



Electrode for proton exchange membrane fuel cells: A review

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ABSTRACT

The electrode is the key component of the membrane electrode assembly (MEA) of proton exchange membrane fuel cells (PEMFCs). The electrochemical reaction of hydrogen (fuel) and oxygen that transform into water and electrical energy occurs at the catalyst site. Attempts to improve the performance and durability of electrodes have sought to overcome the challenges arising from utilizing PEMFCs as an efficient and competitive energy source. To accomplish this goal and to solve the problems related to using PEMFC electrodes, the structure and function of each component and the manufacturing method must be comprehensively understood, and the electrode performance and durability of the cell must be characterized. Therefore, in this paper, we discuss the components, preparation, functions and performance of the electrodes used in PEMFCs. This review aims to provide comprehensive information regarding PEMFC electrodes.

1. Introduction

The proton exchange membrane fuel cell (PEMFC) is the most promising candidate for renewable and sustainable energy conversion devices due to its zero CO₂ emissions. This technology is expected to be an efficient energy source that is free of pollutants, and it has a high energy density compared to conventional energy sources. Considering the global energy demand from the human population, the threat of fossil fuel shortage is a major concern. Therefore, extensive research and development have been focused on renewable energy sources as well as reducing of CO₂ emissions. PEMFCs are widely used as clean energy conversion devices, especially in vehicles and in stationary and portable power generation systems. Because of their power density, energy efficiency, lack of pollutant emissions and low operating temperature [1–7]. However, the commercialization success of PEMFCs depends on their ability to demonstrate optimal fuel to electricity conversion with a high current density [8].

In PEMFCs, hydrogen (H₂) gas feed at the anode is oxidized to

release protons and electrons. The electrons generate electricity at the external circuit connected to the load. The hydrogen ions (protons) migrate through the polymer electrolyte (proton exchange membrane) to recombine with electrons and oxygen to produce water at the cathode. There are two important key factors that slow the development and commercialization of PEMFCs: cost and durability. The high cost of PEMFCs is largely due to the use of noble metals (platinum) as catalysts, which accounts for 55% of all PEMFC manufacturing costs [9]. While researchers are currently seeking alternative catalysts, platinum is still the most commonly used catalyst because it is very effective and has a high chemical stability, exchange current density and work function [10]; however, platinum is expensive and easily poisoned [11]. Therefore, the development of fuel cells is directed toward developing new electrodes and reducing the use of platinum catalysts [11,12], perhaps even by replacing platinum with a non-noble metal, such as Co, in the anode [9]. Although their performance is not as high as that of a platinum catalyst, Co catalysts in PEMFCs may decrease the cost of PEMFC manufacturing. The durability of a PEMFC varies

Abbreviations: AB, acetylene black; AST, accelerate stress test; BL, backing layer; CCM, catalyst coated membrane; CFGDL, carbon-filled gas diffusion layer; CL, catalyst layer; CV, cyclic voltammetry; CNF, carbon nanofibers; CNT, carbon nanotubes; DOE, department of energy; DSO, dimethyl silicon oil; ECSA, electrochemical active surface area; EIS, electrochemical impedance spectroscopy; EDX, energy dispersive X-ray; FE, four electrode; FPE, four point electrode; FEP, fluorinated ethylene propylene; FCTESTNET, fuel cell testing and standardization thematic network; GDB, gas diffusion backing; GDL, gas diffusion layer; GDM, gas diffusion media; HALS, hindered amine light stabilizers; HDP, homogeneous deposition precipitation; IBAD, ion beam assisted deposition; LSV, linear sweep voltammetry; MPL, macroporous layer; MEA, membrane electrode assembly; NIOSH, national institute of occupational safety and health; OCV, open circuit voltage; ORR, oxygen reduction reaction; PBI, polybenzimidazole; PFSA, perfluorosulfonic acid; PTCDA, perylenetetra-carboxylic dianhydride; PPA, polyphosphoric acid; PTFE, polytetrafluoroethylene; PVDF, polyvinylidene fluoride; PED, preferential pulsed electrodeposition; PEMFC, proton exchange membrane fuel cells; SEM, scanning electron microscope; TEM, transmission electron microscopy; XPS, X-ray photoelectron microscopy; XRD, X-ray powder diffraction

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according to its application mode. In 2015, the DOE (Department of Energy; USA) declared a lifetime target of 5000 h for vehicle energy systems and 40,000 h for stationary power systems. However, the actual PEMFC technology achievements to date are 1700 and 10,000 h, respectively [13]. Some durability test standards have been developed by the DOE and the Fuel Cell Testing and Standardization Thematic Network (FCTESTNET). In addition to endurance tests, accelerated tests are also used for conditioning or fuel cell incubation to achieve optimum conditions. Depending on the membrane electrode assembly (MEA) type, conditioning can require several hours or even days. Some theoretical aspects and considerations for conditioning include activating and cleaning the catalyst from impurities, cleaning the remaining catalysts and membranes from the dry and wet membrane electrode structures and activating the Nafion ionomer as a proton conductor [14]. A commonly used method involves heating the cell without a gas supply followed by applying a shortcut connection between the anode and cathode for a few minutes. After the hydrogen and oxygen gas flow and the voltage reach 1.0 V OCV (open circuit voltage), the fuel cell is operated under a load for 6 h. The conditioning process is considered complete when the voltage changes by < 1 mV.

Discussing the structure, components, manufacturing, and characterization methods of electrodes is very important for highlighting and solving the problems related to PEMFC electrodes. Researchers have attempted to investigate electrodes to improve their performance and durability, but a limited number of review papers exist regarding all aspects of the electrode. For example, Litster et al. [12], focused on the catalyst layer (CL) in a very constructive manner but presented a limited discussion on the gas diffusion layer (GDL). In fact, the current research has focused on the durability of electrode materials, but the degradation of the PEMFC cannot be avoided. However, the degradation can be minimized by understanding the degradation mechanism and cell components [14].

This paper intends to review the prior studies on electrodes regarding the function of the catalyst components, manufacturing and characterization methods, electrode performance and cell durability to provide a reference for future electrode researchers. Moreover, the aim of this work is to provide a critical review as well as a comprehensive discussion on the electrode synthesis process, the function of electrode components in terms of cell performances, and electrode system management that could be effective for PEMFC researchers.

2. Electrode structure and components

Generally, electrodes are constructed with three layers: the backing layer (BL), gas diffusion layer (GDL) and catalyst layer (CL) [15]. Some researchers consider that an electrode is composed of two layers, the GDL and CL, while the GDL consists of two layers, namely, a microporous layer (MPL) of carbon powder and hydrophobic / hydrophilic agents and a backing layer composed of carbon paper or carbon cloth. The electrode structure according to Park [16,17] is presented in Fig. 1.

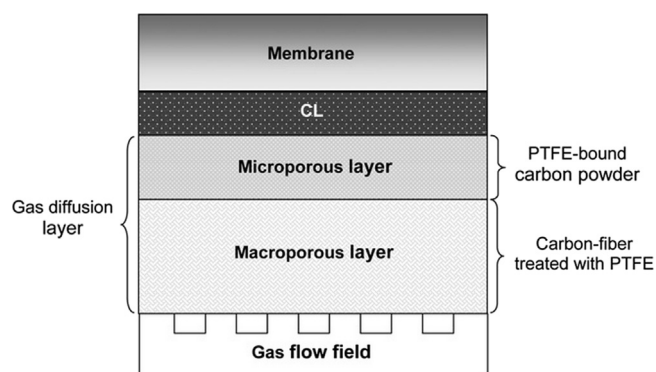


Fig. 1. Basic structure of PEMFC electrode (Reprint permission from Ref. [16]).

During electrode preparation, the GDL (GDL = BL + MPL) and CL should be carefully considered. The MPL that supports the catalyst layer should have the following features: high electrical conductivity, good interactions as a catalyst support, a high surface area, the ability to repel water and prevent flooding, corrosion resistance and the ability to easily restore catalyst functions [10]. The GDL and CL can be prepared using several methods, including casting, painting and brushing [4,19]; injection molding [5,20]; impregnation [11,21]; spraying [17,22]; thin layer deposition; using a catalyst coated membrane (CCM) [23,24]; and electro deposition [25,26].

In addition to serving as electrochemical reaction centers, electrodes must continuously allow electrons to flow from anode to the cathode side. In accordance with this function, an electrode must fulfill three aspects. First, the electrode must have appropriate pores for the reactants. Second, the electrode should contain a chemical catalyst to break the bonds of the fuel to form more reactive ions. Third, the electrode must conduct electrons to the external circuit. A proper PEMFC electrode structure has three phases: the gas phase reactant, catalyst particles and ionic conductors. The catalyst particles must directly contact the electron conductor. Electrons will flow through the carbon on which the catalyst particles are immobilized [27].

In MEA, the electrodes included an anode and a cathode. The anode is the site where the hydrogen gas (feed) is oxidized and split into protons (H^+) and electrons. Protons can pass through the proton exchange membrane to the cathode site. In contrast, electrons flow through the outer circuit to reach the cathode, thus generating electrical energy. Meanwhile, oxygen from the air is reduced at the cathode to produce water (H_2O). The electrochemical reaction that occurs at the cathode is $4H^+ + O_2 + 4e^- \rightarrow 2H_2O$ as shown in Fig. 2.

Many electrode studies have focused on the electrochemical reactions that occur on the cathode [21,28–34]. For example, the oxygen reduction reaction (ORR) is slower than the hydrogen oxidation reaction [35], requires a high catalyst content to maintain a tolerable response speed [32], requires a large over potential and causes the loss of 80% of the cell voltage [36]. In addition, PEMFC applications (especially for vehicles) require more oxygen from the air at the cathode. Since the oxygen content in air is approximately 20%, more oxidant intake is required, and the catalyst must have a high affinity for oxygen. Due to the increased oxidant flow, more impurities are introduced that can cause catalyst poisoning. Yu et al. [36,37] comprehensively discussed the catalyst activity and durability of Pt/C catalysts for PEMFC cathodes. Moreover, many researchers [38–42] have focused on catalyst poisoning by CO and H_2S gases at the anode. Meland et al. [43]

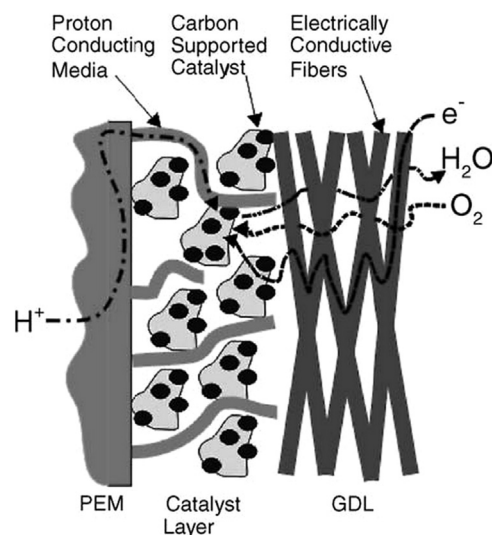


Fig. 2. Electrochemical processes forming water at the cathode (Reprint permission from Ref. [12]).

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