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Homogeneous polymer/filler composite membrane by spraying method for enhanced direct methanol fuel cell performance

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ABSTRACT

In this work, composite membranes for a direct methanol fuel cell (DMFC) were prepared using a spraying method to improve cell performance especially at a high methanol concentration. Nafion polymer and mordenite as a filler were used for the composite membrane preparation using a spraying method and a conventional solution casting method and the membranes from the two methods were compared. SEM images showed that a more homogeneous composite membrane could be obtained using the spraying method. The effect of mordenite content was also studied. The membranes were consequently characterized and tested in DMFC operation. The results were compared to those prepared using the solution casting method at 30, 50, and 70 °C with methanol concentrations of 2, 4, and 8 M. It was found that the membrane with 5 wt.% mordenite from the spraying method showed a vast improvement in DMFC performance. When the cell was operated at 70 °C, the maximum power density of 5 wt.% mordenite from the spraying method was higher than that of commercial membrane and 5 wt.% from the solution casting method. Power densities from the 5 wt.% sprayed membrane were higher by around 29%, 40%, and 60% at 2, 4, and 8 M methanol concentration, respectively.

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Introduction

A fuel cell is an electrochemical device that offers the prospect of a greener future to advance global energy resources with environmental and economic benefits [1]. Fuel cells have been extensively acknowledged as a clean and an efficient technology for generating electricity [2,3] as they have several advantages in terms of high-energy efficiency, high-energy density, low noise, near-zero pollutants, and short charging time [3–5]. Direct methanol fuel cells (DMFCs) have attracted much interest due to their advantages of inexpensive raw materials, high energy conversion efficiency, and low operating temperature [6–9]. The transportation of methanol fuel is more convenient than that of hydrogen fuel because methanol is in liquid form at room temperature and atmospheric pressure making it having high energy density [3,7]. The benefit of using liquid energy carrier is enable long operating times without refilling and cheaper cost compared to pressurized hydrogen gas storage [3,7]. Furthermore, methanol has a simple molecular structure and its electrochemical activity is relatively high. Thus, DMFCs are promising for proton exchange membrane fuel cell (PEMFC) applications [10]. Polymer electrolyte membrane or proton exchange membranes (PEMs) is a key components used in PEMFC [11] as it plays a vital role in transporting protons and blocking electrons from anode to cathode as well as preventing fuel going through during the fuel cell operation [10,12]. The limitation of using PEMs in DMFC is that methanol can permeate through the polymer electrolyte membranes from the anode to the cathode and this methanol crossover is possibly the most severe problem in DMFCs [12–15].

The polymer electrolyte membrane has two important functions as a proton conductor and a fuel and gas barrier [12]. Therefore, a good conducting membrane should have key properties of high proton conductivity, low methanol permeability, good chemical resistance, good thermal and mechanical stability [5]. Many types of sulfonated polymers have been used as polymer electrolyte membranes such as Nafion, chitosan, sulfonated poly(arylene ether sulfone), and sulfonate poly(ether ketone) [16–20]. Ahmed et al. [10] and Zakil et al. [21] presented overview of membranes for direct methanol fuel cell application. Nafion has been the most widely used as a proton-conducting membrane because of its excellent chemical, mechanical, thermal stability, and high proton conductivity, (0.1 S cm^{-1}) [22]. However, Nafion exhibits high alcohol permeability leading to reduced DMFC performance [23]. Many approaches have been studied to improve the alcohol resistance in a Nafion membrane, such as developing an alternative polymer electrolyte [24,25] and Nafion membrane modification [26–31].

According to many researchers, membrane modification by incorporating fillers into the Nafion matrix could enhance DMFC performance [15,32–35]. Zeolites have a highly crystalline structure and are resistant to an organic solvent due to their hydrophilicity. Mordenite (MOR) is one of the zeolites which can withstand an acidic medium and high temperature (above 800 °C) [26]. However, the weak interaction between an organic component and the zeolite in the composite membrane causes poor interfacial properties [22,36,37]. Many researchers have reported that the incompatibility can attributed to pinholes between the filler and the Nafion matrix [26,36,37]. These pinholes facilitate alcohol transport or fuel crossover [32]. The interfacial properties can be improved by grafting a silane coupling agent onto the surface of MOR [38].

There are several methods to fabricate membranes for DMFC such as the sol-gel method, dip coating, and the solution casting method (SCM) [39–41]. The sol-gel method has not been widely used because of its chemical complexity [42]. Thus, SCM with the addition of zeolites has become of interest because of its simplicity. However, the SCM has limitations because it takes time to evaporate the solvents. Therefore, the additive cannot be suspended and this results in deposition at the bottom of the composite membrane [43,44]. Thus, to overcome this problem, a new method was used to fabricate the composite membrane, which was intended to improve the dispersion of MOR in the polymer matrix.

Xing et al. [45] successfully used a solution-spray process to fabricate Nafion/polytetrafluoroethylene composite membrane. Our previous work reported a preliminary study on applying a spraying method to fabricate Nafion/filler composite membrane [46]. It was found that sprayed membranes can result in a more homogeneous composite membrane compared to that produced using the solution casting method. In this work, the spraying method (SM) was the focus and was used to investigate the composite membrane morphological properties with different filler loadings, as it was expected that a higher filler loading could lead to lower alcohol permeability while the membranes maintained high proton conductivity. Consequently, the performance of DMFCs using sprayed composite membranes may be improved. Therefore, this study investigated a series of membrane characterizations and tests of the membranes in DMFC operation over a range of temperatures from low to high. Furthermore, the DMFC performance test was carried out at a high methanol concentration.

Materials and methods

The 20 wt.% Nafion solution was purchased from Ion Power. Mordenite-Na with a Si/Al molar ratio of 5 and an average particle size of 2.7 μm was purchased from Zeolyst International. 3-Mercaptopropyl triethoxysilane (MPTES), sulfuric acid, hydrogen peroxide, ethanol, methanol, N,N-dimethylformamide (DMF), dichloromethane, ammonium chloride, and toluene, were purchased from Sigma-Aldrich. De-ionized water was used throughout the study.

Mordenite preparation

Mordenite preparation was carried out following the procedure described in Prapainainar et al. [38,44,46]. The MOR-Na powder was ground using a zirconia ball mill (0.5 mm diameter) to reduce the particle size. After grinding, an average particle size of 0.317 μm was obtained. The ground MOR-Na particles were protonated in 1 M NH_4Cl at room temperature for 24 h, then washed with deionized water, and dried at 105 °C for 24 h. Then, it was calcined at 550 °C for 5 h with a heating rate of 2 °C·min^{-1} to produce MOR-H. The MOR-H was

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