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Designing the Next-Generation of Proton Exchange Membrane Fuel Cells

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Abstract

With the rapid growth and development of proton exchange membrane fuel cell (PEMFC) technology there has been an increasing demand for clean and sustainable global energy applications. While there are many device-level and infrastructure challenges still to be overcome before wide commercialization can be realized, increasing the PEMFC power density is a critical technical challenge, with ambitious goals proposed globally. For example, the short-term and long-term goals of the Japan New Energy and Industrial Technology Development Organization (NEDO) are 6 kW L^{-1} by 2030 and 9 kW L^{-1} by 2040, respectively. To this end, we propose technical development directions required for next-generation high power density PEMFCs. This perspective comprehensively embraces the latest advanced ideas for improvements in the membrane electrode assembly (MEA) and its components, bipolar plate (BP), integrated BP-MEA design, with regard to water and thermal management, and materials. The realization of these ideas is expected to be encompassed in next-generation PEMFCs with the aim of achieving a high power density.

Key words: proton exchange membrane fuel cell, power density, membrane electrode assembly, bipolar plate, clean and sustainable energy.

Introduction

Addressing the increasing global energy consumption and environmental pollution caused by fossil energy usage has spurred a worldwide growth in renewable and eco-friendly energy solutions¹. The hydrogen economy based on renewables, including hydrogen production, hydrogen storage, and conversion of hydrogen to electricity, is considered as a promising solution for the future of energy by many authorities. In the hydrogen economy, fuel cell vehicles (FCVs) are a critical segment for delivering low-carbon transport, and the well-to-wheels greenhouse gas emissions are reduced to near zero when hydrogen is produced from renewables²⁻⁴.

As the two low-carbon transport routes, FCVs and battery electric vehicles (BEVs) are often compared⁵. Batteries are energy storage devices, whereas fuel cells are energy conversion devices that typically use hydrogen for energy storage. As a storage medium, hydrogen has inherent advantages over lithium-ion batteries, exhibiting higher energy density and shorter refueling time for FCVs⁶. The FCV also outperforms the BEV under subzero temperature conditions, as the BEV often exhibits significantly reduced discharge capacity^{7,8}. The cost of FCVs is currently higher than that of BEVs for short-range vehicles (below 200 miles, 322 km); however, their cost is comparable to or lower than that of BEVs with high annual production rates, particularly for long-range vehicles (above 300 miles, 483 km)^{6,9}. On the downside, the efficiency of FCVs is considerably lower than that of BEVs on the usage side, and the hydrogen infrastructure is still at the preliminary stage¹⁰⁻¹². Based on the differences in their technical characteristics, the prevailing opinion is that FCVs are better suited to heavy-duty and long-distance transportation as well as for other commercial vehicles, such as forklifts, whereas BEVs are more suitable for light-duty and short-distance transportation (Figure 1)¹³. Large-scale expansion of the BEV market has been achieved with the rapid technical development of lithium-ion batteries in the past two decades combined with grid at-home recharging. As the core component of FCVs, proton exchange membrane fuel cells (PEMFCs) have technical barriers that need to be overcome; moreover, the performance, cost, and durability of the PEMFC stack significantly affect the large-scale commercialization of FCVs. Improving power density is vital for the development of FCVs^{3,5,8}. The stack power densities with and without end plates of the second-generation MIRAI, which is the latest Toyota FCV launched at the end of 2020, achieved 4.4

and 5.4 kW L⁻¹, an increase of 42% and 54% in comparison with those in the previous generation, respectively^{8,14,15}. According to the Japan New Energy and Industrial Technology Development Organization (NEDO), the 2030 and 2040 targets of the stack power density for automotive applications are 6.0 and 9.0 kW L⁻¹, respectively¹⁶. The European Union Fuel Cells and Hydrogen 2 Joint Undertaking (EU-FCH2JU) recently demonstrated their PEMFC stack with a power density of 5.38 kW L^{-1} (with end plates) at 2.67 A cm⁻² and 0.6 V¹⁷, and the goal is 9.3 kW L⁻¹ by 2024¹⁸.

An overview of the progressive improvements needed for PEMFCs to meet the future high power density requirements is illustrated in Figure 2, accompanied by a schematic explanation of the working principle. The current technical status and the widely acknowledged prospects of PEMFCs are listed based on individual components. A typical single PEMFC generally comprises a membrane electrode assembly (MEA) and bipolar plates (BPs), wherein the MEA comprises a gas diffusion layer (GDL) with a microporous layer (MPL), catalyst layer (CL), and proton exchange membrane (PEM). In addition to the desired electrical output, water and heat are generated by electrochemical reactions, which affect the PEMFC operation. It is vital to maintain an appropriate water content and a suitable operating temperature, through water and thermal management, to attain a desirable trade-off among sufficient PEM hydration, unobstructed reactant supply, high catalyst activity, and adequate component lifetime. The GDL is a layer of carbon paper, which plays multiple important roles in gas distribution, mechanical support, and electrical connection. It is typically attached to another layer made up of carbon black and polytetrafluoroethylene (PTFE), commonly known as the MPL, which assists in the timely removal of electrochemically produced water. The CL is the site wherein the hydrogen oxidation and oxygen reduction electrochemical reactions occur through a series of coupled physiochemical processes. Platinum (Pt)-loaded carbon, which is finely dispersed with ionomer, is the most commonly used catalyst owing to its excellent activity and durability in an electrochemical environment. Decreasing the platinum group metal (PGM) loading is an unchanging goal for the development of CL owing to its high cost and limited resources. The core component of a PEMFC is the PEM, which functions as a proton conductor, an electrical insulator, and a gas separator. Better temperature- and humidity-range tolerances, as well as sufficient mechanical strength, are necessary to meet the future higher power density and durability demands. The BP is a

rigid support structure in the PEMFC stack onto which the reactant and coolant flow fields are fabricated, and is responsible for current collection and heat dissipation. However, the BP also constitutes a large proportion of the stack volume and cost. A thinner structured BP with better mass transfer capacity, electrical conductivity, thermal conductivity, and lower cost is an inevitable design strategy to achieve higher power density and lower-cost PEMFCs¹⁵. A more detailed perspective of potential expectations and possible avenues for implementation at the component level, as depicted in Figure 2, are elaborated in the following sections.

According to the state-of-the-art FCV products and worldwide fuel cell programs, the volume power density of the stack with end plates is expected to reach 6 kW L⁻¹ in the next 5–10 years, with the working current density and cell voltage increasing to 3–4 A cm⁻² and 0.7–0.8 V, respectively. Furthermore, an ultimate stack target of 9 kW L⁻¹ may be achieved with further developments, with cell operating points raised to even higher levels, such as 4–5 A cm⁻² and 0.8–0.9 V. The mathematical conversion between the two description indices of the PEMFC performance (volume power density of stack kW L⁻¹ and current density of active area A cm⁻²) is based on public domain data and a calculation of the number of cells and the thickness of the end plates. The value of the volume power density throughout this paper refers to that of a stack with end plates, unless specified. To achieve the significant boost in the PEMFC stack power density required, a comprehensive understanding of the current challenges and potentials involving all the components of a PEMFC needs to be clarified, which is the objective of this perspective.

Membrane electrode assembly

The MEA consists of several individual components, namely the electrically conductive GDL that allows gases to permeate through, the CL in which the redox reactions occur, and the PEM, which acts as the electrolyte. With the aim of elevating the power density and promoting the more extensive commercial adoption of PEMFCs, it is of great importance to fabricate MEAs with higher performance, better durability, and lower cost.

Gas diffusion layer

In the foreseeable future, carbon paper is expected to continue to be the mainstream choice for GDLs owing to its comprehensive advantages in electrical conductivity, mechanical strength, chemical resistance, and fabrication cost¹⁹. Structural modification of a GDL, such as laser perforation, has been tested, and may provide a potential future direction²⁰. Owing to scale-discrepant transport characteristics between the BP and MEA (pore sizes range from the micron- to nano-scale), mass transfer for gas permeation and water management is complex. It is speculated that GDLs with a gradient pore size, possibly via manipulation of the carbon fiber arrangement, are expected to establish a more effective bridge between the flow field and MEA and improve the mass transfer ability of the MEA itself, as depicted in Figure 3a-I²¹⁻³¹. For example, decreasing the porosity on one or both sides of the GDL may reduce the contact resistance and create pore gradients inside the GDL to facilitate reactant supply and water removal.

Another important factor is the interfacial resistance between components, which mainly depends on the material properties and assembly process. The contact resistance between the BP and GDL is the main source of electrical impedance, which is approximately two orders of magnitude higher than the resistance of the GDL itself (estimated with a GDL conductivity of 10^3 S m⁻¹, thickness of 150 µm, and contact resistance of $10^{-6} \Omega$ m²). A metal sheet GDL has been used to lower the ohmic loss³²; however, the problem of corrosion still needs to be solved. With the consideration of alleviating or even eliminating the interfacial effect, the concept of component integration or unification is proposed, which entails replacing the BP and GDL with another component to meet the requirements of electric conduction, gas distribution, and water management simultaneously³³. The mass transfer path becomes shorter (expected through-plane distance, from 0.5–0.6 mm to 0.3–0.4 mm) to fulfill the demand of higher current density (3–4 A cm⁻²). This so-called integrated BP-MEA or GDL-less design, for example, adopting foam materials (see Figure 3a-II), is further discussed in the section Integrated porous BP-MEA design.

The proposed future power density target $(6-9 \text{ kW L}^{-1})$ also imposes higher requirements on water management capability. The MPL coating (typically 20–40% PTFE content) on the GDL impedes liquid water from pooling at the CL/MPL interface, thereby diminishing the blocking effect of liquid

water on the gas transport³⁴. Nevertheless, with the continuous improvement of membrane materials to tolerate higher temperatures and lower humidity, water management can be simplified¹⁶. Consequently, hydrophobic coating is not necessarily the only choice, and the design of MPL and GDL wettability, and their microstructures should be adapted to the evolving practical requirements. For instance, a hydrophilic anode and hydrophobic cathode or MEA divided into hydrophilic and hydrophobic zones may better fit the operation of a PEMFC stack operating without an external humidifier. It is anticipated that the development of GDLs together with MPLs will make ~10% contribution toward the improvement of power density, owing to the improved matching of other components (see Supplementary Information for the estimation).

Catalyst layer

The maximum power density is dominated by the performance of the CLs. To achieve a power density of 9 kW L^{-1} at a low catalyst loading, a high cell output voltage over 0.8 V and current density of 4.4 A cm⁻² are required¹⁶; however, this requires significant breakthroughs in catalyst activity and CL design, as depicted in Figure 2 and Figure 3b.

One main approach to increasing the specific activity (SA) or mass activity (MA) of the catalyst is to design a novel catalyst architecture (nanocage²¹, core-shell²², nanoframes²³, nanowire²⁴, nanocrystals²⁵, etc.). A representative example is Pt_3Ni nanoframe catalysts with extended platinum surfaces, which display enhancement factors of 36 in MA and 22 in SA in comparison with commercial Pt/C catalyst²³. Thus far, the highest SA and MA values reported are 11.5 mA cm⁻² and 13.6 A mg_{Pt}⁻¹ respectively, by using ultrafine jagged platinum nanowires²⁴. However, these catalysts are typically evaluated using the rotating disk electrode (RDE) method, with little practical application being reported at the MEA level. Recently, a breakthrough catalyst with an ultra-low concentration of Pt alloy supported on PGM-free materials (denoted as LP@PF-1) achieved an MA of 1.08 A mg_{Pt}⁻¹ and retained 64% of its initial value after 30,000 cycles in a fuel cell. However, MA decreased by one order of magnitude in comparison with the value measured by RDE²². Furthermore, some Pt nanoparticles are considered to be metastable, and the shape geometries diminish with catalyst age³⁵. Thus, a future challenge is the stabilization of catalyst particle shapes to enhance their

durability and simultaneously maintain their ultra-high SA or MA in an operating fuel cell environment.

Ionomers, in contact with the catalyst, ensure accessibility and transport of protons to and from the catalyst particles at the cathode and anode. Their distribution has a pronounced effect on ionic conductivity and Pt catalyst utilization²⁶. For example, recent research showed that Nitrogen-doped carbon supports could ensure unprecedented uniform coverage of the ionomer due to coulombic attraction between the ionomer and N groups on the carbon support, eventually achieving fuel cell power densities of up to 1.39 W cm^{-2} for a pure Pt catalyst²⁶. Furthermore, carbon supports with a preferred internal pore opening of 4-7 nm simultaneously conferred excellent ORR activity and transport properties²⁷. These results indicated that appropriate modification of the carbon supports could lead to marked improvements in PEMFC power densities. Interestingly, with regard to the catalyst/ionomer interface, shortening the side-chain length of ionomers also enhanced proton transport, but decreased local O₂-transport²⁸, suggesting another promising approach to achieving a balanced trade-off between proton transport and oxygen transport as well as promoting full utilization of the Pt catalyst³⁶. In addition, given that the water swelling/de-swelling of the ionomer may cause interfacial degradation between the CL and PEM³⁷, the ionomer must be dimensionally and physically compatible with the PEM with which it is in contact³⁸. A previous study demonstrated that it is of great significance to develop ionomers with high stability and reliable water retention ability for fabricating MEAs with improved durability and good performance in the future³⁹.

Order-structured MEAs possess highly efficient mass transport pathways and reaction sites as well as high utilization of Pt, enabling high power densities at relatively low catalyst loadings. The 3M nanostructured thin film (NSTF) catalyst is a well-known example of an extended surface area catalyst in an order-structured MEA^{40,41}, which has achieved a total PGM loading (0.094 mg cm⁻², 0.106 g_{PGM} kW⁻¹) for the MEAs, meeting the U.S. Department of Energy (DOE) target for 2020⁴². However, the NSTF CL has a wetting issue, making it unsuitable for integration into a CL. The mechanism of proton transport in this CL is currently unknown, and no commercial applications have been achieved. In addition, order-structured MEA based on vertically aligned carbon nanotube

CNT arrays and ionomers is another promising structure, which can deliver a high current density $(2.6 \text{ A cm}^{-2} \ @0.6 \text{ V})$ with low cathode Pt loading $(0.1 \text{ mg cm}^{-2}, 0.064 \text{ g kW}^{-1})^{43}$. These results indicate that order-structured MEAs are a promising approach to achieving high-power fuel cells with ultra-low catalyst loading. It is expected that improvements in CL design can account for about 40% improvement in power via advanced catalysts and CL architecture (see Supplementary Information for the estimation).

Proton exchange membrane

An ideal membrane for high-power PEMFCs is one with high proton conductivity under low humidity conditions and good electrochemical and mechanical stability⁴⁴. To improve the power density of fuel cells by optimizing the PEMs, the main strategy is to reduce the membrane thickness of commercial Perfluorosulfonic acid (PFSA)-based membranes. For example, the first-generation MIRAI adopted a state-of-the-art reinforced ultra-thin membrane (~10 μ m)⁴⁵, which not only reduced the proton and water transport path, but also achieved self-humidification to avoid anode dryness; however, thin membranes also face challenges of mechanical damage or electrochemical degradation^{46,47}. Thin perfluorinated membranes reinforced with PTFE exhibited enhanced mechanical and dimensional stability, while the first-generation MIRAI used a porous media flow field over the cathode GDL to further enhance the mechanical support of the thin PEM⁴⁵. Another classic strategy to extend the stability of thin membranes, which is used in commercial practice, is to incorporate cerium salt into the membrane 4^{48} . For example, the second-generation MIRAI adopted the new Gore-Select[®] Membrane⁴⁹ containing cerium salt, which achieved improved performance and mechanical durability, despite being 30% thinner than the previous version. Furthermore, polydopamine-treated composite membranes with self-supported CeO_x radical scavengers exhibited simultaneously enhanced chemical/mechanical durability, which might be an innovative approach for stabilizing future high-performance PEMFCs (as depicted in Figure 3c)²⁹.

Sulfonated hydrocarbon polymers, including sulfonated poly(ether ether ketone)s (SPEEK)⁵⁰, sulfonated poly(ether sulfone) (SPES)⁵¹, sulfonated polyimides (SPI)⁵², and sulfonated polyphenylene-based PEMs⁵³ have also been considered as promising alternatives owing to their

high proton conductivity and thermal stability at elevated temperatures and high humidities. However, many hydrocarbon-based PEMs commonly suffer from relatively low conductivity at low RH, for example, <40% RH, compared with PFSA^{54,55}. Examples of some of the most promising hydrocarbon-based PEMs in terms of stability or fuel cell performance under low RH conditions are sulfonated polyphenylenes⁵⁶⁻⁵⁸, phosphoric acid quaternary amine-biphosphate ion pair membranes⁵⁹ and interpenetrating network membranes⁶⁰. To address low RH and elevated temperature operation, cactus-inspired nano-crack-structured self-humidifying membranes have been reported³⁰. Thin (~nm) hydrophobic layers with nano-cracks on the surface of hydrocarbon FEM can regulate water retention, even at low humidities and elevated temperatures (~120 °C). Recently, through-plane oriented proton transport channels in hydrocarbon PEMs demonstrated efficient proton conduction even at significantly low RH (20–40%) and exhibited a fairly high power output at 120 ℃ (Figure 3c)³¹. Most interestingly, these PEMs with oriented proton transport structures also displayed considerably high water retention due to their microporous structures and unexpected durability compared with that of Nafion[®] 212⁶¹. Furthermore, ferrocyanide groups incorporated into polymeric structures imparted both proton conduction and a high resistance to oxidative free radical degradation, suggesting that this strategy may be a new avenue toward ferrocyanide-stabilized inexpensive durable hydrocarbon-based PEMs⁶². An OCV held at 90 °C and 30% RH on MEAs displayed only ~2.0% loss after 80 h, in comparison with a 28.2% loss for Nafion[®] after 50 h.

PFSA-based polymer membranes are expected to continue to play a dominant role for the next 5–10 years, and continuous improvements in PEMs are expected to contribute 10–20% to the improvement of power density (see Supplementary Information for the estimation). However, with significant step-change improvements and innovations in stability, less expensive non-perfluorinated PEMs are expected to gain more prominence. Furthermore, PEMs capable of operating under low RH conditions may reduce the humidification requirements, thus indirectly improving the power density.

Bipolar plate

Since the adoption of PEMFCs for automotive application in the late 20th century, BP development and innovation have been constant^{63,64}. However, further improvements in the stack power density to 6.0 and 9.0 kW L⁻¹ will inevitably introduce additional technical challenges and still require an additional ~20% contribution of power density from BP innovation (see Supplementary Information). Here, an introduction of the BP and its technical limitations is presented with a discussion of individual technical design challenges combined with U.S. DOE and Japan NEDO reports^{16,65-68}. Using these reports and products from Toyota and Honda as examples, a comprehensive overview of these technical challenges over the past two decades is plotted in Figure 4^{15,16,69-72}. Both carbon and metal BPs have been considered because of their long-term development and dramatic progress.

The mass transport capacity is an important criterion for a BP design, which is mainly dependent on the flow field structure⁷³. Currently, there are two established design routes for BP structural designs, as shown in Figure 4. One is to modify and narrow the channel-rib structure, and the other is to develop flow fields with no ribs but with micro baffles or porous structures. These routes have shown some advantages over conventional designs, but they are also accompanied by new complications. The 3-D fine-mesh BP of the first-generation MIRAI with baffles stamped in a fish-scale pattern, for example, had demonstrated efficient mass transfer⁶⁷. However, the tearing process also caused fracture surface cracks, exposing the metallic substrate to an acidic electrochemical environment⁷⁴. Furthermore, progressive stamping of refined meshes increases the BP costs. The U.S. DOE requires that the BP should achieve a permeability to hydrogen and oxygen lower than 2×10^{-6} m² by 2020, while the current flow channels/fields with a depth lower than 0.5 mm can easily reach a permeability of 10^{-8} m². Toyota has replaced the 3-D flow field by a 2-D wave channel on the second-generation MIRAI. These indicate that the mass transport capacity may not be a major concern for the current stacks. However, there is still a gulf between the current status (4.4 kW L⁻¹ for second-generation MIRAI) and the power density goal as high as 9 kW L⁻¹, and the current density goal of 4.4 A cm⁻² (by Japan NEDO)¹⁶. A concomitant increase in the gas flow rate can exacerbate the pressure drop and maldistribution in the flow channels/fields, leading to an increase in parasitic power loss, local fuel starvation, and thus mass transfer loss. Another issue is the liquid removal induced by a high electrochemical water production rate. Although modified MEAs

can be operated at lower humidification levels, water condensation and accumulation are still difficult to avoid, particularly at current densities of 2 A cm^{-2} and above⁷⁵. However, at operating temperatures of 100 °C and above, this issue might be largely mitigated by evaporation. In summary, the enhancement of mass transfer remains a dominant challenge for the BP design of next-generation PEMFCs.

Heat and electron conduction are two other challenges in BP design. Because the heat dissipated from the cell edges or removed by mass flow is almost negligible, most of the waste heat must be transferred through the MEA and BP and then removed by the external radiator coolant through thermal convection⁷⁶. For a stack with a power density of ~4 kW L^{-1} , the BP has a thermal conductivity ~30 times higher than that of the GDL or CL, but the interfacial thermal resistance between the BP and GDL can be ~10 times larger than that in BP⁷⁷. The interfacial electrical resistance between the BP and GDL (~ $10^{-6} \Omega m^2$) is nearly four orders of magnitude higher than that in BP (~10⁻¹⁰ Ω m²). While current metal separators can easily reach thermal and electrical conductivity values of 50 W m⁻¹ K⁻¹ and 1.4×10^6 S m⁻¹, respectively, far exceeding the U.S. DOE criteria for 2020, the interfacial resistance is only comparable⁶⁸. Japan NEDO proposed that the output voltage and operating temperature should be further improved from the current status, that is, 0.65 V and 90–100 %, to 0.85 V and 120 % in 2040¹⁶. Thus, the major concern is to reduce the thermal and electrical interfacial contact resistance between the BP and GDL, which is sensitive to the cell stack compactness, contact area between BP and MEA, BP surface roughness, and substrate conductivity. In addition, the current coolant channels must be integrated with hydrogen and/or air distributors for a compact cell structure, inferring a conflict between the cooling capacity and thickness reduction.

The durability of BP is predominantly influenced by electrochemical corrosion and mechanical degradation. A further increase in the current density (~3.8 A cm⁻² by 2030, Japan NEDO¹⁶), may cause an increase in the electrochemical corrosion of the components may be a leading cause of cell degradation, especially for metal BPs. For carbon BPs, corrosion also occurs, but is almost negligible under normal operating conditions. To mitigate corrosion, the BP is generally treated with a passive

layer by incorporating materials capable of forming low-resistance oxide films or by the deposition of corrosion-resistant coatings. The efficacy of corrosion resistance is mainly determined by the uniformity of defect-free coatings, stable passive films, or nitride layers⁷⁸. According to the U.S. DOE criteria, the corrosion current density of the anode and cathode BPs should be limited to <1 µA cm⁻² in 2020⁶⁵. The current coatings, for example, multi-layer chromium carbide coating and amorphous carbon coating, on SS 316L (or graphite) already meet the criteria⁶⁶. However, an advanced coating technique is still necessary to achieve better corrosion resistance, lower contact resistance, and, most importantly, lower coating cost^{66,67}. Mechanically, the swelling and shrinking of the MEA and pressure variations of the gas and coolant may induce load and pressure fluctuations of the metal BP, leading to plastic deformation and fatigue failure⁷⁹. The U.S. DOE set criteria for 2020 for flexural strength and impact strength of BP of 25 MPa and 40.5 J m⁻¹, respectively⁶⁴. Typical BP substrate materials, such as stainless steel (by Honda⁷⁰) and flexible graphite (by Ballard⁸⁰), have already met these criteria. However, localized thickness reduction, cracks, and susceptibility to plastic deformation may occur during manufacturing processes or after long-term operation. Furthermore, for refined ribs, ultra-thin plates, meshes, baffles, or metal ligaments, a high compact pressure can result in BP deformation and weak contact between the BP and GDL⁸¹.

While achieving target transport capabilities, the fabrication capacity of BPs should correspond with the industrial base for the mass production of fuel cell products. The cost and volume of BPs can account for over 30% and 70% of the entire PEMFC stack, respectively, depending on the substrate material, fabrication capacity, and coating technique, etc⁶⁴. The U.S. DOE criterion for 2020 for the total BP cost, inclusive of materials, forming, and coating, is \$3 kW⁻¹. However, for the substrate material, such as SS 316L, the cost alone is close to \$3 kW⁻¹ (~\$2.7 kW⁻¹), making it difficult to achieve this goal⁶⁷. Furthermore, utilizing ultra-refined plates has recently become a widely adopted approach for BP design, but it requires high precision and rapid fabrication processes for metal or graphite substrates, such as progressive stamping and compression molding. The precision and uniform manufacturing of both BPs and coatings are anticipated to be major technical obstacles for high-volume production. Thus, both Japan NEDO and U.S. DOE have emphasized the cost reduction of BP for the further development of fuel cell technology and the FCV industry^{16,67}.

Integrated porous BP-MEA design

Recently, a new type of BP has been proposed that utilizes a metal/graphene porous foam as a reactant distributor⁸². With appropriate mechanical properties, uniform mass and heat distribution can be achieved with a reduced volume and weight of a porous BP^{83} . These porous materials can be fabricated at a considerably lower cost than that of refined ribs or baffles, and their geometric parameters, including porosity, pore density, and pore shape, are controllable⁸⁴. This may extend to the complete removal of the GDL and its replacement with porous material solely for mass transport between the CL and external environment, i.e., an integrated porous BP-MEA design. In addition to a more compact cell structure, this integration may advantageously eliminate the interfacial transport of mass, heat, and electrons between the BP and GDL, thereby avoiding interfacial transport resistances. Tanaka et al.³³ introduced a GDL-less cell design using a corrugated SS mesh flow channel rather than a porous material and a freestanding MPL, demonstrating the feasibility of integration. Park et al.⁸⁵ adopted graphene foam to fabricate a unified flow field/GDL without an MPL. The cell thickness decreased by 82% while achieving a higher power output over a wide current density range. Although better performance is achieved with a thinner cell through the integrated BP-MEA design, porous flow field materials with high electrical conductivity are susceptible to chemical corrosion in acidic environments. Consequently, effective coating materials and coating methods for 3-D porous structures are necessary to achieve long-term operational stability. Moreover, porous materials exhibit a lower stiffness than conventional BPs. A BP stiffness enhancement process, for example, compression, is necessary to avoid over-deformation of the flow field. In summary, integrated BP-MEA or GDL-less designs can simultaneously improve mass transport, alleviate water flooding, and decrease the volume of PEMFC stacks and may provide a promising avenue for ultra-high power density (9 kW L⁻¹) owing to significant improvements in stack compactness.

Summary and outlook

PEMFCs have recently gained attention for automotive propulsion owing to their advantages of FCVs and remarkable technical progress in recent years. This perspective aims to highlight the development directions of PEMFC components in the entirety of their interrelationship and design, to achieve the goal of a step change in power density for next-generation PEMFCs. In other words, advancing the power density from the current status (~4 kW L⁻¹) to the short-term target of 6 kW L⁻¹ and long-term target of 9 kW L⁻¹.

Future developments of GDLs and MPLs should focus on the optimization of cross-scale and cross-component transport, while being compatible with improvements in other components regarding the control of structure and wettability. For the CL, the activities of novel catalysts are sufficiently large at the RDE level but still need significant improvement at the MEA and stack levels. Modification of carbon supports and catalyst/polymer interfaces based on molecular arrangements is promising for improving the ionomer distribution and catalyst utilization. Order-structured MEA is a strong candidate for the future owing to its unique merit of enabling high power density at ultra-low catalyst loadings. In the next 5-10 years, PFSA-based polymers with enhanced durability and adaptability are expected to continue to dominate the PEM market. The future direction of BP design is to solve the corrosion resistance, fabrication cost and interfacial contact resistance issues. The mass transport capacity needs to be enhanced for future ultra-high power density operations. The integrated BP-MEA design is expected to provide a promising path toward ultra-high power density owing to its advantages of interface elimination and volume reduction.

Improving power densities, reducing costs, and increasing the durability of PEMFCs will directly promote large-scale commercialization, and the three criteria are highly coupled to, and sometimes constrained by, each other. The balance among the three criteria in the development of different fuel cell products should also be considered. Overall, establishing refined and controllable structural designs that can be readily manufactured is the key development direction based on the existing framework of materials, and the development of new materials is expected to have revolutionary effects in the long run.

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Author contributions

K.J., J.X., Q.D., Z.H., and M.D.G. conceived the study. All authors contributed to the writing and commented on the manuscript. K.J., Z.B., B.X., B.W., Y.Z., L.F., and M.D.G. contributed to the preparation of the figures.

Declaration of interests

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

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This study proposes an integrated design of gas diffusion layer and flow field with boosted fuel cell performance, demonstrating that such design is promising for increasing the power density.

50 10 2 1	Vehicle size / tonne		FCV do	main
0	200	400	600	800
	BEVs		FCVs	
	Storage Media	Li-ion batteries		Hydrogen
	Energy Density*	<200 Wh kg ⁻¹		~300 Wh kg ⁻¹
	Refueling Time**	Hours		Minutes
*	Low temperature	Severe degradation		Demonstrated at -30 °C
C	Efficiency***	~90%		~60%
1	La Infrastructure	Grid & char	ging piles	Hydrogen stations

* Energy density of the powertrain system, which includes the battery pack and battery management system for BEVs, and the fuel cell system and hydrogen tank for FCVs.

** The refueling time of fuel cell passenger vehicles is approximately 5-10 min. The normal charging time of the similar scale BEVs is several to dozens of hours. The fast charging of BEVs is not considered here because it substantially compromises cycle life and cannot be used for daily charging.

*** The efficiency of BEVs is charging efficiency, while that of FCVs is hydrogen to electricity efficiency.

Figure 1 Expected application domains of the BEV and FCV in future automotive transportation, and technical characteristic comparisons between FCVs and BEVs⁶⁻¹³. The advantages of FCVs include better energy medium, higher energy density, less refueling time, lower cost and safety risks with increasing energy density, and better low-temperature performance. BEVs have higher efficiency on the usage side and a wider infrastructure.



Figure 2 Overview of progressive improvements in PEMFC to meet future high power density requirements and a schematic explanation of the working principle. Owing to the largely proprietary nature of many commercial technologies, some estimations of the present technology status and potential for the future are given. Seven components of a PEMFC are presented, including BPs with flow channel/field, GDLs and CLs at the anode and cathode, respectively, and the central PEM. During PEMFC operation, hydrogen is fed to the anode and air (or oxygen) is fed to the cathode. The hydrogen oxidation reaction dissociates the hydrogen molecules into protons and

electrons at the anode CL. Protons transport to the cathode CL through the PEM, while electrons travel through the external circuit. The oxygen reduction reaction (ORR) occurs at the cathode CL among protons, electrons, and oxygen molecules, which generates water, heat, and electricity. In the lower part of this figure, the BP thickness refers to the distance between two adjacent MEAs in the fuel cell stack.



Figure 3 State-of-the-art and next-generation MEA designs. **a** Two potential approaches for future GDL development: I Pore size gradient by manipulation of carbon fiber arrangement, II Integrated BP-MEA design or GDL-less design adopting foam materials. **b** Some key CL advances: I Novel catalysts with special shapes and ultra-high activities achieving higher power density and lower catalyst loadings (Pt_{1.5}Ni nanocage²¹, PtCo core shell²², PtNi nanoframe²³, Pt nanowire²⁴, Transition metal–doped Pt₃Ni octahedra²⁵), II Modification of carbon supports enabling the uniform distribution of ionomers and high catalyst utilization (Nitrogen-doping of carbon supports²⁶ and carbon with accessible mesopores²⁷), III Boosting Pt/ionomer interface via molecular arrangements²⁸; **c** New design approaches for PEMs: I Ce-doping membrane enhancing membrane stability²⁹, II Nanocrack-regulated self-humidifying membrane regulating water-retention³⁰, III Membrane with through-plane oriented proton transport channels enabling efficient proton conduction even at an extremely low relative humidity (RH)³¹. Images adapted with permission from refs 21-25 (the American Association for the Advancement of Science), ref. 27 (American Chemical Society), ref. 28 (Multidisciplinary Digital Publishing Institute), ref. 29 (John Wiley and Sons), and refs 26, 30, 31 (Springer Nature).



Figure 4 Development trend of BPs on FCVs, obtained from the FCV products of Toyota Motor and Honda Motor ^{15,69-72}, and the dotted lines represent the trend predictions. The power density by volume is evaluated with the end plates of a stack. Superscripts distinguish four types of BP: ^{*} a carbon straight flow channel, ^{***} a metal straight flow channel, ^{****} a metal (refined) wave flow channel, ^{*****} a metal 3-D fine mesh flow field. The BPs with wave channels were first utilized by Honda FCV in 2006 and have undergone constant refinements in each generation. The 3-D fine-mesh flow field was developed by first-generation (1st gen) MIRAI in 2014, presenting an innovative approach to BP design. However, it was simplified into a 2-D structure for the BPs on second-generation (2nd gen) MIRAI in 2020, possibly due to cost restraints. Japan NEDO predicted that the target power density should be achieved with a current density of 3.0 A cm⁻² in 2025¹⁶.