogen supply system. The other suggestion is that the model-based methods or data-driven methods, the method based on the output voltage diagnosis which are proposed in [109] or [110], and the method based on the pressure decay diagnosis which is proposed in [112] are used together for the diagnosis of hydrogen leakage in the stack.

In the future, the diagnosis methods based on environmental hydrogen concentration and model-based diagnosis methods or data-driven diagnosis methods may play more important roles in on-line hydrogen leakage diagnosis of FCVs. With a decrease in the volume of hydrogen concentration sensors and their cost, it is possible to install more hydrogen concentration sensors in hydrogen FCVs. An increase in the number of hydrogen concentration sensors can greatly reduce the hydrogen leakage diagnosis time, and the leakage area can be estimated from the output values of sensors installed in different locations. In addition, with the improvement of the operational speed of embedded systems and the anti-interference capability of sensors, the diagnosis time of the model-based methods or data-driven methods will be shorter, and they can diagnose hydrogen leakage at a smaller flow rate in the future.

This review can help readers understand the principles and research status of different hydrogen leakage diagnosis methods for PEMFC system, and it can provide important references for researchers and engineers engaging in the related work.

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