Power-to-Hydrogen by Electrolysis in Carbon Neutrality: Technology Overview and Future Development

Zhiyao Zhong, Jiakun Fang, *Senior Member*, *IEEE*, *Senior Member*, *CSEE*, Kewei Hu, Danji Huang, Xiaomeng Ai, *Member*, *IEEE*, Xiaobo Yang, Jinyu Wen, *Member*, *IEEE*, *Fellow*, *CSEE*, Yuan Pan, and Shijie Cheng, *Life Fellow*, *IEEE*, *Fellow*, *CSEE*

***Abstract*—Power-to-hydrogen by electrolysis (PtHE) is a promising technology in the carbon-neutral evolution of energy. PtHE not only contributes to renewable energy integration but also accelerates decarbonization in industrial sectors through green hydrogen production. This paper presents a comprehensive review of PtHE technology. First, technical solutions in PtHE technology are introduced to clarify pros and cons of one another. Besides, the multiphysics coupling and the multi-energy flow are investigated to reveal the insight mechanism during operation of compactly assembled industrial PtHE plants. Then, the development trends of core components in PtHE plants, including electrocatalysts, electrode plates and operation strategy, are reviewed, respectively. Research thrusts needed for PtHE in carbon-neutral transition are also summarized. Finally, three configurations of the PtHE plant in energy system integration are introduced, which can achieve renewable energy integration and efficient energy utilization.**

***Index Terms*—Carbon neutrality, power-to-hydrogen by electrolysis (PtHE), multiphysics coupling, multidisciplinary.**

I. INTRODUCTION

DUE to depletion of fossil fuels and environmental concerns about carbon emissions, a consensus has been reached worldwide that a clean energy carrier is needed to replace fossil fuels [1]. Several countries have announced their policies for a decarbonized society and China set a goal of reaching carbon neutrality by 2060 [2], [3]. For these purposes, evolution of energy mix occurs in the energy system. The primary energy source shifts from fossil fuels to renewable energy, such as wind and solar energy [4]. Penetration of renewable energy in power systems has risen to achieve carbon neutrality. Integration of renewable energy also brings challenges to safety and stability of power systems [5].

Under this circumstance, hydrogen is considered to play an important role in energy transition for distinguished advantages [6]. First, mass energy density of hydrogen is up to 120 MJ/kg, almost triple those of conventional fuels, such as diesel and kerosene [7]. Second, only hydrogen and oxygen are involved in the process of combustion or electrolysis, and they are carbon-free. Therefore, hydrogen can replace fossil fuels as the energy carrier in carbon neutrality. Besides, hydrogen can be utilized in many scenarios, including transportation [8], [9] and industry [10], [11]. In transportation, fuel cell electric vehicles using hydrogen have advantages in long driving distance and fast refueling, compared with battery electric vehicles [12]. Hydrogen is also widely used as a propellant in railway trains [13] and aerospace flights [14]. In industry, utilization of hydrogen is currently dominated by petroleum refining, chemical synthesis and metallurgy production [15], [16]. Petroleum refining requires large demand for hydrogen to remove sulfur, nitrogen and metal impurities. For example, crude oil consumption in Lebanon reaches 150,000 bbl/day, nearly 20,000 Mt/day, which accounts for 65,000 Mt/year of hydrogen [17]. Hydrogen is consumed to synthesize methanol and ammonia for chemical applications, where ammonia is regarded as a hydrogen carrier with low cost and potential for large-scale applications [18]. In metallurgy production, hydrogen is used to reduce iron oxides by producing water [19]. No carbon emission means a sustainable development of the production of iron and steel [20].

As a clean and sustainable energy carrier with various applications, hydrogen has attracted research attention worldwide [21], [22]. Efforts are made to promote rapid development of hydrogen energy in energy transition [23], [24]. A hydrogen supply chain containing production, storage, transmission and application is established [25], [26]. Hydrogen is produced from primary energy sources, such as hydrocarbons, biomass and water, where hydrocarbons are currently dominant [27], [28]. Then, hydrogen is widely compressed in pressure vessels or salt caverns for storage, while other options are rapidly developing, such as liquid hydrogen and metal hydride [29], [30]. Hydrogen stored in tanks is transported by

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Z. Y. Zhong (ORCID: https://orcid.org/0000-0001-9008-4081), J. K. Fang (corresponding author, email: jfa@hust.edu.cn; ORCID: https://orcid.org/0000-0002-8208-5938), K. W. Hu, D. J. Huang, X. M. Ai, J. Y. Wen, Y. Pan, and S. J. Cheng are with the State Key Laboratory of Advanced Electromagnetic Engineering and Technology, Huazhong University of Science and Technology, Wuhan 430074, China.

X. B. Yang is with Hitachi Energy Research, Hitachi Energy (China) Limited, Beijing 100015, China.

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trucks, which can achieve energy transfer across space [31]. In addition, hydrogen transmission is also available in gas pipelines [32]. Fuel cell (FC) is the main application of hydrogen and it can replace the internal combustion engine for sustainable transportation [33], [34]. Besides, FC can generate electrical power to mitigate shortages in power system [35], [36]. Most studies point out hydrogen production is the key step in supply chain [37]. However, producing hydrogen from hydrocarbons will cause massive carbon emissions, which is not compatible with carbon neutrality. Power-to-hydrogen by electrolysis (PtHE) is a clean hydrogen production method and has attracted attention [38]. Although PtHE is cost-competitive with renewable energy in the niche application, it is not competitive for industrial-scale supply [39], [40].

To enhance the techno-economic feasibility of PtHE, its performance should be improved. Improvement in either efficiency or production rate requires multidisciplinary research, including catalyst materials [41], [42], structure designs [43], [44] and operation strategies [45], [46]. From the material perspective, there is a trade-off between catalysis performances and investment cost of materials [47]. Catalysts based on noble metals, such as Pt and Ir, can achieve remarkably low overpotential ($<$300 mV), realizing low energy consumption of hydrogen production. But scarcity and high investment cost of noble metals impede commercial application of PtHE [48]. From the design perspective, the structure of electrode plates can influence the flow field pattern of electrolytes [49]. According to [50], performances of PtHE are improved by uniformly distributed flow field, which can be brought by a well-designed plate. From the operation perspective, PtHE is affected by internal multiphysics based on electrochemistry research [51], [52], when it serves as a flexible load in power system to convert surplus renewable energy into hydrogen [53]. In addition, PtHE can be further applied in the integrated energy system to promote renewable energy integration and energy sector coupling [45], [54].

However, existing reviews of hydrogen energy and PtHE technology are limited to examining individual facets of the technology. An integrated comprehension, which encompasses all elements of PtHE, remains elusive. For example, performances of catalysts are significantly affected by multiphysics coupling, while the scheduling in power system usually considers a single physical process. As a result, comprehensive guidance for PtHE is lacking to coordinate research development in various aspects, such as novel catalysts, high-capacity units and advanced strategies.

To bridge this gap, we present a multidisciplinary overview of PtHE technology, aimed at its rapid development in carbon neutrality. The main contributions of this paper are as follows. First, a comprehensive review of PtHE technology is carried out to highlight advantages and disadvantages of technical solutions with a maturity level higher than TRL4. Secondly, a full understanding on the multiphysics coupling of water electrolysis reaction and multi-energy flow in PtHE plants is established based on industrial processes. Then, research thrusts ranging from fundamental materials and structure design to macroscopical operation strategies are synthesized to promote multidisciplinary research and bridge the current gap. Finally, three configurations of energy system integration with PtHE plants are presented to accommodate various hydrogen applications and facilitate efficient energy utilization. This paper is expected to serve as a leading outline for rapid development of PtHE technology and further contributes to hydrogen energy transition in carbon neutrality.

II. Overview of Power-to-Hydrogen by Electrolysis

As a secondary energy source, there are various methods to obtain hydrogen. These methods are categorized into three groups: grey hydrogen, blue hydrogen and green hydrogen, according to carbon emissions [7]. Grey hydrogen is produced mainly by breaking C-H bonds in hydrocarbons, including partial oxidation (POX) of oil [27], steam methane reforming (SMR) of natural gas [55] and coal gasification [56]. This kind of technology currently accounts for most of hydrogen production worldwide, where natural gas contributes more than 45% of hydrogen [57]. However, breaking C-H bonds to produce hydrogen leads to carbon emissions. For example, there are 10–15 kg CO_2 emissions along with 1 kg hydrogen produced by SMR [58], [59]. As a result, grey hydrogen is regarded as unsustainable, and it should be replaced in carbon neutrality. Blue hydrogen production uses carbon capture and storage (CCS) to reduce carbon emissions from breaking C-H bonds [60], [61]. Consumption of hydrocarbons only increases by 2% with carbon emission dropping by 80% [62].

To achieve carbon-neutral hydrogen production, biomass or water as raw materials is gathering attention [63], [64]. Biohydrogen is a promising technology but needs research to address challenges in large-scale applications [65]. Water splitting breaks O-H bonds to produce hydrogen, including photocatalysis [66], pyrolysis [67] and electrolysis [68], [69]. Through water photocatalysis, chemical energy in hydrogen is converted from photonic energy under low temperature [70], while catalysts currently available have poor stability and high cost [71]. As a thermochemical reaction under high temperature, water pyrolysis suffers from difficulties including efficiency improvement and toxic byproduct treatment [72], [73]. Different from the above processes, water electrolysis, which is also called power-to-hydrogen by electrolysis (PtHE), is a rising technology with demonstration projects and commercial applications worldwide [74]–[76]. More importantly, PtHE with renewable energy as input can achieve a whole carbon-free process from primary energy (wind or solar) to hydrogen energy [77], [78]. It avoids consumption of conventional electrical power generated from fossil fuels and hydrogen produced by this means is called green hydrogen. Although PtHE only supplies 4% of global hydrogen production [57], both academia and industry are currently focusing on PtHE to make efforts for technology development.

In this section, different *technical solutions* for water electrolysis are reviewed to present the pros and cons of each one. An *industrial PtHE plant* is introduced from three aspects: compact electrolyzer, multiphysics process of the reaction and multi-energy flow in the plant.

A. Technical Solutions for Water Electrolysis

Water electrolysis phenomenon was discovered by William Nicholson and Anthony Carlisle in the 1800s [7]. Electrical power from direct current (DC) power supply is converted into chemical energy of hydrogen in the electrolyzer, which is the core component of PtHE plants. As shown in the reaction equation, the water molecule is decomposed under external voltage, while hydrogen and oxygen are produced on the electrodes, respectively.

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2$$

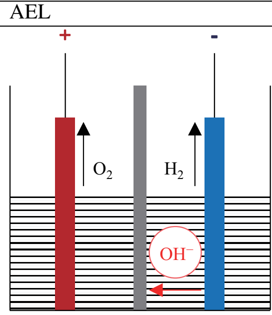
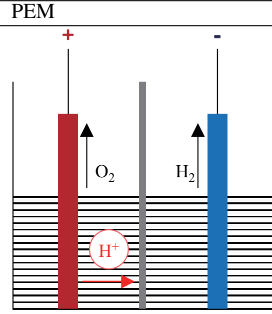
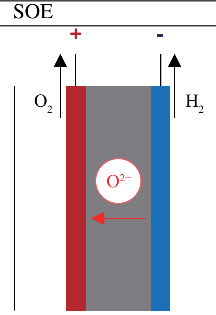
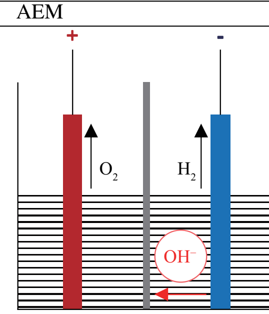
Industrial application of PtHE technology began at the end of the 19th century [79]. Through development over centuries, there are several technical solutions of PtHE technology currently available [69], where alkaline water electrolysis (AEL), proton exchange membrane electrolysis (PEM) and solid oxide electrolysis (SOEC) have technology readiness level (TRL) higher than TRL 4 [80]–[82]. The main differences between each technical solution are charged particles in electrolytes [83], as shown in Table I. Taking AEL as an example, alkaline solutions are used to provide hydroxide particles (OH^-), such as potassium hydroxide (KOH) and sodium hydroxide (NaOH) [38]. Hydrogen evolution reaction (HER) occurs on the cathode connected to the negative terminal of DC power supply, where OH^- and hydrogen are produced. OH^- transported through the diaphragm is consumed on the anode, which is connected to the positive terminal. In the oxygen evolution reaction (OER), oxygen is also produced on the anode [68].

AEL has distinguished advantages in high capacity and low investment cost, which has reached TRL 8–9 representing the maturity [68], [84]. AEL is widely used in commercial applications, where hydrogen production capacity is up to 1500 Nm^3/h [85]. Besides, AEL does not need expensive diaphragms and catalysts [86]. However, the main drawbacks of AEL are low current density and low electrolysis efficiency. This is due to large resistance of the electrolyte and diaphragm in AEL [38]. The partial load range of AEL is also narrow due to hydrogen crossover phenomenon [27], [87]. The fraction of hydrogen on the anode may exceed lower explosion limit (LEL) under a low load rate, resulting in a safety hazard.

As a rapidly developing technical solution in recent years, PEM has high current density, 10000–20000 A/m^2, compared with 2000–4000 A/m^2 of AEL [88]. The proton exchange membrane is quite thin with high proton (H^+) conductivity, leading to low internal resistance. The selective membrane can effectively hinder matter permeation and improve operation range [69]. Besides, pressurized operation is practical for PEM, which has advantages in energy saving of compressors and gas solubility improvement [89]. However, the electrolyte in PEM is acidic with corrosion, therefore, noble metals, usually Pt, serve as the catalyst [90]. Since both the proton exchange membrane and noble metals are expensive, investment cost of PEM is relatively high [86]. As a result, there are fewer commercial applications of PEM than AEL, which is mainly applied in demonstration projects [74], [91].

SOE operates under high temperature in the range of 700–1000 °C [92], making current density and electrolysis efficiency higher than low-temperature PtHE (AEL, PEM). On one hand, high temperature provides a large amount of thermal energy, which reduces demand for electrical power. On the other hand, high temperature makes it easy for activation on the electrode, increasing current density [93]. Reversible operation is another advantage of SOE for switching between electrolyzer mode and fuel cell mode [45]. However, high temperature also brings challenges to SOE. Temperature gradient caused by thermal imbalance may lead to degradation of material performances [41]. Therefore, SOE has a lower TRL

TABLE I
THE COMPARISON BETWEEN FOUR TECHNICAL SOLUTIONS OF PTHE TECHNOLOGY: (A) ALKALINE WATER ELECTROLYSIS (AEL), (B) PROTON EXCHANGE MEMBRANE ELECTROLYSIS (PEM), (C) SOLID OXIDE ELECTROLYSIS (SOE) AND (D) ANION EXCHANGE MEMBRANE ELECTROLYSIS (AEM). (RED: ANODE, BLUE: CATHODE, GREY: DIAPHRAGM)

	AEL	PEM	SOE	AEM
Electrolysis cell				
TRL	TRL 8-9	TRL 8-9	TRL 6-7	TRL 2-4
Charged particles	OH^-	H^+	O^{2-}	OH^-
HER (cathode)	$2H_2O + 2e^- \rightarrow 2OH^- + H_2$	$2H^+ + 2e^- \rightarrow H_2$	$H_2O + 2e^- \rightarrow O^{2-} + H_2$	$2H_2O + 2e^- \rightarrow 2OH^- + H_2$
OER (anode)	$2OH^- - 2e^- \rightarrow H_2O + \frac{1}{2}O_2$	$H_2O - 2e^- \rightarrow 2H^+ + \frac{1}{2}O_2$	$O^{2-} - 2e^- \rightarrow \frac{1}{2}O_2$	$2OH^- - 2e^- \rightarrow H_2O + \frac{1}{2}O_2$
Pros	- high capacity, up to 1500 Nm^3/h; - low investment cost.	- high current density; - pressurized operation.	- high electrolysis efficiency; - reversible operation.	- a combination of AEL and PEM.
Cons	- low current density; - low electrolysis efficiency; - narrow partial load range.	- high investment cost.	- material degradation.	- catalyst stability; - membrane conductivity.

than low-temperature PtHE [94], [95], TRL 6-7, which means it has been validated in the laboratory and there are several demonstration projects around the world [96], [97].

In addition to the above-mentioned three PtHE technologies, a novel technical solution called anion exchange membrane electrolysis (AEM) has been developed rapidly in recent years. AEM can be regarded as a combination of AEL and PEM to achieve high current density and capacity, which uses the anion exchange membrane to effectively transport hydroxide particles (OH^-) [98]. The same alkaline electrolyte is used to avoid high investment cost of catalysts based on noble metals [99]. Despite a promising future, AEM has not reached TRL 4 and it is far from applications [97]. Because AEM still faces technical challenges, such as catalyst stability and membrane conductivity [100].

In summary, each of the above technical solutions has its advantages and disadvantages, as shown in Table I. AEL can be widely applied in large-scale hydrogen production, such as MW and GW-level PtHE plants. PEM is suitable for application in scenarios pursuing high performance, including high current density and electrolysis efficiency. SOE is supposed to be applied in high-temperature conditions for easily accessible thermal energy, such as in nuclear power plants. Besides, efforts are needed to mitigate drawbacks and improve performance of various technical solutions.

B. Industrial PtHE Plant

Despite differences between technical solutions, the industrial PtHE plant has a similar structure. To fully understand the operation principle of industrial plants, the following aspects are reviewed, including *compact structure* of electrolyzers, *multiphysics coupling* of water electrolysis reaction and *multi-energy flow* in PtHE plants.

1) Compact Structure of Electrolyzers

As the core component of the industrial PtHE plant, water electrolysis reaction occurs in the electrolyzer. Unlike the laboratory prototype, the industrial electrolysis cell has a multi-layer structure, consisting of four parts: end plates, flow channels, electrodes (anode and cathode) and the selective diaphragm, as shown in Fig. 1 [7]. The end plate is used to support mechanical stability of electrolysis cells. The electrolyte flows in the channel, contacting the catalyst on electrodes. Then, water electrolysis reaction occurs to produce hydrogen and oxygen. At the same time, charged particles are generated or consumed on electrodes, requiring transportation across the selective diaphragm. The move of charged particles forms the current circuit of electrolysis cells along with connection between electrodes and DC power supply [38]. Besides, the flow channel in cells has a narrow gap, causing a lower resistance and higher current density than laboratory prototypes [85]. In industrial electrolyzers, electrolysis cells are stacked to expand capacity, as shown in Fig. 1, where the number of cells is up to several hundred in limited space [101]. The electrolyzer has a common inlet of the electrolyte, which is subsequently supplied to each electrolysis cell. After water electrolysis reaction in the cell, produced hydrogen and oxygen exit from the electrolyzer, respectively.

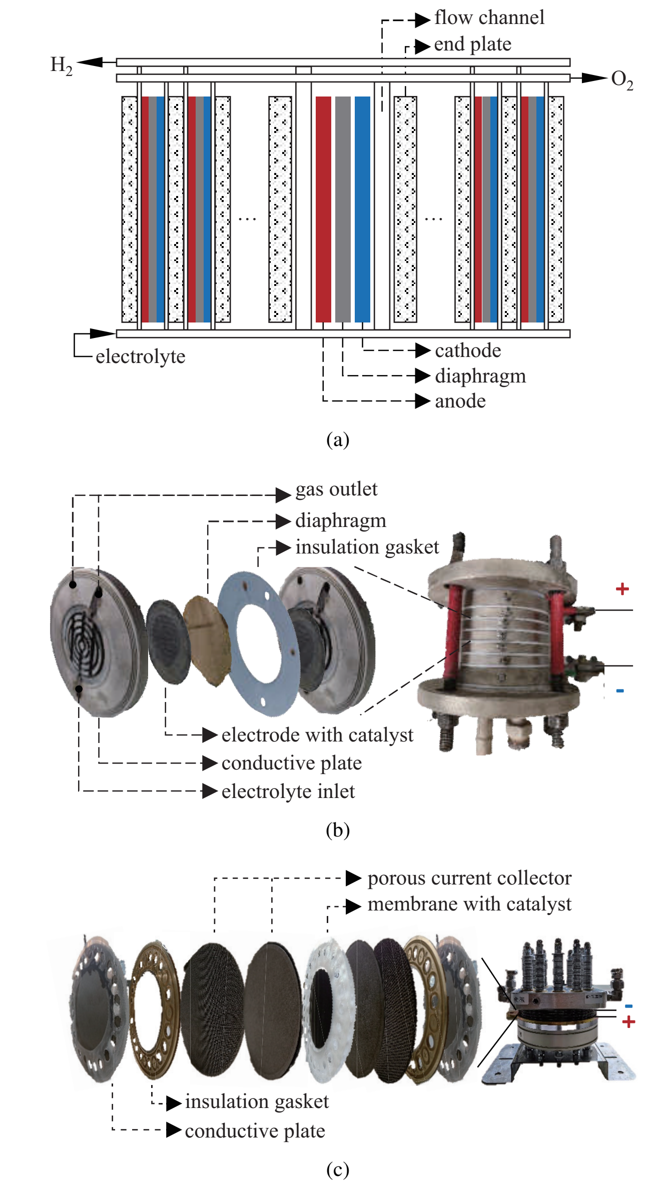


Fig. 1. The compact structure of industrial electrolyzers. (a) Multi-layer electrolysis cells and the stacked electrolyzer. (b) The construction of industrial electrolyzers using AEL. (c) The construction of industrial electrolyzers using PEM.

2) Multiphysics Coupling of Water Electrolysis Reaction

In electrochemistry research, water electrolysis and its inverse reaction, hydrogen fuel cell, have been the focus [102]. Research shows performances are affected by several physical processes [83], [103]. Involved processes include electrochemical reaction on the electrode surface [104], mass transfer of reactants and products [105], fluid mechanics in the flow channel [106] and thermodynamics of the electrolyzer [107]. These processes couple with each other through specific physical quantities to form multiphysics coupling [83], as shown in Fig. 2.

The main physical process is electrochemical reaction expressed by current density field. In the field, current density and electrical potential in the electrolyzer are determined by current conservation and Ohm's law [108], referring to hydrogen production rate and energy consumption. Other processes,

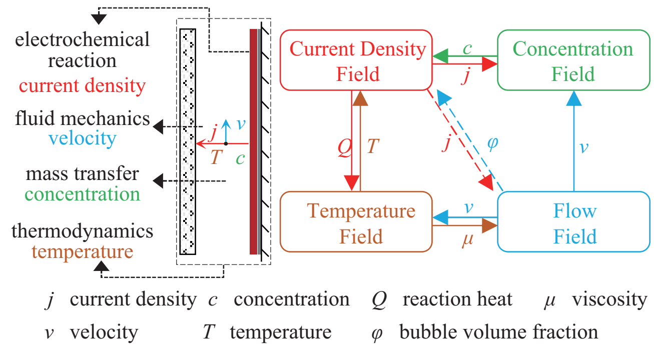


Fig. 2. Multiphysics coupling of water electrolysis (PtHE).

including mass transfer, fluid mechanics and thermodynamics, are represented by concentration field [109], flow field [110] and temperature field [107], respectively. Distribution of physical quantities in each field is determined by mass, momentum and energy conservation in electrolyzers.

Each field couples with the other one to affect physical quantity distribution [103]. For example, the electrical potential on the electrode is influenced by substance concentration and electrolyte temperature [104], according to Nernst equation. Besides, there is gas-liquid diphasic flow in low-temperature PtHE (AEL and PEM) [111], where the gas bubble may accumulate on the electrode surface to reduce effective area and hinder reaction, called bubble effect [112]. Meanwhile, the change rate of substance concentration or bubble volume fraction, if diphasic flow exists, is proportional to current density, according to Faraday's law [113]. The convection process in mass and energy conservation are both affected by flow velocity, coupling flow field with concentration field and temperature field [114].

Currently, research is about multiphysics coupling in water electrolysis. In conclusion, the internal multiphysics coupling has been attracting research attention, which is significant for PtHE plants. However, the following research points need to be considered. On the one hand, 3D multiphysics simulation at the industrial level is urgently needed, especially low-temperature PtHE. Partial approximation of PDEs and joint simulation based on CFD software can accelerate computation [131], [132]. On the other hand, existing comparison of technical solutions lacks consideration of multiphysics coupling [83]. The difference between solutions needs to be explained by respective multiphysics coupling, in favor of application in various scenarios.

3) Multi-energy Flow in PtHE Plants

The industrial PtHE plant also has several auxiliary components, including DC power supply, circulating pump, heat exchanger and gas-liquid separators [68], as shown in Fig. 3. First, the electrolyte is added to the PtHE plant and driven by the circulating pump. Then, the temperature of electrolytes is maintained in the preset range by the heat exchanger for high reactivity and gas purity [101]. Finally, water electrolysis reaction occurs in the electrolyzer under electrical power input, producing hydrogen and oxygen. Through the separator, pure hydrogen and oxygen are obtained with the remaining electrolyte flowing back to the pump for the next cycle [7].

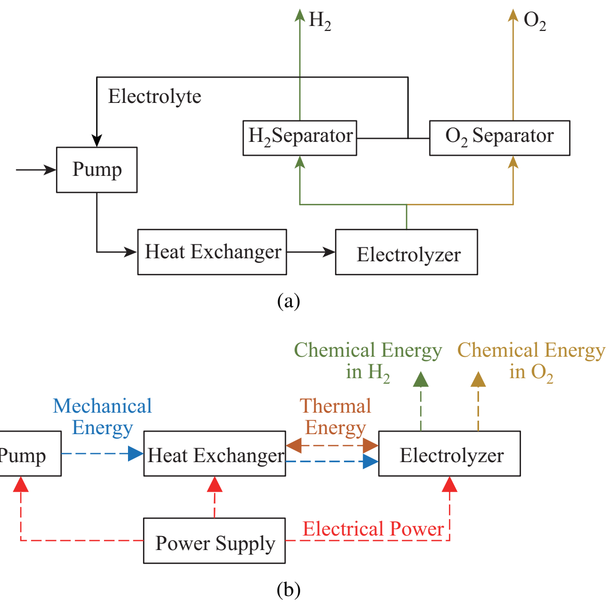


Fig. 3. Matter and energy flow in the industrial PtHE plant. (a) The matter flow with circulating electrolyte. (b) The multi-energy flow including electrical power, thermal energy and mechanical energy.

Energy consumption of internal components in PtHE plants is provided by DC power supply. On the one hand, the electrolyzer requires electrical power from DC power supply. On the other hand, mechanical energy and thermal energy are needed for circulating flow and temperature maintenance of electrolytes. They are converted from electrical power in the pump and heat exchanger, respectively [46]. Therefore, there is a multi-energy flow during water electrolysis reaction, where mechanical energy, thermal energy and electrical power are gathered and converted into chemical energy. Each form of energy flow has an impact on performance of PtHE plants, such as hydrogen production rate and energy conversion efficiency [133].

Mechanical energy determines flow velocity of electrolytes. According to the above multiphysics coupling, fast flow velocity guarantees a sufficient mass convection process. It ensures a large number of reactants in water electrolysis reaction [107], [109]. Besides, fast flow velocity can relieve the bubble effect caused by diphasic flow in low-temperature PtHE [46]. As a result, electrical potential under the same current density decreases and electrolysis efficiency of the electrolyzer is improved [113]. However, massive electrical power is required for the pump, which makes total energy consumption increase and energy conversion efficiency of PtHE plants decrease. Meanwhile, a large amount of thermal energy is needed for SOE to heat the electrolyte, reaching the operating temperature [83]. For low-temperature PtHE, the reaction is exothermic and electrolyte temperature can be self-sustained. However, temperature may exceed the safety limit in some cases, requiring cooling of electrolytes [103]. Both heating of SOE and cooling of low-temperature PtHE can be achieved with electrical power consumption in the heat exchanger. Similar to the pump, energy conversion efficiency of PtHE plants may decrease due to extra consumption [134]. Electrical power supplied to electrolyzers can be expressed by operation voltage. Increasing operation voltage can obtain high hydrogen production rate of the PtHE plant, while electrolysis efficiency decreases [133].

In summary, each form of energy can improve performance to some extent, while it may harm energy conversion efficiency of PtHE plants. With a given electrical power of power supply, the key issue of high energy conversion efficiency is to optimize distribution of different forms in the multi-energy flow. Mechanical energy and thermal energy in the industrial PtHE plant are converted from electrical power in auxiliary components. Therefore, high efficiency is determined by distribution of electrical power from DC power supply, which requires coordination between the electrolyzer, circulating pump and heat exchanger during operation.

III. Development Trends and Research Thrusts

In carbon neutrality, green hydrogen consuming renewable energy has a promising future to replace grey hydrogen and become the mainstream [135]. Green hydrogen can address challenges brought by high penetration of renewable energy and accelerate decarbonization in various energy sectors, while it is currently cost-competitive with the cost dropping in renewable energy and PtHE plants [136]. Despite the promising future, the natural characteristics of renewable energy, such as uncertainty and intermittency, bring power fluctuations to the PtHE plant, which may make operation of PtHE plants different [7], [45]. Therefore, adaptability to power fluctuations from renewable energy is a significant indicator for performance evaluation of PtHE plants, the same as hydrogen production rate and energy conversion efficiency. In this section, the effect of the *electrocatalyst*, *electrode plate* and *operation strategy* on performance of PtHE plants are reviewed to summarize the development trend. Corresponding research thrusts are prospected to achieve high performance of the PtHE plant in carbon neutrality.

A. Advanced Electrocatalyst for Water Electrolysis

In water electrolysis reaction, the electrocatalyst is used to activate electrodes under external voltage [7]. External voltage relies on activation overpotential, which is determined by what kind of electrocatalysts is used. Besides, electrocatalysts should maintain their performances during long-term operation and be easy to manufacture for commercial applications of PtHE plants [47], [137]. Conventionally, noble metals, such as platinum (Pt), iridium (Ir) and silver (Ag), are used as the electrocatalyst in PtHE plants for their lower overpotential and higher stability than other catalysts [42]. However, scarcity of noble metals greatly hinders large-scale application [138]. Therefore, it is necessary to develop alternatives to noble metals as the electrocatalyst in PtHE plants.

Extensive research efforts have been devoted to a catalyst from a material perspective regarding different technical solutions of PtHE. For AEL, several transition metals, including iron (Fe), manganese (Mn), nickel (Ni), and their oxides are used to replace noble metals for both HER and OER [48], [139]. There are several strategies for the design of transition metal-based catalysts [140]. For example, an electrocatalyst called NiFe LDH with nanopore structure is proposed to enhance activity of OER [141]. Through these efforts, performances of electrocatalysts based on the transition metal are close to or even exceed noble metals in a certain dimension, such as low overpotential and high stability [142]–[144]. Besides, transition metals are abundant on earth, so they are easy to obtain, which is the key reason AEL is widely applicated in projects worldwide [145]. For PEM, internal voltage loss is low, which is at the cost of expensive catalysts. Currently, it is difficult to develop low-cost alternatives [146]. The main reason is PEM has acidic electrolytes and very few metals are stable enough in this environment, such as Au and Ir [147], [148]. Although other metals cannot be used in PEM, they can serve as supporting materials to reduce the amount and hence the cost of electrocatalysts [90]. For SOE, the cathode plays a crucial role, where steam electrolysis occurs with a gas mixture of H_2, CO_2 and CH_4 [149]. A global effort is needed to find suitable cathode electrocatalysts, obtaining high-purity hydrogen for SOE [150]. Besides, key issues about stability under high temperature cannot be ignored [47]. The electrocatalyst that can endure temperature gradients and performance degradations needs to be developed urgently, especially under thermal imbalance [41].

In summary, development of electrocatalysts in PtHE plants has a common goal for efficient hydrogen production, and each technical solution has its obstacles to achieve this goal. However, green hydrogen production is the mainstream in carbon neutrality [40], where renewable energy brings another challenge for the electrocatalyst. It requires the effect of power fluctuations on electrocatalysts, especially the lifetime of electrolyzers, which is hardly mentioned in existing research.

B. Structure Design of Electrode Plate

The electrode plate structure determines the flow channel of electrolytes, where electrolytes contact with catalysts and the reaction occurs [49]. On the one hand, a well-designed structure ensures an adequate supply of reactants [151]. On the other hand, a uniform flow field determined by the plate structure can avoid bubble accumulation on electrodes [152]. As a result, hydrogen production rate and energy conversion efficiency of PtHE plants can be improved. Generally, the structure of electrode plates is divided into parallel configurations and serpentine configurations [50]. Results show the serpentine flow field has higher current density than the parallel one. Channel dimensions, such as the width and depth, also affect characteristics of the flow field and further performance of PtHE plants [153].

Although efforts have been devoted to this field, research thrusts remain. First, a quantitative theory for the structure design of electrode plates in PtHE plants is still lacking. Currently, comparison between different design options is necessary to select the optimal one [43], which requires extensive simulations and experiments. Second, a simplified CFD model is needed to represent the flow field during structure designs [50]. Because the narrow flow channel with a high aspect ratio in industrial PtHE plants leads to a heavy computation burden of existing CFD solving [44], [152]. Finally, current structure designs are only intended for fixed power operation condition. The effect of power fluctuations on structure design of electrode plates is still missing, which is similar to electrocatalysts.

C. Operation Strategy Under Power Fluctuations

The operation strategy of PtHE plants needs to determine an operation point referring to high hydrogen production rate and energy conversion efficiency [7]. Generally, operation point relies on several parts, such as electrical power supply, heat management, gas-liquid separation, pressure and flow rate control [154]–[156]. For example, heat management is vital for SOE to obtain high-temperature steam as input [157], [158], while gas-liquid separation can ensure high-purity gas generation in low-temperature PtHE [159]. Besides, the operation range of AEL can be extended by control of pressure and flow rate [160]. Obviously, there are various studies on operation strategies to improve performances [161], [162], among which industrial processes involving electrical power, thermal energy and mechanical energy are dominant, as shown in Fig. 3. Coordination of multi-energy flow can be regarded as distribution of electrical power from power system in most industrial PtHE plants [163]. Therefore, the scheduling of PtHE plants in power systems is the main focus of this paper. Moreover, green hydrogen production, which takes renewable energy as input, brings power fluctuations to the PtHE plant. Therefore, the operation strategy of PtHE plants needs to adjust their operation points in real-time, improving adaptability to renewable energy [45].

By coordinating the PtHE plant with other flexible sources, such as energy storage and demand response, renewable energy integration and economic operation of power systems can be achieved [164]–[166]. For instance, a renewable-electrolysis system is formed, where the PtHE plant is directly connected to wind farms or photovoltaic stations [167], [168]. Battery energy storage is usually introduced in the system to assist in suppressing power fluctuations, realizing maximum hydrogen production [169] and grid auxiliary services [170] with investment cost increasing. The PtHE plant, which is a significant source of hydrogen, also serves as a coupling unit between different energy sectors to establish the integrated energy system (IES) [171]–[173]. Each energy sector in IES has the ability of flexible operation, such as compression of natural gas in pipelines [174]. Energy allocation in IES can be coordinated through these flexible sources and coupling units, such as PtHE plants, aimed at large-scale renewable energy integration and efficient energy utilization [175]–[177].

However, the above studies are still insufficient for application of PtHE plants in carbon neutrality. The electrolyzer or PtHE plant is mostly modeled to provide computational conveniences, such as an energy conversion unit with constant efficiency [166] and a nonlinear electrical circuit [170]. Further, a few studies consider the internal physical process during scheduling [178]. For instance, scheduling of PtHE plants in [179] considers thermodynamics of electrolytes. As an important indicator, temperature can represent performance of PtHE plants based on kinetics and be used as a criterion to start up and shut down the electrolyzer [180]. Besides, the crossover phenomenon caused by fluid mechanics is considered to prevent gas impurity, especially on the anode [181], [182]. In these models, coupling between different physical processes, also called the multiphysics coupling, is hardly considered. Each process has its time scale and ignoring the coupling cannot reflect multi-time-scale dynamic behavior. This may lead to a lack of precision in the characteristic analysis of PtHE plants under power fluctuations. Besides, there are few experiments on the PtHE plant under power fluctuations [183]. The result shows fluctuation with appropriate amplitudes and frequencies can improve hydrogen production rate, while energy conversion efficiency slightly decreases [184]. To fully understand this phenomenon, the interaction between PtHE plants (the internal multiphysics coupling) and renewable energy (power fluctuations) is an urgent research thrust, which needs in-depth study.

In conclusion, current operation strategies of the PtHE plant hardly consider thermal inertia of electrolytes and fluid dynamics inside electrolyzers, which significantly affects its performance. Therefore, a dynamic model of PtHE plants considering the internal multiphysics coupling is significant for application in carbon neutrality, which can represent characteristics under power fluctuations. In addition, the operation strategy needs the ability of real-time decision-making to adapt to varying conditions brought by high penetration of renewable energy. As a real-time method, model predictive control (MPC) is widely used, especially in the energy storage system [185], [186], while it cannot guarantee global optimality [187]. Approximate dynamic programming (ADP) adopts value function approximation (VFA) and so on [188] for solving Bellman's equation to avoid the curse of dimensionality in dynamic programming (DP) [189]. It has been used in the real-time operation of BES and IES, where optimality is improved by adequate training [190]. By embedding the real-time method and the dynamic model into the operation strategy of PtHE plants, efficient hydrogen production under renewable energy can be achieved. PtHE plant can participate in power system and IES to improve operation flexibility.

D. Brief Summary

In this section, a review of the electrocatalyst, electrode plate and operation strategy in industrial PtHE plants is presented, as summarized in Table II. Research about electrocatalysts and electrode plates, which mainly focuses on micro level with a scale of m and nm, mostly ignores the effect of power fluctuations. On the contrary, operation strategy, which focuses on macro level with a scale of km, less considers internal multiphysics coupling. Obviously, there is a research gap between the micro level and the macro level, where studies at each level are independent, lacking a unified consideration. Therefore, multidisciplinary research on the industrial PtHE plant is needed. The key to this research is interaction between the multiphysics coupling at the micro level and power fluctuations at the macro level. Then, the PtHE plant with distinguished performances can be obtained, promoting technology development in carbon neutrality.

IV. Energy System Integration with PtHE

Energy sectors including electrical power system, natural gas system and heat transfer system, are integrated to form IES in carbon neutrality, which can achieve efficient energy utilization. Meanwhile, hydrogen produced by PtHE plants can be utilized in several sectors, meaning PtHE has a promising future in energy system integration. In this section, three configurations of energy system integration with PtHE plants are introduced. Due to high energy density, hydrogen is widely used as fuel on the demand side and in seasonal energy storage. As a result, among these configurations, the first one is related to hydrogen fuel transportation and the remaining two are related to hydrogen energy storage. Besides, research thrusts are summarized for development of these configurations and IES in carbon neutrality.

A. Integrated Gas and Power System with Hydrogen Blended

The PtHE plant using renewable energy can obtain hydrogen with no carbon emissions, which is a distinguished fuel on the demand side. Among various methods, blending in natural gas pipelines can achieve large-scale transmission and storage and avoid costly construction of hydrogen pipelines [191], [192]. Integrated gas and power system with hydrogen blended is established, which will be the main form of energy systems in carbon neutrality [193]. There are several demonstration projects worldwide, especially in Europe, to conduct research on mixture of hydrogen and natural gas [194]. Basic information and features of these projects are summarized, as shown in Table III. As a coupling unit, the operation of PtHE plants can greatly affect characteristics of the integrated system [195].

Research on integrated gas and power system started early and there has been a close relationship between the adequacy of natural gas systems and the reliability of electrical power systems [204]. By introducing PtHE plants, hydrogen blending in natural gas pipelines becomes the research topic, which has significant impact on operation of integrated gas and power system [205], [206]. Most studies adopt Weymouth formulation to describe steady-state energy flow in pipelines [207], [208]. A few studies consider different response times of gas transmission system and electrical power system to avoid the phenomenon energy flow in pipelines may not reach a steady state during the dispatch period [174]. Dynamic gas diffusion process in the pipeline is described by Naiver-Stokes equations [209].

According to demonstration projects worldwide, there may be a safety hazard of hydrogen blending, which is closely related to hydrogen volume fraction in the pipeline [210]. However, the fraction limit in the existing studies is based on average value of the whole pipeline network, lacking a dynamic distribution of hydrogen in each pipeline. Thus, a dynamic safety evaluation of hydrogen blending is missing, causing a threat to operation of gas transmission system [32]. Besides, gas consumers on the demand side, such as gas turbine, have various requirements for fuel, including heat value

TABLE II
THE SUMMARY OF THE ELECTROCATALYST, ELECTRODE PLATE AND OPERATION STRATEGY IN INDUSTRIAL PTHE PLANTS

Electrocatalyst (nm scale)	Electrode plate (m scale)	Operation strategy (km scale)
key issues - low activation overpotential; - low investment cost; - long lifetime.	key issues a well-designed structure for: - an adequate supply of reactants; - a uniform flow field to weaken the bubble effect.	key issues achieve renewable energy integration and efficient energy utilization as: - a flexible load in power system; - a coupling unit in IES.
main challenges - high cost of metals stable in acid (PEM); - poor lifetime under temperature gradient (SOE); - lack of the effect from power fluctuations.	main challenges - lack of a quantitative theory for the design; - heavy computation burden in CFD solving; - only intended for the fixed operation condition.	main challenges - less consideration on multi-time-scale dynamic behaviors caused by internal multiphysics coupling.

TABLE III
DEMONSTRATION PROJECTS OF INTEGRATED GAS AND POWER SYSTEM WITH HYDROGEN BLENDED [194], [196]–[203]

Project	Country/Region	Timeline	Features
NATURLHY	Europe	2004–2009	First demonstration project worldwide; Systematical study on the impact of hydrogen blended; Publish the equipment aging and repair methods; Improve standards and criterion for gas mixture.
HyBlend	USA	2021–2023	Develop public tools that assess the risks of blending; Develop a tool that evaluates the opportunities and costs; Analyze life-cycle greenhouse gas and pollutant emissions.
HyDeploy	UK	2017–2025	Inject hydrogen into both gas distribution and transmission network; Explore feasibility of injection without equipment modifications; Reach 20% blended hydrogen in 2019.
HyNet	UK	–	The infrastructure to produce, transport and store low carbon hydrogen; Both upgrade existing infrastructure and develop new infrastructure; Over 40 organisations sign up to decarbonise through HyNet.
H2HoWi	Germany	2020–2023	Add hydrogen to natural gas in the distribution networks; Disconnect a medium-pressure gas pipeline and connect to a storage facility.
H2-PIMS	Germany	–	Develop PIMS for the transport of hydrogen mixtures; Develop the conversion methods of existing natural gas infrastructures; A safety concept for long-distance transport; Recommend standards for the infrastructure operation.
GRHYD	France	2014–2021	Reach 20% blended hydrogen.
Sustainable Ameland	Netherlands	–	Blend hydrogen into the natural gas distribution network for residential use; No combustion problems under 20% blended hydrogen.

and volume fraction of hydrogen [54], [211]. In conclusion, hydrogen injection brings constraints to gas pipelines and gas consumers, which should be considered in operation of integrated gas and power with hydrogen blended. Then, energy demand of electrical or gas consumers is satisfied, while transmission safety is ensured. Furthermore, high penetration of renewable energy can be consumed by coordinated operation of the integrated system.

B. Combined Heat and Hydrogen Storage Station

The PtHE plant can also serve as the charging unit of hydrogen storage system (HSS), along with the storage unit (hydrogen tanks) and the discharging unit (hydrogen fuel cells or hydrogen turbines). Due to high energy density of hydrogen, HSS is regarded as a seasonal energy storage option in power systems [212]. However, HSS faces the dilemma of cycle energy efficiency, which is lower than common energy storage, such as battery energy storage [213]. On one hand, even if SOE has highest theoretical efficiency among various technical solutions [38], [134], heating for high-temperature steam may lead to low efficiency as the charging unit [83]. On the other hand, overall generation efficiency of hydrogen turbines is less than 60%, considering combined cycling technology of the exhaust steam [214]. As a result, HSS is seldom used to realize energy shifting.

To solve this problem, an idea for full utilization of exhaust steam from hydrogen turbines is proposed, where large heat demand is satisfied to maintain high temperature in SOE and cycle energy efficiency of HSS is improved. Based on this idea, a power plant named combined heat and hydrogen storage station is proposed, as shown in Fig. 4, consisting of the PtHE plant using SOE, gas storage tanks, combined cycling generation unit and thermal storage.

In the station, components interact with each other, which forms an energy cycle with electrical power, hydrogen energy and thermal energy. During the charging process of the station, SOE converts surplus electrical power into chemical energy to produce hydrogen and oxygen under high temperature [7]. The produced gas is compressed in hydrogen and oxygen storage tanks, respectively. The discharging process aims at meeting shortage of electrical power, which is mainly realized by the combined cycling generation unit. First, hydrogen and oxygen in the tanks are supplied to hydrogen turbines for combustion, driving generators for electrical power generation [215]. Then, high-temperature exhaust steam from hydrogen turbines can be fed into the steam turbine for extra electricity generation. At the same time, steam can also be stored in thermal storage, avoiding waste of thermal energy [216]. Thermal storage, where phase change materials (PCMs) act as storage media [217], provides steam with high temperature (500°C–600°C) to the PtHE plant and may satisfy heat demand. Then, temperature in SOE is maintained, which ensures high electrolysis efficiency. Coupling between power system and heat transfer system is further enhanced. In a word, exhaust steam of hydrogen turbines is fully utilized by coordination between internal components, including the PtHE plant, generation

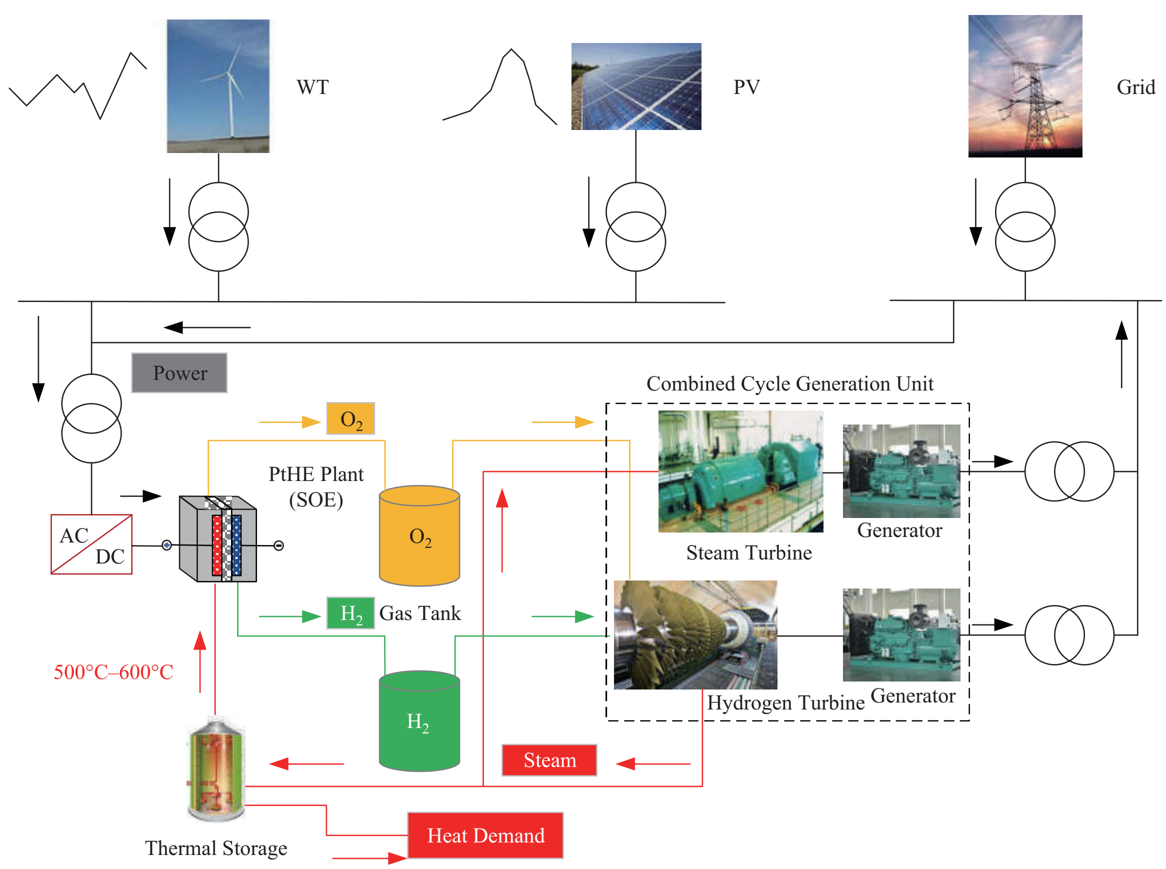


Fig. 4. Combined heat and hydrogen storage station.

unit and thermal storage, improving the cycle efficiency of combined heat and hydrogen storage station.

To participate in auxiliary service in power systems, including renewable energy integration, frequency regulation and flexibility improvement, the following research thrusts in the proposed station need to be studied. First, the internal components require technical modification to accommodate each other, such as pure oxygen combustion in hydrogen turbines [218] and cost-effective materials for high-temperature thermal storage [219], [220]. Second, multi-time-scale dynamic process in the station, including the multiphysics coupling in PtHE plants, electromagnetic transient and electromechanical transient in generators, as well as thermal dynamics in thermal storage, need in-depth analysis. Finally, an optimal operation and control strategy is needed to fully coordinate hydrogen storage and thermal storage, achieving seasonal energy storage in power systems with high penetration of renewable energy.

C. Liquid Hydrogen Coupled Superconducting Magnetic Energy Storage (LIQHYSMES)

In addition to low cycle energy efficiency, HSS still has a long response time compared with other energy storage systems, lacking the ability of instantaneous power support for frequency stability [221]–[223]. To address this problem, a hybrid energy storage system called liquid hydrogen coupled superconducting magnetic energy storage (LIQHYSMES) is proposed [224], which combines fast response of SMES with high capacity of HSS. Thus, power fluctuations with different time scales from seconds to days and months can be suppressed. In LIQHYSMES, liquid hydrogen acts as both the energy carrier and coolant for superconductors, avoiding additional cooling equipment for low investment cost [225].

LIQHYSMES is composed of three main parts: power conversion and control (PCC) unit, electricity-hydrogen bidirectional conversion (E-HBC) unit and multi-energy storage (MES) unit [226], [227]. There is an in-depth coupling of substances and energy in these units of LIQHYSMES, as shown in Fig. 5. During the charging process, the PtHE plant in E-HBC unit consumes electrical power to produce hydrogen. Then, gaseous hydrogen transforms into liquid hydrogen through the hydrogen phase change system, which is stored in LH_2 tank of MES unit. On the contrary, liquid hydrogen in the tank transforms into gaseous hydrogen for the discharging process of LIQHYSMES. Subsequently, electrical power is generated from hydrogen combustion in E-HBC unit to mitigate the shortage, which is achieved by the fuel cell or hydrogen turbine. Meanwhile, superconducting magnetic energy storage (SMES) of MES unit can smooth rapidly changing power [212], [228]. During the charging and discharging process, temperature of superconductors is maintained by a cooling system based on liquid hydrogen, achieving zero resistance and no heat loss [229]. Because the superconductor has a higher stability margin and critical current in the temperature region of liquid hydrogen than of liquid helium [230], [231]. In PCC unit, the multi-port power electronic converter is designed for distribution of electrical

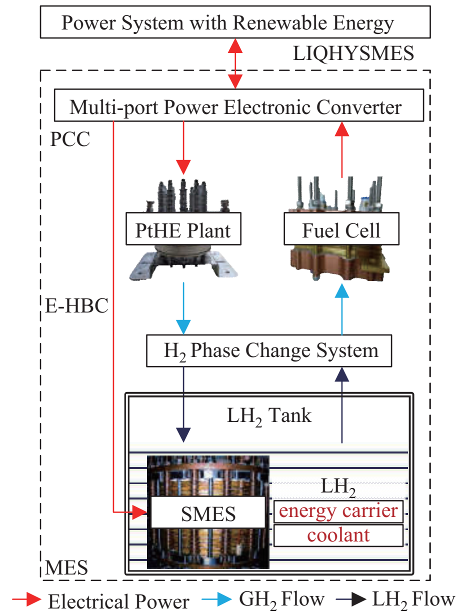


Fig. 5. Liquid hydrogen coupled superconducting magnetic energy storage (GH_2: gaseous hydrogen, LH_2: liquid hydrogen).

power between the internal units of LIQHYSMES and power system.

Currently, research on LIQHYSMES is still limited in the feasibility analysis through simulation [227], [232], where HSS and SMES are simply superimposed. Therefore, an in-depth study considering substance and energy coupling is urgently needed for the operation principle of LIQHYSMES. A design approach is also needed to determine power and energy capacity, such as dimension match of LH2 tank and superconductor for compactness. Then, various forms of energy in LIQHYSMES should be coordinated to achieve efficient energy conversion and storage. Combined with the above studies, a laboratory prototype of LIQHYSMES can be developed. LIQHYSMES can be an energy storage solution for high penetration of renewable energy in power systems. Furthermore, it can also be an energy router of IES to make full use of hydrogen in carbon neutrality.

V. Conclusion

In this paper, a comprehensive review of power-to-hydrogen by electrolysis (PtHE) in carbon neutrality is presented. First, different technical solutions of PtHE technology are introduced. Each solution has advantages and disadvantages, which should be considered in selection of their application scenarios. Second, based on the compact structure of electrolyzers, the multiphysics coupling of water electrolysis reaction and the energy flow in industrial PtHE plants are introduced. For development in carbon neutrality, the electrocatalyst, electrode plate and operation strategy of the PtHE plant are reviewed, respectively. Green hydrogen produced by PtHE with renewable energy will be the mainstream, where efforts are made to develop PtHE plants with high hydrogen production rate and energy conversion efficiency. However, there is a gap in the research on PtHE between micro level and macro level. Multidisciplinary research is needed to bridge the gap by studying interaction between internal multiphysics coupling and power fluctuations from renewable energy. Finally, three configurations of the PtHE plant are introduced, including integrated gas and power system with hydrogen blended, combined heat and hydrogen storage station and liquid hydrogen coupled superconducting magnetic energy storage (LIQHYSMES). These plants can not only assist renewable energy integration in power systems but also achieve efficient energy utilization in IES.

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Zhiyao Zhong received the B.Eng. degree in Huazhong University of Science and Technology (HUST), Wuhan, China, in 2020. He is currently pursuing the Ph.D. degree in Electrical Engineering with the Huazhong University of Science and Technology, Wuhan, China. His research interests include the optimal dynamic control of power-to-hydrogen (PtH) plants and the optimization of power system operation with PtH.

Jiakun Fang (SM'19) received the B.Sc. and Ph.D. degrees from Huazhong University of Science and Technology (HUST), Wuhan, China, in 2007 and 2012, respectively. From 2012 to 2019, he was with the Department of Energy Technology, Aalborg University, Aalborg, Denmark. He is currently a Professor with the School of Electrical and Electronics Engineering, Huazhong University of Science and Technology. His research interests include the optimization of integrated energy transmission systems for electricity, refined oil and natural gas, power-to-hydrogen and its applications, and partial differential constrained optimization.

Kewei Hu received the B.Eng. degree in Electrical Engineering from Huazhong University of Science and Technology (HUST), Wuhan, China, in 2020. He is currently pursuing the Ph.D. degree in Electrical Engineering with the Huazhong University of Science and Technology, Wuhan, China. His research interests include the multiphysics modeling and optimization of the power-to-hydrogen electrolyzers.

Danji Huang received the B.Eng. degree in the Huazhong University of Science and Technology (HUST), Wuhan, China, in 2017. He is currently pursuing the Ph.D. degree in Electrical Engineering with the Huazhong University of Science and Technology, Wuhan, China. His area of research is centered on the multiphysics modeling and optimal design of water electrolyzer, with a focus on its large-scale implementation in energy systems.

Xiaomeng Ai (M'17) received the B.Eng. degree in Mathematics and Applied Mathematics and the Ph.D. degree in Electrical Engineering from Huazhong University of Science and Technology (HUST), Wuhan, China, in 2008 and 2014 respectively. He is currently an Associate Professor with HUST. His research interests include robust optimization theory in power system, renewable energy integration, and integrated energy market.

Xiaobo Yang recived the M.S. and Ph.D. degrees in Power Electronics from the Department of Electrical Engineering, Yanshan University, Qinghuangdao, China, in 2003 and 2006, respectively. He joined in ABB Corporate Research Center, Beijing, as Research Scientist in 2007, and had been Corporate Research Fellow in 2018. Since 2019, he has been with Hitachi Energy, where he is currently Center Manager of Hitachi Energy Research China. His research interests include power electronics converter topology and control, HVDC system, FACTS, energy storage system, green hydrogen, and renewable energy generation.

Jinyu Wen (M'10) received the B.S. and Ph.D. degrees in Electrical Engineering from Huazhong University of Science and Technology (HUST), Wuhan, China, in 1992 and 1998, respectively. He was a Visiting Student from 1996 to 1997 and Research Fellow from 2002 to 2003 all at the University of Liverpool, Liverpool, UK, and a Senior Visiting Researcher at the University of Texas at Arlington, Arlington, USA, in 2010. From 1998 to 2002 he was a Director Engineer with XJ Electric Co. Ltd. in China. In 2003, he joined the HUST and now is a Professor with the School of Electrical and Electronics Engineering, HUST. His current research interests include renewable energy integration, energy storage, multi-terminal HVDC and power system operation and control.

Yuan Pan received the B.Eng. degree from the Department of Electrical Power, Huazhong Institute of Technology, Wuhan, China, in 1955. He was with the Institute of Atomic Energy, Southwest Institute of Physics, and the Plasma Physics Institute, Chinese Academy of Sciences, Beijing, China. He was an Elected Academician of CAE in 1997. He is currently a Professor and a Ph.D. Instructor. He is an Expert in magnetic confinement fusion technology and high-power pulse electrical source technology. Professor Pan is a member of the Chinese Expert Committee of International Thermonuclear Experimental Reactor (ITER).

Shijie Cheng (LF'11) received the B.S. degree from Xi'an Jiaotong University, Xi'an, China, in 1967, the M.Sc. degree from Huazhong University of Science and Technology (HUST), Wuhan, China, in 1981, and the Ph.D. degree from the University of Calgary, Calgary, AB, Canada, in 1986, all in Electrical Engineering. He has been a Professor with the School of Electrical and Electronics Engineering, HUST, Wuhan, China, since 1991. His research interests are power system control, stability analysis, application of Artificial Intelligence, and energy storage. Professor Cheng is Academician of the CAS.