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Electrocatalysts for low temperature fuel cells

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ABSTRACT

Low temperature fuel cells technologies are currently shifting very fast from fundamental research to real growth. The development of electrocatalysts plays a vital role in the electrocatalytic reactions involved in these devices, because the catalyst determines the overall reaction efficiency, durability and cost. This article review progress in the research and development of electrocatalysts for low temperature fuel cells technologies, with especial attention in the contribution of our research teams over the last 15 years or so. The intensive research efforts in reducing or replacing Pt-based electrodes in fuel cells have been focus in the use of carbon nanomaterials as electrocatalytic supports, including carbon nanostructures tailored by surface modification or building in particular dopants/defects. Recent research effort has also led to the use of electronic conductivity noncarbon support materials. In addition, carbon-composite materials are proving to be a robust, inexpensive and active electrocatalysts, where the synergetic effect between the carbon nanomaterials and the ceramic or polymer nanostructures can lead to a superior electrocatalytic performance and durability for low temperature fuel cells. Perspectives on these catalysts and possible pathways to address current remaining challenges are also discussed.

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1. Introduction

Low temperature fuel cells are among the most promising candidates for the reliable and efficient conversion of hydrogen or alcohol into electric power in automotive, distributed power generation, and portable electronic applications on a large scale [1–5]. The one-step (from chemical to electrical energy) nature of the electrochemical process, in comparison to the multi-step (e.g. from chemical to thermal to mechanical to electrical energy) processes involved in combustion-based heat engines, offers several unique advantages, making them one of the strongest candidates to be the energy conversion device of the future [2,5]. Since the development of perfluorinated cation exchange membranes, such as Nafion[®], proton exchange membranes have dominated as the electrolyte for polymer electrolyte membranes fuel cells (PEMFCs) and direct alcohol fuel cells (DAFCs). This electrolyte conducts protons from the anode to the cathode, where the oxygen is reduced to obtain water (Fig. 1).

However, some major obstacles have restrained more rapid development and applications of PEM fuel cells, like the high cost of membranes and noble metal catalysts, and the relatively low activ-

ity and complex reaction mechanism, predominantly during the oxygen reduction reaction (ORR) at the cathode. Anion exchange membrane fuel cells (AEMFCs), which oxidize fuel directly on the anode in alkaline media, have attracted attention again in recent years because of the potential solutions to overcome the problems in the PEM fuel cells. As polymer electrolyte membrane fuel cells, anion exchange membrane fuel cells (AEMFCs) have similar structures with PEMFCs [6,7]. However, the most important difference between them lies on that AEMFC uses an alkaline polymer electrolyte membrane, which can conduct OH⁻ anions. The AEMFCs present several potential advantages compared to its acidic Nafion-based counterparts, including much improved kinetics of electrochemical reactions, materials stability, and easy water management. Thus, AEMFCs may hold great potential to become the most efficient low-temperature fuel cell in the near future, providing opportunity to use a host of non-platinum group metal catalysts [8–11].

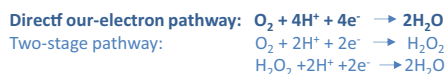
The electrocatalytic processes involved in the low temperature fuel cells can be achieved at temperature lower than 120 °C and atmospheric pressure, and the reaction rate and selectivity are possibly controllable by using an appropriate electrocatalyst. Therefore, the electrocatalyst play a crucial role in this technology [12]. To play its essential function, an electrocatalyst needs to provide high intrinsic activities for the electrochemical oxidation of a fuel at the anode, whether this is hydrogen or alcohol (methanol,

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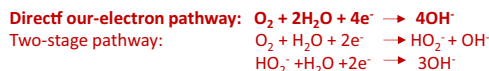
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Oxygen Reduction Reaction

Acid electrolyte



Alkaline electrolyte



Scheme 1. Reaction pathways of oxygen reduction reaction in both acid and alkaline electrolytes.

ethanol), and for the electrochemical reduction of oxygen at the cathode. Other requirements include high electrical conductivity, appropriate physical and electrical contact with the ionomer, suitable reactant/product gas access/exit, and high stability in the highly corrosive working medium [13,14]. To ensure that a fuel cell delivers maximum efficiency, both electrode reactions need to take place close to their thermodynamic potential as possible.

The oxygen reduction reaction (ORR) is the primary electrochemical reaction occurring at the cathode of low temperature fuel cells, and is central to these technologies because it is the main source of efficiency losses [2,15]. Oxygen is converted to water at the cathode via the oxygen reduction reaction with two possible reaction routes: the direct four-electron pathway and the two-stage pathway with two electrons transferred in each stage and a peroxide intermediate (Scheme 1)

Highly dispersed metal catalysts, commonly platinum or platinum-based catalysts, supported on carbon-based materials, are used as electrocatalysts for oxidation and reduction reactions in PEMFCs fuelled with hydrogen or small organic molecules such as methanol or ethanol. The intrinsically high cost of Pt, however, is a serious roadblock in the large-scale roll-out of ORR-involving technologies since the catalyst (i.e. Pt) usually accounts to around 14% of the total cost [9]. This has led to a very high interest for inexpensive catalysts alternative to Pt (e.g. carbon and non-noble transition metals) that were introduced as either a supporting material or as an alloying element to reduce the usage of Pt [16–24].

In the past decade, the advances of nanotechnology and material science have opened new ways to develop novel electrocatalysts for fuel cells [25,26]. The use of nanomaterials in fuel cell systems can significantly improve the electrocatalytic performance to achieve high energy density and high power density while reducing the manufacturing cost. It is the aim of this article to review recent progress on nanostructured electrocatalysts for application to low temperature fuel cells, with the special attention on the contribution of our research teams in the advancement and study of electrocatalysts based on novel nanostructure carbons and noncarbon materials.

2. Carbon-based nanomaterials as electrocatalysts supports

Over past decades, carbon materials have been widely used as electrodes in low temperature fuel cells, not only because they have extraordinary physical properties such as high surface areas and good electronic conductivity, but also owing to their abundance, processibility and environmental friendliness [27–30].

In nature, carbon is found free in three main allotropic forms: amorphous carbon, graphite and diamond [31]. Depending on distinct types of crystal structures, carbon atoms can form a variety of allotropes endowed with different properties [27], as shown in Fig. 2. The simplest graphite material is a two-dimensional “graphene” sheet, which is essentially a very large polyaromatic hydrocarbon. Graphite stands for typical sp^2 hybridization. Diamond represents sp^3 hybridized carbon materials exhibiting different electrochemical properties from sp^2 -related carbon materials. The most common fullerenes used as an electrode material are carbon nanotubes, which amount to single or multiple layer of graphene sheets “rolled up” to form tubes of varying diameter, length, and termination [32].

In general, sp^2 -hybridized carbon materials exhibit a high diversity in crystallinity, morphology, porosity and texture. These structural parameters play a crucial role in determining and optimizing the electrochemical performance when carbons are used as electrodes [33]. On the other hand, the completely sp^3 hybridized, tetrahedral bonding of diamond results in both its hardness and low electrical conductivity, with the latter property making single- or polycrystalline diamond uninteresting as an electrode material. However, the introduction of heteroatoms, as boron or nitrogen, into diamond structure can increase the conductivity of diamond

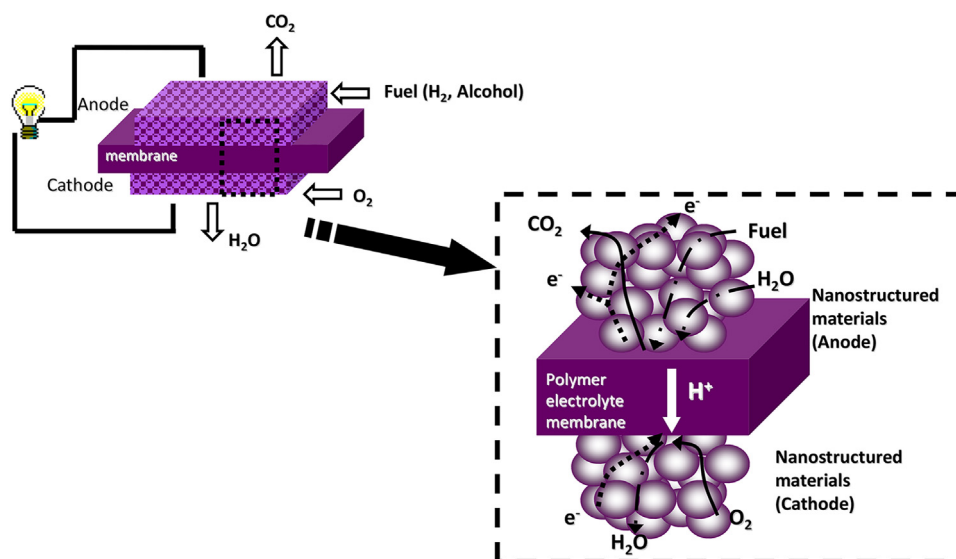


Fig. 1. Schematic illustration of a polymeric electrolyte membrane fuel cell (PEMFC) and/or direct alcohol fuel cell (DAFC).

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