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Review (Special Issue on Electrocatalysis Transformation)

A review of applications of poly(diallyldimethyl ammonium chloride) in polymer membrane fuel cells: From nanoparticles to support materials

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ARTICLE INFO

Article history:

Received 11 March 2016

Accepted 26 April 2016

Published 5 July 2016

Keywords:

Polyelectrolyte

Nanoparticle

Support

Controllable synthesis

Functionalization

ABSTRACT

Polymer membrane fuel cells represent important sustainable energy devices because their operation involves zero emissions and low temperatures and their components exhibit low toxicity. Among the various components of such cells, the electrocatalyst plays the vital role of enhancing the output power density and/or working lifetime. Over the past several decades, numerous strategies have been proposed to address the challenges of electrocatalyst activity and/or durability. Herein, we review the applications of polyelectrolytes in electrocatalysts, including the enhancement of both catalytic nanoparticles and support materials. The effects of polyelectrolytes with regard to controlling the size, composition and morphology of catalytic nanoparticles, as well as the modification of support materials were summarized. In addition, the future possibilities for the research and development of polyelectrolytes in the field of catalyst design and synthesis are discussed.

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1. Introduction

Polymer membrane fuel cells (PMFCs) are a promising means of addressing the present challenges of environmental pollution and energy shortages. Both anode and cathode electrocatalysts, typically composed of precious metals, are key components of PMFCs and play central roles in enhancing the output power density and extending the working lifetime [1–4]. It is notable that the performance of precious metal nanoparticles is limited primarily by several controllable parameters, including size distribution [5], composition [6–9] and morphology (a factor related to preferential facets) [10–13]. Each of these parameters is critical to the electrocatalysis process and

significantly affects the reaction pathways and/or activities [14–19]. However, the size, composition and morphology of metal nanoparticles are difficult to tune during synthesis due to the nature of the precursors that are employed, and so various controlling agents, such as polymers and polyols, are often used. The as-prepared electrocatalysts also tend to suffer from agglomeration, dissolution, detachment of catalytic nanoparticles and, especially, severe corrosion of support materials during long-term operation [20–29]. As a result of these phenomena, PMFCs incorporating these materials may not meet the requirements for commercialization. At present, carbon materials such as 1D carbon nanotubes (CNTs) and fibers [30–38], 2D graphene [39–49] and 3D carbon [50–61] are still the most

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This work is supported by the National Natural Science Foundation of China (21276058, 21433003) and the State Key Laboratory of Urban Water Resource and Environment (Harbin Institute of Technology) (2014DX10).

DOI: 10.1016/S1872-2067(16)62480-4 | <http://www.sciencedirect.com/science/journal/18722067> | Chin. J. Catal., Vol. 37, No. 7, July 2016

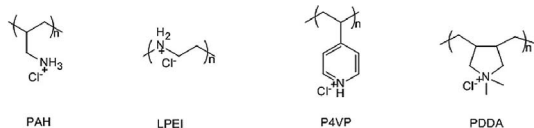
widely used supports due to their balanced performance, although they undergo more severe corrosion under fuel cell operating conditions [25–29,62–68] compared with other novel supports such as metal oxides [69–75] and metal carbides [76–82]. Graphitization is efficient at enhancing the stability of carbon materials, and can significantly reduce surface defects, although it also results in a decrease in the loading sites available for nanoparticles. Therefore, the development of catalytic nanoparticles (commonly precious metals and their alloys) exhibiting high activity and stability, as well as support materials with high stability, electrical conductivity and strong interactions with nanoparticles, is urgently required [3,83–93].

Polyelectrolytes have the potential to address the above challenges, and so are widely used in fabricating membrane electrode assembly (MEA) devices based on layer-by-layer self-assembly. Several excellent reviews of this subject have been published [94,95] in addition to other reports regarding catalytic nanoparticles and/or support materials [96–105]. However, only minimal attention has been paid to the effects of polyelectrolytes with regard to tuning catalytic nanoparticles and functionalizing support materials. Herein, we focus on the applications of polyelectrolytes, particularly poly(diallyldimethyl ammonium chloride) (PDDA), in developing novel catalytic nanoparticles and carbon support materials with remarkably enhanced performance.

2. Polyelectrolytes

Polyelectrolytes, or polymeric electrolytes, are generally recognized as helpful agents in the design and synthesis of

Polycations



Polyanions

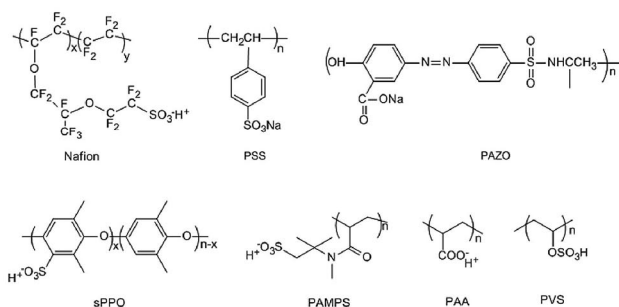


Fig. 1. Polycations: poly(allylamine hydrochloride) (PAH), linear poly(ethylene imine) (LPEI), poly(4-vinylpyridine) (P4VP), poly(diallyldimethyl ammonium chloride) (PDDA). Polyanions: Nafion, poly(sodium styrene sulfonate) (PSS), poly(1-(4-(3-carboxy-4-hydroxyphenylazo)benzene sulfonamido)-1,2-ethanediy), sodium salt) (PAZO), sulfonated poly(2,6-dimethyl-1,4-phenylene oxide) (sPPO), poly(2-acrylamido-2-methyl-1-propanesulfonic acid) (PAMPS), poly(acrylic acid) (PAA), poly(vinyl sulfate) (PVS). Reproduced with permission from Ref. [94]. Copyright (2012) Royal Society of Chemistry.

electrocatalysts, due to their unique properties and/or charged states in aqueous solutions. Based on their charge states, polyelectrolytes can be classified as polycations, polyanions or polyampholytes, among which the polycations and polyanions are potentially useful in the field of PMFCs (Fig. 1) [94].

Polyelectrolytes in aqueous solution typically dissociate into long polymer chains and ions having opposite charges. Taking PDDA as an example, the polymer dissociates into Cl^- ions and positively charged polymer chains (Fig. 2). Within the PDDA chains, the positively charged regions are hydrophilic amino groups, while the remainder of each chain is composed of hydrophobic hydrocarbyl regions (Fig. 2(a)). These charged polymer chains can form micelles, the shape and size of which depend on the concentration of the polymer. When the PDDA concentration is high, positively charged hydrophilic groups move outward to form a spherical micelle structure with exposed amino groups (Route I, Fig. 2(b)). At lower concentrations, the polymer chains will form a twisted structure (Route II, Fig. 2(c)) [106]. In addition, long chains with functional groups can adsorb on the surfaces of both catalytic nanoparticles and support materials, which is the basis for using polyelectrolytes to modify such materials. Interestingly, the modified nanoparticles and supports can be either positively or negatively charged by the polymer chains, depending on the polyelectrolyte type. Oppositely charged regions will tend to gather together based on electrostatic attraction, which leads to numerous possibilities with regard to synthesizing novel electrocatalysts and MEAs. The various uses of polyelectrolytes are all based on these unique properties.

It should be emphasized that most reports of polyelectrolyte in PMFCs are associated with layer-by-layer self-assembly, which involves non-covalent integration between two oppositely charged regions to produce highly ordered structures on the MEA level [94,95], rather than on the catalyst level. However, as researchers have developed a more profound understanding of polyelectrolytes, more and more investigations have focused on the application of polyelectrolytes to catalyst synthesis. Although the main object of our attention is PDDA at present, there is still much to be learned in this field regarding

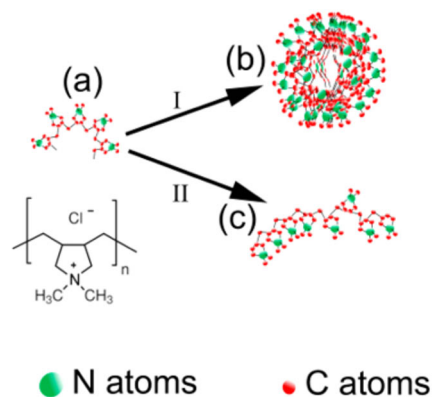


Fig. 2. Illustrations of a PDDA molecular chain and the PDDA monomer (a), the micelle structure at high PDDA concentrations (b), a twisted, long PDDA chain at low concentrations (c). Reproduced with permission from Ref. [106]. Copyright (2014) American Chemical Society.

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