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# Review article

# Materials, components, assembly and performance of flexible polymer electrolyte membrane fuel cell: A review



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# HIGHLIGHTS

# G R A P H I C A L A B S T R A C T

- Crucial insights and information on flexible PEMFC are provided.
- Materials and components for achieving flexibility are summarized.
- Assembly techniques for flexible single cell and stack are presented.
- Performance and durability under various bending conditions are analyzed.
- Remaining challenges and future development on flexible PEMFC are discussed.

## ARTICLE INFO

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## ABSTRACT

With emerging demand of potable and wearable electronic devices, reliable and flexible energy suppliers are inevitable. Polymer electrolyte membrane fuel cells (PEMFCs) attract great attention due to high energy density and sustainability. However, non-bendability limits their application in flexible electronic devices. To make PEMFCs adaptable and flexible, considerable efforts have been devoted to developing various bendable components or advanced techniques. This review, therefore, focuses on the advancement of components and relative techniques of flexible PEMFCs, which determine the performance and durability, while achieved little concern in other reviews. The components and techniques include membrane, flexible catalytic layer, flexible gas diffusion layer, flexible pipolar plates, assembly of single cell or stack, store or supply of fuel and oxidant. In each section, the materials or techniques of flexible PEMFCs and then proceeding to the development of flexible components and relevant techniques of flexible PEMFCs. Subsequently, the flexible PEMFCs' performance and durability are presented, reaching to 100–200 mW cm<sup>-2</sup> and dozens of hours, respectively, still far lower than those of conventional PEMFCs. Finally, a brief perspective on remaining challenges and future development of flexible PEMFCs are provided.

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# 1. Introduction

As the demand for the application of flexible electronic devices [1-3](e.g., wearable devices and roll up displays) increases and is expected to increase greatly in the coming years, safe and reliable energy supplies is necessary to guarantee their long-term operation [4-6]. The commonly used energy supplies for electronic devices include lithium-ion batteries, supercapacitors, or solar cells, which are always too heavy, rigid, or bulky to fit the flexible electronic devices. Therefore, the energy supplies with stretchability and flexibility were developed, such as flexible lithium-ion batteries, supercapacitors, or solar cells, and successfully applied in the flexible electronic devices [7–13]. However, there are still several barriers need to be overcome for the flexible energy supplies in order to widely apply in practical products, e.g., limited energy storage (flexible lithium-ion batteries or supercapacitors), lower mass power density, lifetime (including electrochemical and mechanical durability, especially mechanical durability), or limited by sun light (flexible solar cells).

Fuel cell [14–16], an energy conversion device, could directly convert the chemical energy into electricity. It shows a variety of advantages, e.g., high energy density, high energy conversion efficiency, sustainability, low pollution emission, and without the necessity of charging. Therefore, the obstacles that the flexible energy supplies face may be resolved by reasonably designing and fabricating flexible fuel cells. Among the various fuel cells, proton exchange membrane (PEM) fuel cell [17–21] exhibits a significant superiority in the flexible power generators due to the flexible and solid electrolyte membrane [22] commonly used (e.g., Nafion membrane [23]), low operation temperature (less than or equal to 80 °C), or compact design. However, although the PEM is flexible, the catalyst layers (CLs) [24–30], gas diffusion layers (GDLs) [18,31,32], and bipolar plates [33–35] traditionally used are rigid, brittle, heavy, bulky, or expensive, limiting the application of PEMFC in the flexible energy supplies.

To realize the flexibility of PEMFC, there are many attempts to develop light, and flexible CLs, GDLs, and bipolar plates. For example, to allow the CL to bend or twist, the carbon fiber cloth (CFC), capable of bending, was introduced to support catalyst [36]; the flexible GDLs have been prepared by using CFC or modified CFC instead of the brittle carbon paper commonly used in the tranditional GDL [37]; the flow channel patterned polymer films were used as the substrate to forming the flexible bipolar plates [38].

The flexible PEMFCs have been fabricated by assembling these flexible components and show promising performance and durability [39,40]. To supply satisfactory power and keep the flexibility in practice, numerous flexible PEMFCs are usually tiled, forming the flexible PEMFC stack [41]. This is different from the traditional PEMFC stack, which is always assembled by stacking the multiple single cells one upon one.

Except the flexible components and the assembly, the storage or supply of fuel [42,43] and oxidant are also essential for the practical application of the flexible PEMFC in flexible devices. To fit the flexible electronic devices, some novel fuel containers with light weight, compact structure, and flexibility have been developed for the flexible PEMFC systems [40,44]. Due to no need for the air supply system, the self-breathing PEMFC shows a great advantage in the flexible devices [45,46]. With the development of the major components, assembly technique, and the fuel and oxidant storage or supply, the size and weight of flexible PEMFC have been greatly decreased, and its flexibility, performance, and durability have been significantly improved. The thickness and weight of the flexible PEMFC were declined by using an ultra-thin and flexible GDL composed of carbon nanotubes, leading to the high volume-specific power density (15600 W  $L^{-1}$ ) and weight-specific power density (9660 W kg<sup>-1</sup>) [47]. The flexible direct methanol fuel cell (DMFC) containing the flexible GDL based on TiC/carbon nanofibers films exhibited a maximum power density of 20.2 mW cm $^{-2}$  and a good stability in the repetitive bending test [44]. Yoo

et al. implemented a flexible PEMFC with a rubber-like photopolymer-based flow-field plates, which could provide a high power density of 87.1 mW cm<sup>-2</sup> in maximum bending, larger than that in flat position [48].

To summarize the development of the flexible PEMFC, Yang et al. focused on the operational system, fabrication approach, powergeneration property, and application demonstration of the flexible fuel cells [49]. However, there are few reviews focusing on the major components, assembly process, as well as the fuel and oxidant storage or supply system, which determine the performance, durability, and cost of the flexible PEMFC. Hence, this review will focus on the current development of the flexible components, assembly technique of the flexible PEMFC (single cell and stack), the fuel and oxidant storage or supply system, as well as the performance and durability of the flexible PEMFC. Subsequently, a brief perspective for future research and development of the flexible PEMFC are provided.

# 2. Materials and components for flexible PEMFC

A single PEMFC is commonly constituted by two bipolar plates and membrane electrode assemblies (MEAs). The MEAs include a PEM, two CLs, and GDLs. Except for PEM, the other components are unflexible and could not be used to fabricate the flexible PEMFC. Therefore, it is necessary to introduce the flexible materials with the same features (e. g., conductivity, permeability) to replace the tranditional CLs, GDLs, and bipolar plates to realize the flexibility of PEMFC. Besides, the novel flexible PEMs with improved proton conductivity under low humidity and low cost also need to be developed to decrease the fuel cell's cost. In this section, the recent development of the PEM as well as the flexible CLs, GDLs, and bipolar plates will be present in detail.

#### 2.1. Flexible proton exchange membranes

In PEMFC, PEM needs to effectively transport protons from anode to cathode and separate the fuel and oxidant. The optimum PEM should have the features of high proton conductivity, the ability to prevent gas/ liquid penetration and thermal/mechanical stability. From the time being, the commonly used PEMs include perfluorinated sulfonic acid membrane (PFSA membrane, e.g., Nafion membrane most commonly used) in the low-temperature PEMFC and phosphoric acid-doped polybenzimidazole film (PBI film) in the high-temperature PEMFC. Due to the relatively high operation temperature, the PBI-based PEMFCs are commonly unsuitable for flexible energy suppliers. Therefore, the present review is focused on PFSA membranes and the PEMFCs in this review is only low-temperature PEMFCs. PFSA membrane is always flexible so that it can be used directly in the flexible PEMFC. However, there still exist some disadvantages limiting its application. For example, the Nafion membrane's performance is greatly sensitive to the humidity and temperature. Because of a significant reduction in size, cost, and weight of PEMFC system, self-breathing mode is generally adopted in the flexible PEMFCs as mentioned in section 5, which means a low relative humidity. The proton conductivity of Nafion membrane will greatly decrease with a decrease of humidity. Besides, the expensive price of PFSA membrane will impede the widespread application of the flexible PEMFCs, which are a problem of the conventional PEMFCs as well [50,51]. Not only conventional PEMFCs but also flexible PEMFCs still face the limited mechanical stability of PFSA membrane in the temperature and humidity cycling as well as its chemical degradation caused by the production of  $H_2O_2$  and metal ions [52,53].

To improve the proton conductivity under low humidity and durability as well as decrease the cost of PEMs, several novel polymer membranes have been developed and successfully applied in the flexible PEMFC. Yoon et al. [54] synthesized two kinds of polymer electrolytes with excellent thermal and chemical stability (Fig. 1a). When being angled, these membranes exhibited more excellent ion exchange capacity and water absorption ability at 80 °C and the relative humidity of



Fig. 1. (a) Fabrication method of the blend polymer membrane [54]; (b) the b-SS glass electrolyte fabrication procedure and unique structure [56]; (c) structure of the flexible DMFC with different components [57].

80% than the commercial Nafion 117 membrane. A PEM based on photosensitive SU-8 and Cr/Au metal was prepared and successfully applied in flexible direct methanol micro-fuel cell (FDMMFC) [55]. The PEM showed increased mechanical stability and performance compared to the commercial Nafion 117 membrane in the bending test.

In addition to the polymer-based PEMs, the silicate-based thin films, capable of transporting proton, are the usually used PEM. However, the high price and brittleness limit their application in the flexible PEMFC. To improve the mechanical bendability and performance of the silicatebased PEMs, Lee, et al. [56] used polyimide nonwoven fabrics as the substrate and then impregnate 3-trihydroxysilyl-1-propanesulfonic acid (THPSA) and 3-glycidylox-ypropyl trimethoxysilane (GPTMS), followed by the in situ sol-gel synthesis, finally obtaining the sulfonic acid functionalized silicate (b-SS) glass electrolytes (Fig. 1b). This silicate-based PEM shows the enhanced mechanical bendability and improved performance at low humidity and medium temperature. Besides, its proton conductivity could be modulated by changing the ratio of THPSA and GPTMS. Li et al. [57] reported a flexible and high-performance DMFC (Fig. 1c) with quasi-solid kalium polyacrylate hydrogel electrolyte. This pre-swollen electrolyte exhibits excellent flexibility and capacity of preventing methanol penetration, enhancing the flexible fuel cell (8.86 mW cm<sup>-2</sup> at 30 °C).

Although most PEMs prepared in the literatures have shown good performance and durability in a short period than the commercial PEMs, their long-term durability is a challenge in the long-running process of the flexible PEMFC. Besides, it is another challenge to prepare these novel PEMs in mass production.

#### 2.2. Flexible catalyst layers

The conventional CLs are commonly fabricated by depositing the ink composed of Pt/C catalysts, ionomer, and solvent onto GDL or membrane. The prepared CLs have poor interfacial contact with membrane or GDL and the interaction between catalyst-based agglomerates in the CL is always poor. The CL may peel off or the integrity of CLs will be destroyed when the PEMFC is bended or twisted, resulting in the increased interfacial resistance or internal resistance in CL and thus the decreased performance. Therefore, the CL suitable for flexible PEMFC needs to rationally designed and fabricated by improve the structure or composition in order to improve its flexibility and strengthen its interfacial contact with membrane and GDL.

The three-dimensional (3D) networks made of one-dimensional (1D) nanowires, e.g., carbon fiber cloth (CFC), are always bendable due the flexibility of nanowire and the intertwined structure. Therefore, the flexible CLs could be prepared through introducing the flexible 3D networks, such as the CL based on the AuPd nanowire network loaded on CFC [58] (Fig. 2 a, b) and the CL with CFC supported PtCu alloy nanotube arrays (Pt/Cu ANTAs/CFC) [59] (Fig. 2 c). These CLs shown improved performance in the twisted test compared to the conventional catalyst particle-based CLs, resulting from the excellent flexibility.

## 2.3. Flexible gas diffusion layers

In PEMFC, the gas/liquid fuel or air in the flow channel need to diffuse into the CL through GDL [60,61], the reaction products are drained through GDL, and the electron is conducted from/to CL through GDL. The commonly used GDL in PEMFC is constituted by carbon paper as substrate layer and microporous layer composed of carbon powder and binder (e.g., polytetrafluoroethylene (PTFE)) [62,63]. The GDL should have high porosity, good permeability, and excellent electron conductivity. Some commercial carbon papers often applied in GDLs and their physical properties are listed in Table 1. It is worth being pointed that the physical properties of GDL inside PEMFCs often differ from that outside PEMFCs, e.g., porosity or permeability, as the GDL in PEMFCs bears compression stress under assembly force and bending stress in bending. Under compression and bending stress, the GDL's pore structure or porosity will be changed to a certain extent [64-67]. The carbon paper-based GDL's structure could be destroyed due to the irreversible fracture occurs in low bending stress or small bending angle, resulting in the decrease of performance, such as decreased electron conductivity. Therefore, the conventional GDL cannot be used in the flexible PEMFC. Preparing the flexible GDL is necessary for the flexible PEMFC. The flexible GDLs used for flexible PEMFCs and their features were summarized in Table 2.



Fig. 2. (a) The fabrication procedure of Au–Pd alloy NWs, (b) the fabrication Au<sub>95,1</sub>-Pd<sub>4.9</sub> alloy NWNs@ CFC [58], (c) the fabrication progress of Pt/Cu ANTAs/CFC [59].

Properties of some commercial carbon papers commonly used in GDL.

| Material   | TGP-H-060 | TGP-H-090 | SIGRACET (GDL-20BA) | Ballard (AvCarb EP40) | Spectracorp (2050-A) | LyFlex (C332) |
|--|-----------|-----------|---------------------|-----------------------|----------------------|---------------|
| Thickness (mm)   | 0.19      | 0.28      | 0.22                | 0.19                  | 0.26                 | 0.33          |
| Bulk Density (g/cm <sup>3</sup> )  | 0.44      | 0.44      | 0.29                | 0.21                  | 0.48                 | 0.37          |
| Porosity (%)   | 78        | 78        | 83                  | /                     | /                    | 79.5          |
| Gas Permeability (mL <sup>·</sup> mm $\text{cm}^{-2} \text{ h}^{-1} \text{ mmaq}^{-1}$ ) | 1900      | 1700      | 1800                | 1800                  | 1620                 | /             |
| Electrical resistance (through plane) (m $\Omega$ cm)                                    | 80        | 80        | 10                  | /                     | 70                   | 251           |
| Electrical resistance (in plane) (m $\Omega$ cm)   | 5.8       | 5.6       | /                   | 13                    | /                    | 24.1          |
| Tensile strength (N $cm^{-1}$ )  | 50        | 70        | /                   | 75                    | /                    | 594           |

To prepare the flexible carbon paper, the cellulose fiber was mixed with carbon fiber, achieving the carbon–cellulose fiber-based composites [37]. The carbon–cellulose fiber-based composite GDLs have superior flexibility, uniformity, and porosity. However, although the mechanical strength of the composite GDL will increase with an increase of cellulose fiber amount, its electron conductivity will decrease (electrical resistance >0.035  $\Omega$  cm<sup>2</sup>). As mentioned in the last section, CFC is

flexible. Besides, CFC has excellent capacity of transferring gas/liquid and electron, which will be a good substitute of carbon paper-based GDL [36,58]. In order to further enhance the mechanical properties of CFC, Ag nanowires (NWs) was introduced, preparing the Ag NWs-carbon fiber composited fiber cloth [69]. Ag NWs could well guarantee the GDL's conductivity and integrity when the carbon fibers are broken in bending. However, the mass transport resistance of GDL is increased due to the

Summary of the flexible GDLs and their properties.

| Materials for flexible GDL                         | Physical properties  | Remark  | Reference |
|--|--|---|-----------|
| Carbon cloth                                       | Flexibility  | Repetitively bended<br>with different<br>bending radius (83,<br>58, 47 mm)                            | [37]      |
| Carbon fiber-felt<br>and carbon fiber-<br>paper    | High thermostability and flexibility   | 1   | [68]      |
| Carbon–cellulose<br>fiber-based<br>composite       | Increased tensile<br>strength, high<br>uniformity and<br>porosity, and high<br>surface area  | /   | [37]      |
| Carbon paper<br>coated with Ag<br>NWs              | High electron<br>conductivity, low<br>electron resistance<br>(<0.28 Ω cm <sup>2</sup> ) and<br>flexibility   | More than 100 times<br>repetitive bending<br>under the maximum<br>bending radius (about<br>2 cm)      | [69]      |
| Composite of CNT<br>films, CNT<br>powder, and PTFE | Low thickness (<40 $\mu$ m), bending radius:<br>< 0.17 mm, high modulus and electron conductivity (22880 $\pm$ 3480 S m <sup>-1</sup> ), super-<br>hydrophobicity. | Low fabrication<br>temperature, low<br>cost, and scalable   | [47]      |
| TiC/carbon<br>nanofibers (CNFs)<br>film            | High porosity,<br>decreased charge<br>transfer resistance  | Simple<br>electrospinning<br>method; stable<br>voltage after 50 times<br>repetitive bending at<br>75° | [44]      |

addition of Ag NWs, thus decreasing the output power. Hoshi et al. [70] coated a graphene layer on CFC to increase effective surface area, and as a result, the flexible ascorbic acid fuel cell (AAFC) provided 34.1  $\mu$ W cm<sup>-2</sup> at 0.46 V with 100 mM ascorbic acid (AA) at room temperature.

Except the CFC-based GDLs, several novel flexible GDLs have been developed recently. For example, the flexible and porous carbon nanotube (CNT) membrane was introduced and combined with the carbon paper, forming the composite GDL (Fig. 3a) [46]. The composite GDL is flexible and porous, as well as shows good mechanical stability in bending and thus improves the performance of flexible PEMFC. Additionally, the GDLs prepared with CNT film, CNT powder and binder (Fig. 3b and c) [47] or TiC/CNT film [44] exhibited excellent flexibility and high performance for flexible PEMFCs. Furthermore, nickel foam was used to show excellent performance, which provided more options for fabricating flexible GDL.

# 2.4. Flexible bipolar plates

In PEMFC, the MEA is compressed between the bipolar plates to guarantee the excellent contact between components and thus minimize the interfacial contact resistance. The bipolar plates mainly provide the pathway for the flow of gas/liquid fuel or oxidant to MEA [71,72], harvest the current, separate the fuel and oxidant in the adjacent cells of the stack, introduce the coolant and conduct heat from the MEA to the cooling cell, as well as supply the mechanical support for the cell. Therefore, the bipolar plates should be conductive (2025 DOE targets: electron conductivity >100 S cm<sup>-1</sup>, areal specific resistance <0.01  $\Omega$ cm<sup>2</sup>), impermeable to liquid and gases (2025 DOE targets: H<sub>2</sub> permeability  $2 \times 10^{-6}$  cm<sup>3</sup> s<sup>-1</sup> cm<sup>-2</sup>), or heat-conductive, and possess sufficient strength (2025 DOE targets: flexural strength >40 MPa) [73-75]. The bipolar plates commonly used in PEMFC are made by graphite [76-78], metal [79-81], or composite material [82-84]. The conventional bipolar plates are heavy, bulky, and rigid, thus unsuitable for the flexible PEMFC which requires the light, thin, and flexible bipolar plates. The bipolar plates used in flexible fuel cells and their properties were summarized in Table 3.

To prepare the flexible bipolar plates, the polymer film could be used as the flow field plate and then coated with a thin-film metal layer acting as current collector [36,69,86,87,90]. For example, the flexible bipolar plates could be prepared by applying a flexible current collector layer composed of Ag nanowire percolating network to the flow channel patterned polydimethylsiloxane (PDMS) film (Fig. 4a) [69], or depositing an Au layer on the cycloolefin-based flow field plate (Fig. 4b) [38]. These polymer film-based bipolar plates are usually not only flexible but also light-weight, which are appliable for the flexible PEMFC. However, their electron conductivity ( $<10^5$  S m<sup>-1</sup>) is lower than the conventional bipolar plates ( $>10^5$  S m<sup>-1</sup>) [74,75], limiting the performance improvement of the flexible PEMFC. In addition, Fu-Kuang et al. developed a novel current collector composed of the metal wires



Fig. 3. (a) The fabrication procedure and SEM images of a composite electrode for the flexible air-breathing PEMFC [46], (b) overall morphology of the flexible PEMFC with different flexible components, (c) the fabrication procedure of the CNT-composite GDL [47].

Summary of the materials used to prepare the flexible bipolar plates, size and properties.

| Materials   | Size   | Properties   | Reference    |
|---|--|--|--------------|
| Cycloolefin polymer<br>(COP) film<br>Carbon fiber with            | Flow channel:<br>200*100 mm (w/<br>d), Thickness: Au<br>layer: 200 µm, Ti<br>layer: 30 nm<br>Plate: 3*1*4 cm (w/                 | Low thickness, cost<br>and weight, small<br>surface area, wide<br>applications and<br>simple fabrication.<br>High electron   | [38]<br>[85] |
| metal wires   | d/1)   | conductivity, low<br>contact resistance<br>(<80 m $\Omega$ cm <sup>2</sup> ) and<br>flexibility, stable<br>output with bending<br>radius (10, 17 and 45<br>cm).  |              |
| PDMS and Au–Ni<br>film  | Plate: 45*45 mm<br>(w/l); Thickness: Ni<br>layer: 880 nm, Au<br>layer: 3.8 μm; SC  | High electron<br>conductivity<br>(resistance <0.5 $\Omega$ ),<br>maximum bending<br>curvature: 1.8 m <sup>-1</sup> .   | [45]         |
| PDMS and Ag NWs   | Symmetric plate<br>thickness: A: 5 mm,<br>CA: 5 mm,<br>Asymmetric plate<br>thickness: A: 4 mm,<br>CA: 6 mm,<br>SC                | High mechanical<br>stretch and electron<br>conductivity decrease<br>oxygen reduction<br>reaction resistance<br>(about 2.03 $\Omega$ cm <sup>2</sup> ),<br>bending radius of<br>about 15.6 cm.                        | [86–88]      |
| A thin<br>polycarbonate<br>(PC) film                              | Plate thickness:<br>200 µm, Stainless<br>steel sheet: 50 µm<br>thick, 1*1 mm<br>square mesh with<br>20 nm thick gold<br>film, SC | Ultra-lightness, thin,<br>large bending angel<br>(180° and 360°) and<br>radius of 2 cm. No<br>significant decrease in<br>performance with S-<br>shape or rolled-up<br>state  | [39]         |
| PDMS and carbon<br>fiber powder                                   | Plate: 26*1*42 (w/<br>d/l)   | Tubular, high<br>flexibility, 180°<br>bending angel for 100<br>times.  | [40]         |
| Carbon-fiber-<br>reinforced<br>thermoplastic<br>composite         | Plate: 100*0.4*100<br>mm (w/d/l)   | Excellent corrosion<br>and damage<br>resistance, thermo-<br>plasticity and<br>thermostability.<br>Simple fabrication<br>procedure.   | [83]         |
| Non-woven carbon<br>felt and a cyanate<br>ester-modified<br>epoxy | Plate thickness: 0.96 $\pm$ 0.03 mm  | Reduced thickness and<br>weight, good contact<br>and reduced areal<br>specific resistance.<br>Stable with high<br>pressure and high<br>temperature.  | [84]         |
| Pure Cu foil  | Plate: 15*0.1*20<br>mm (w/d/l), SC   | Low thickness and<br>high electron<br>conductivity.  | [89]         |
| Carbon fibers with<br>Ag NWs                                      | Ti gauze: 280 μm<br>thick, SC  | High electron<br>conductivity and<br>stretchability. Steady<br>performance during<br>100 times repetitive<br>bending.  | [69]         |
| PDMS film   | Hydrogen<br>generator: 75 *2<br>mm (w/d), PC   | High flexibility and low weight.   | [41]         |
| PDMS and Al foil  | SC   | High electron<br>conductivity,<br>decreased ohmic<br>resistance and<br>fabrication resistance<br>$(0.954 \ \Omega \ cm^2 \ and$<br>$0.737 \ \Omega \ cm^2)$ , bending<br>radius:14.1 cm. Stable<br>output during 300 | [90]         |

Table 3 (continued)

| Materials  | Size  | Properties   | Reference |
|--|---|--|-----------|
| PDMS and<br>aluminum electric<br>wires   | Plate: 60*10*60<br>mm (w/d/l), SC   | times repetitive<br>bending.<br>High electron<br>conductivity,<br>decreased ohmic and<br>charge transfer<br>resistances ( $0.738 \Omega$<br>cm <sup>2</sup> and $0.882 \Omega$ cm <sup>2</sup> ).<br>3D-printed flow flied,<br>higher output in<br>bonding | [48]      |
| Polymethyl<br>methacrylate<br>(PMMA), PDMS<br>and copper-clad<br>polyimide (PI)<br>substrate | Plate: 36*0.1*50<br>mm (w/d/l),<br>Stainless steel<br>sheet: 0.7 mm thick | Low thickness and<br>weight, small volume.   | [91]      |

w: width; d: depth; l: length; A: Anode; CA: Cathode; SC: Serpentine Channels; PC: Parallel Channel.

embedded in several bunches of carbon fiber arrays [85]. The metal wire between the carbon fiber bunches provides the required flexibility. The soft and flexible carbon fibers provide the good contact with the electrode in the bent condition.

# 3. Assembly of flexible PEMFC (single cell and stack)

The various components have different features and always show different response to the temperature, humidity, pressure, or bending radius. The interfacial contact between components is usually poor or the gap between them may be formed when the operation condition changes or bending, significantly increasing the contact and mass transport resistance and then decreasing the fuel cell's performance. Therefore, how to effectively assemble the flexible components and ensure them in good contact in bending is essential to achieve a flexible PEMFC with high performance and durability how to effectively assemble the flexible components and ensure them in good contact in bending.

In the conventional PEMFC, the single cell or stack are usually clamped and fixed with bolts and nuts, leading to a certain assembly force on the rigid bipolar plates to make sure the components intimately contact and thus obtain a minimum contact resistance. Different from the conventional PEMFC, the flexible PEMFC is bendable and the components are flexible, light, and thin. A large compression force may lead to the uneven tension distribution or even damage the cells' structure in bending. Therefore, a small assembly force is necessary to guarantee the close contact between the components and the flexibility of PEMFC. Hence, the novel assembly methods should be developed for the flexible PEMFC.

To obtain a flexible single cell, there are commonly two pathways. I) MEA sandwiched between two bipolar plates is directly clamped by paper clips, spring-retainer, or clamps without additional adhesions. II) MEA is adhered to bipolar plates with sealant (e.g., silicone sealant or dichloromethane) or sealant tape; to improve the contact of each component, the assembly could be hot-pressed under a given pressure at a specific temperature or clamped with clips or other clamp.

Since a single fuel cell always has too low power to drive the electronic devices, multiple cells often need to be connected together (in series or in parallel), forming a PEMFC stack. For the conventional PEMFC, the fuel cell stack could be fabricated by stacking the multiple single cells layer by layer. However, this assembly approach is inapplicable for the flexible fuel cell stack as this will cause the loss of flexibility. Therefore, to reserve the bendability of the single fuel cell in the stack and make the stack bendable, multiple single flexible fuel cells are commonly tiled. For example, the flexible single fuel cells are assembled side by side, forming a flexible fuel cell stack (in series) for powering



Fig. 4. (a) The fabrication procedure of bendable fuel cell [69], (b) (b) the fabrication process of flexible fuel cell on COP film, (c) the practical morphology of flexible fuel cell on COP film [38].

LED lights or charging a mobile phone (Fig. 5a) [46]. Park et al. [39] tiled the flexible single PEMFCs together, achieving the stack (in series) to charge smartphone and operate an electric LED fan simultaneously (Fig. 5b). To decline the volume of the flexible PEMFC stack, the assembled stack (in series) could be rolled up or folded up (Fig. 5c).

Although many flexible PEMFC single cell or stack have been fabricated successfully, the effective assembly pathways need to be developed to keep the components intimately contact, especially in bending, which is crucial for improving the performance and stability of the flexible fuel cell.

# 4. Flexible PEMFC system

For the PEMFC systems, the storage or supply of fuel and oxidant are vital in practical application as well. The fuels, e.g.,  $H_2$  or methanol, are usually stored in the high-pressure gasholder, tank, plastic can, and so on in the traditional PEMFC system. Besides, some accessories are required for the supply of fuel and oxidant, e.g., fuel pumps, fuel sensors, or air pumps. These fuel storage pathways and the complex and energy-cost accessories cannot be sufficient for the flexible electronic devices which always require a light, compact, and even flexible fuel reservoir and avoid the use of any accessories. Therefore, the storage or supply of fuel and oxidant need to be advanced for the flexible PEMFC systems.

To fabricate the flexible fuel cell stack, Wang et al. [41] invented a

flexible H<sub>2</sub> generator through preparing a novel bifunctional aerogel catalyst composed of the porous silica aerogel as the substrate and catalyst immobilized in the pore of silica aerogel. This could produce H<sub>2</sub> with formic acid stored in the pore of silica aerogel under the catalytic function of the catalyst. The theoretical energy density of the flexible PEMFC pack (in parallel) containing the flexible H<sub>2</sub> generator (the flexible PEMFC pack (as shown in Fig. 6a) reaches to 722 Wh kg<sup>-1</sup>. To fabricate a safe and flexible DMFC system, Zou et al. [92] developed an agar gel/wood sponge composite material as methanol container, which is flexible and adaptable and shows high absorption rate and capacity of methanol (1.5% agar gel could reserve about 90% of methanol solution at 29.4 kPa).

Due to no need for air pumps, self-breathing PEMFC shows the advantage in the flexible energy suppliers. Besides, it only needs a simply current collector without the demand of the complex flow channel at cathode, which will reduce the cost of the cathodic bipolar plate (Fig. 6 b). Therefore, self-breathing PEMFCs have been widely applied in the field of the flexible PEMFC [38,39,45,46].

# 5. Performance and durability of flexible PEMFC

The performance, especially the performance in bending, and durability, e.g., the stability in bending or during multiple bending, are two important parameters to evaluate the flexible PEMFC. They are



**Fig. 5.** Applications of flexible PEMFCs stacks. (a) 53-LED lights powered with a flexible PEMFC stack, (b) a mobile phone powered by a flexible PEMFC stack [46], (c) The practical photo of a flexible PEMFC stack charging a smartphone [39].



Fig. 6. (a) Fabrication procedure and application of a flexible fuel cell pack including a flexible fuel cell stack and a flexible hydrogen generator [41], (b) structure of the traditional PEMFC, traditional air-breathing PEMFC, and flexible air-breathing PEMFC [46].

influenced by various factors, e.g., but not limited to, the properties of the flexible components, the assembly technique, the types, store or supply of fuel and oxidant, the bend or twist conditions, the number of bends, as well as operation conditions.

Although the flexible PEMFC is in the initial stage, the performance and durability of flexible PEMFC have been improved greatly in the last decade with the advancement of the related materials and techniques. The performance and/or durability of flexible PEMFC reported in the literatures have been summarized in Table 4. Here are a few examples. Chang et al. studied a bendable PEMFC with a performance of 82 mW  $cm^{-2}$  at a bending radius 15.6 cm by using the flow-patterned PDMS film coated Ag NWs as the bipolar plates and employing the bipolar plates with difference thicknesses for anode and cathode [86]. Kang et al. [69] increased the bending durability of GDL by coating the carbon paper with Ag NWs. The as-fabricated flexible PEMFC shows a peak power density of 68.6 mW  $cm^{-2}$  at the bending radius of 47 mm and maintains almost original performance after bending more than 100 times with a bending radius of  $\sim 2$  cm (the power density of fuel cell containing carbon paper GDL decreases 44%) (Fig. 7a-c) [69]. Ning et al. reported a flexible, durable, light PEMFC by inventing a novel flexible composited gas diffusion electrode [46]. It has a high specific volume power density of 5190 W  $L^{-1}$  and its performance only decrease 10.9% after 600 times bends and remains the initial power density after falling down 5 time from 30 m (Fig. 7d-f) [46].

By wrapping up the literatures, it can be found that the flexible PEMFCs usually show lower performance in the flat condition than the bend state. Besides, the performance commonly increases with a decrease of the bending radius in a given range (Fig. 8a and b) [48,69, 86,90,93]. This is mainly because the assembly force commonly used is small, leading to the poor contact between components, however, the compressive stress caused by bend, which increases over the curvature

increases, will improve their contact, beneficial in decreasing the interfacial contact resistance and then resulting in the improved performance (Fig. 8c) [90]. Since the flexible PEMFC will work in different situations, not only in bending but also in the flat state, it is essential for steadily supplying energy to maintain the performance stable, i.e., the performance should be almost constant with or without bends. Therefore, the more effective assembly technique needs to be advanced further in order to allow the components close contact in various conditions.

As we all know, the conventional PEMFCs are the early commercialization stage now. Their power density and durability have reached to 0.86 kW kg<sup>-1</sup> and 4130 h until 2020 [94]. Although there are no specific standards or targets values of performance and durability have been made for flexible PEMFCs, unlike the conventional PEMFCs (the ultimate targets of the system specific power and system durability are 0.9 kW kg<sup>-1</sup>and 8000 h [94], respectively, which were made by US DOE), they need to be improved further in order to realize the practical application of flexible PEMFCs as flexible energy supplier.

#### 6. Conclusions and remarks

In this review, recent progress related to the flexible PEMFC have been examined, including the flexible components, assembly of single cell and stack, the storage or supply of fuel or oxidant, as well as the performance and durability.

To fabricate the flexible PEMFC, various flexible materials have been developed and used to prepare the flexible components for the flexible PEMFC. Except the PFSA- or PBI-based PEMs commonly used, some new polymer-based membranes or silicate-based thin films have been developed and shown excellent flexibility and performance as well as decreased cost. To ensure the CL integrity and good contact with

Examples of flexible PEMFCs' active area, test condition, performance and durability.

| Fuel           | Active<br>area           | Test condition   | Performance   | Durability   | Ref. |
|----------------|--------------------------|--|---|--|------|
| Methanol       | 10*10<br>mm <sup>2</sup> | 60 °C,<br>methanol:<br>0.14 mL<br>min <sup>-1</sup> ,1 M;<br>humidified O <sub>2</sub> :<br>8 mL min <sup>-1</sup>                                   | PPD: 19.0 mW<br>cm <sup>-2</sup>  | /  | [55] |
| Methanol       | 2.5<br>cm <sup>2</sup>   | Methanol: 1<br>mL, room<br>temperature   | Peak power:<br>38 mW  | The power<br>decreased<br>about 7.8%<br>after 100<br>times<br>bending with<br>180°   | [40] |
| Methanol       | /                        | Methanol: 5 M,<br>30 °C; bending<br>angles: 0°, 90°,<br>180°   | PPD: 8.86 mW cm <sup>-2</sup>   | 54 h at 0.5<br>mA cm <sup>-2</sup>   | [57] |
| Methanol       | 2*2<br>cm <sup>2</sup>   | Methanol: 2.5<br>M; bending<br>angle: 75°  | PPD: 20.2 mW cm <sup>-2</sup>   | Voltage<br>decreases<br><10% after<br>50 times<br>bending                            | [44] |
| Methanol       | 0.096<br>cm <sup>2</sup> | Methanol: 3 M,<br>0.6 mL min <sup>-1</sup><br>at 25 °C; air:<br>RH 65%   | PPD: 10 mW<br>cm <sup>-2</sup>  | /  | [91] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | $H_2$ : humidified<br>at 20 °C with<br>50 sccm   | PPD: 20.5 mW<br>cm <sup>-2</sup> ; OCV:<br>1.0 V                        | 30% decrease<br>at a bending<br>curvature of<br>1.8 m <sup>-1</sup> .                | [45] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | Humid $H_2$ : 0.5<br>L min <sup>-1</sup> ;<br>humid air: 1.0<br>L min <sup>-1</sup> ;<br>bending<br>radius: 15.6 cm                                  | PPD: 72<br>(symmetric)<br>and 82 mW<br>cm <sup>-2</sup><br>(asymmetric) | 1  | [86] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | Humid H <sub>2</sub> : 0.5<br>L min <sup><math>-1</math></sup> ;<br>humid air: 1.0<br>L min <sup><math>-1</math></sup> ;<br>bending<br>radius: 15 cm | PD:117 mW<br>cm <sup>-2</sup>   | /  | [87] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | H <sub>2</sub> and air: RH<br>94%, 3.33 cm <sup>3</sup><br>s <sup>-1</sup> at 25 °C;<br>bending<br>radius: 83, 58,<br>47 mm                          | PPD: 35–70<br>mW cm <sup>-2</sup>                                       | Decreasing<br>18–40 mW<br>cm <sup>-2</sup> after<br>repetitive<br>bending            | [36] |
| H <sub>2</sub> | 1*1<br>cm <sup>2</sup>   | $H_2$ : 15 mL<br>min <sup>-1</sup> , 20 °C,<br>atmosphere<br>pressure  | Specific<br>volume PD:<br>5.19 kW L <sup>-1</sup>                       | Specific<br>volume PD<br>maintained<br>89.1% after<br>600 times<br>bending           | [46] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | H <sub>2</sub> and air:<br>94% RH, 200<br>cm <sup>3</sup> min <sup>-1</sup> at<br>25 °C; bending<br>radius: 2 cm                                     | Total power:<br>508 mW; Mass<br>PD: 0.228 W<br>g <sup>-1</sup>          | Power<br>decreases<br><10% after<br>200 times<br>bending                             | [39] |
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup>   | Humidified $H_2$<br>and air: 100<br>and 300 cm <sup>3</sup><br>min <sup>-1</sup> ;<br>bending<br>radius: 83, 58,<br>47 mm                            | PD: 68.5 mW<br>cm <sup>-2</sup>   | No decrease<br>after >100<br>times<br>bending with<br>a bending<br>radius of 2<br>cm | [69] |
| H <sub>2</sub> | 5 cm <sup>2</sup>        | $\rm H_2~12.5~mL$ $\rm min^{-1}$   | Energy<br>density: 135.9<br>W h kg <sup>-1</sup>                        | /  | [41] |
| H <sub>2</sub> | 5*1<br>cm <sup>2</sup>   | $\rm H_2: 15~mL$ $\rm min^{-1}$ at 20 $^\circ \rm C$   | Specific<br>volume PD:<br>15 600 W $L^{-1}$ ,                           | /  | [47] |

Table 4 (continued)

| Fuel           | Active<br>area         | Test condition  | Performance  | Durability  | Ref. |
|----------------|------------------------|---|--|---|------|
| H <sub>2</sub> | 3*3<br>cm <sup>2</sup> | $H_2$ and air: RH<br>94%, 0.2 L<br>min <sup>-1</sup> , 25 °C;<br>bending<br>radius: 14.1 cm                     | Specific<br>weight PD:<br>9660 W kg <sup>-1</sup><br>PD: 88.7 mW<br>cm <sup>-2</sup>                     | Power<br>reduces after<br>300 times<br>bending and<br>no decrease<br>with 100<br>times of | [90] |
| H <sub>2</sub> | 5 cm <sup>2</sup>      | $H_2$ and air: 200<br>sccm, RH<br>100% at room<br>temperature;<br>bending<br>curvature:<br>20.2 m <sup>-1</sup> | PPD: 87.1 mW<br>cm <sup>-2</sup><br>(maximum<br>bending) and<br>30.2 mW cm <sup>-2</sup><br>(no bending) | No<br>significant<br>decrease<br>after 100 or<br>200 times<br>bending                     | [48] |

PDD: peak power density.

membrane and GDL, the flexible CL based on the CFC as the support was studied, which exhibited better performance than the Pt/C particlesbased CL in bending. The flexible GDLs mainly include CFC-based GDLs, modified CFC-based GDLs, or CNT-based composite GDLs. For the flexible PEMFC, the light, thin, and flexible bipolar plates is essential, which could be prepared by using flexible polymer film with flow channel pattens as substrate and depositing a thin metal layer at current collector.

These flexible components would be assembled together usually by clips, spring-retainer, or clamps with or without adhesion. These assembly pathways will lead to a relatively small assembly force, which is necessary to ensure the sufficient contact between the flexible components and the flexibility of PEMFC. To achieve the flexible PEMFC stack, multiple flexible single cells are commonly tiled one by one, different from the conventional PEMFC stack which is generally fabricated by stacking many single cells layer by layer.

For the flexible PEMFC system, the light, compact, safe, and even flexible fuel store as well as simple accessories for the supply of fuel or oxidant are vital. Therefore, some porous materials, e.g., porous silica aerogel- or sponge-based materials, have been used to store liquid fuel. Self-breathing is commonly used in the flexible PEMFC, due to no demand for any air supply accessories.

With the development of various flexible components, assembly technique, and store or supply of fuel and oxidant, the performance and durability of flexible PEMFC have been significantly improved. However, the flexible PEMFC is still at an early stage and there still exists a long way needs to go before applying in the practical flexible electronic devices. The power density, mechanical stability, and durability need to be improved further by developing advanced materials or introducing novel preparation methods for the flexible components to increase the flexibility of each component and their contact, optimizing the assembly techniques for the single cell or stack to make sure the excellent contact between components in bending, and exploiting the safe, adaptable, durable, and flexible fuel storage. For example, chemical deposition of Pt-based catalysts may be appliable to prepare the CL for the flexible PEMFC, due to the increased adhesive strength of the prepared CLs to PEM, improved mass transport resulting from the ultrathin thickness, and high Pt utilization [95,96], which will be beneficial for improving the flexibility of CL and its contact with PEM in bending. Besides, electrospinning is an effective and relatively easy method to prepare the fine fiber, and has been used to fabricate the fibrous membranes [97], which may be introduced to prepare the flexible components for the flexible PEMFC.

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**Fig. 7.** (a) Polarization curves and (b) peak power density of flexible PEMFC containing the novel GDL composed of Ag NWs coated on carbon paper GDLs as well as (c) the peak power density of the PEMFC containing the carbon paper-based GDL at various bending radius before and after being bent repeatedly with a bending radius of  $\sim$ 2 cm for> 100 times [69], (d) the performance of the flexible and traditional PEMFC, (e) the photo of the flexible PEMFC bended by a bending angel of 50°, (f) I-t curves of the novel flexible PEMFC before and after bending 600 times [46].



**Fig. 8.** The performance (a) and impedance plot at 0.5 V (b) of the flexible PEMFC with different bending curvatures [48], (c) the morphology and compress stress distribution of the flexible PEMFC at different bending radius (R = 21.3, 16.5, and 14.1 cm) [90].

# Declaration of competing interest

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.

## Data availability

Data will be made available on request.

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