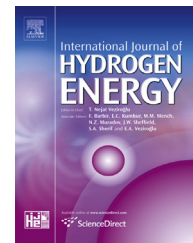




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Novel anion exchange membranes based on pyridinium groups and fluoroacrylate for alkaline anion exchange membrane fuel cells

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ABSTRACT

Novel fluorinated anion exchange membranes with pyridinium salt functionalized groups for alkaline anion exchange membrane fuel cells have been prepared and characterized. These membranes have shown a combination of good thermal stabilities, high ionic conductivities, and excellent chemical stabilities. The ionic conductivity of the membranes can be as high as $2.7 \times 10^{-2} \text{ S cm}^{-1}$ in deionized water at 30 °C. An alkaline H_2/O_2 fuel cell employing the resulting membrane was assembled and revealed a maximum power density of 124.8 mW cm^{-2} at 60 °C. The preliminary performances have demonstrated their potential as electrolytes for alkaline anion exchange membrane fuel cells.

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Introduction

Fuel cell power generation technology is now widely regarded as a potential approach to solve the energy crisis owing to its high energy conversion efficiency and lower pollution [1–3]. The development of polymer electrolyte membrane is very important in the present energy field, not only fuel cell but also other devices [4–6]. Although the proton exchange membrane fuel cell (PEMFC) has significant performance and promising prospects, its commercialization has been hampered by several problems, such as prohibitive cost, sluggish reaction kinetics in acidic condition, fuel (e.g. methanol, ethanol, etc.) crossover and complex water management [7–9]. Unlike PEMFC, the alkaline anion exchange

membrane fuel cell (AEMFC) using anion exchange membrane (AEM) as the polymer electrolyte membrane operates under alkaline condition, which has many merits in cathode kinetics, ohmic polarization and water management [10,11]. The development of AEMFC can lead to higher efficiencies and enable the use of non-noble metal catalysts, such as silver, nickel and palladium, considerably reducing the cost of the cell. In an AEMFC, water is produced at the anode and consumed at the cathode (that is, the consumption and production of water in the reaction processes are significantly different from PEMFC), thus water management can be potentially simplified [12]. Therefore, alkaline anion exchange membrane fuel cell exhibits unpredictable developing potential and has been extensively attracting researchers' interest [13–15].

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The polymer electrolyte membrane is very important in the present energy field, as one of the key components of an AEMFC system, anion exchange membrane (AEM) has been actively studied and a lot of research achievements have been made. Hong et al. obtained PEI-CPP membrane by amination reaction [16]. Zhou et al. synthesized cross-linked anion conductive polysulfone membranes via epoxy functionalities with good physical and chemical stability [17]. The research team of Tongwen Xu developed the organic–inorganic hybrid alkaline membranes based on poly(2,6-dimethyl-1,4-phenylene oxide) (PPO) [18]. Abuin et al. prepared a quaternary ammonium polymer by chloromethylation and amination of a commercial polysulfone which exhibits excellent mechanical properties [19]. In our laboratory, a series of anion exchange membranes based on alkyl quaternary ammonium salts and imidazolium salts have been synthesized and characterized [20–22]. Despite considerable progress has been made by the international scholars over the past five years, still many problems need to be solved before the AEMs can be successfully applied to the fuel cell. Development of novel AEMs with high performance is urgently needed.

Aiming to explore AEMs with sufficient mechanical strength, good stability, suitable ionic conductivity, as well as high cell performance, polymerizable 4-Vinyl pyridine and fluoroacrylate have been using as monomers for polymerization in our works. Different from conventional quaternary ammonium compound, the big π bond conjugated system of the pyridine makes the pyridinium salt possess better thermal and chemical stabilities. Due to the strong water affinity of pyridine group, fluoroacrylate was introduced to tune the hydrophilic–lipophilic balance of the polymers. Furthermore, introduction of fluorine into polymer backbone has been one of the most widely adopted techniques for structure modification leading to substantial property enhancement [23,24]. In order to improve the film forming properties, a small percentage of acrylate was also used as the third monomer. In this paper, we originally report the synthesis and characterization of novel anion exchange membranes based on the copolymer of hexafluorobutyl methacrylate (HFMA), 4-vinyl pyridine (4-VP) and butyl methacrylate (BMA). The copolymer with pendant pyridinium groups has been prepared by free radical copolymerization which is an effective method to control the properties of the reaction products by adjusting mole ratio of the monomers. The chemical structures and the some physicochemical properties of the membranes, such as thermal stability, water uptake, IEC, ion conductivity and chemical stability were also investigated. The single fuel cell using the synthesized membrane was tested at 60 °C and achieved a peak power density of 124.8 mW cm⁻².

Experimental

Materials

Hexafluorobutyl methacrylate (HFMA) ($\geq 96\%$) was purchased from Harbin XEOGIA Fluorine-silicon Chemical Co., Ltd. (China). 4-vinyl pyridine (4-VP) ($\geq 96\%$) was commercial

supplied by Alfa Aesar Company (USA). Butyl methacrylate (BMA) and azobisisbutyronitrile (AIBN) of analytical grade were procured from Chemical Reagent Co., Ltd. (China). HFMA, 4-VP and BMA were washed with 1 mol L⁻¹ NaOH and then distilled under reduced pressure before use. Azobisisbutyronitrile (AIBN) was recrystallized in boiling alcohol then dried in a vacuum oven at room temperature. The other reagents were used as received without further purification.

Synthesis of copolymer with pendant pyridinium groups

The copolymer, poly(HFMA-co-4VP-co-BMA) (PHVB) was synthesized via free radical copolymerization of 4-vinyl pyridine with hexafluorobutyl methacrylate and butyl methacrylate using AIBN as an initiator and N,N-dimethylformamide (DMF) as a solvent, and the reaction process is shown in Fig. 1. The reactant mole ratio of HFMA, 4-VP and BMA was 6:3:1. The copolymerization was carried out in a three-necked round-bottomed flask with magnetic stirrer at 65 °C for 24 h under a nitrogen atmosphere. After precipitating and washing with deionized water, the copolymer was dried in a vacuum oven at 80 °C.

Membrane preparation

The membranes were prepared by a phase-transfer method. The synthesized copolymers were dissolved in DMF to form 5 wt% solutions. The copolymer solutions were cast onto glass plates and dried in a vacuum oven at 80 °C for 12 h. Then, the membranes were peeled off from the glass plates. The fabrication of the OH⁻ form membranes were performed according to Fig. 2. To obtain membrane 1#, the membranes were soaked in 1 mol L⁻¹ HCl for 48 h, then soaked in 1 mol L⁻¹ NaOH overnight at room temperature. And to gain membrane 2#, the membranes were soaked in butyl bromide solution (1 mol L⁻¹, dissolve in cyclohexane) for 24 h at 60 °C, subsequently soaked in 1 mol L⁻¹ NaOH overnight at room temperature. Finally, the obtained OH⁻ form membranes were washed with deionized water thoroughly and preserved in deionized water for further use.

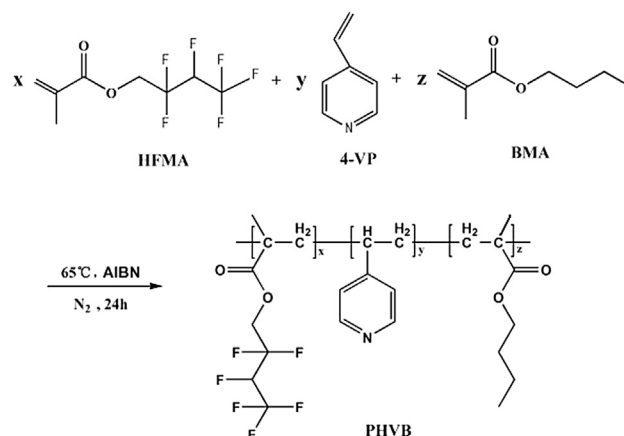


Fig. 1 – Copolymerization of HFMA, 4-VP and BMA.

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