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Investigation on electrical and electrochemical behaviour of cellulose acetate: Ammonium iodide: Silica composite biopolymer electrolyte for proton exchange membrane (PEM) fuel cell performance

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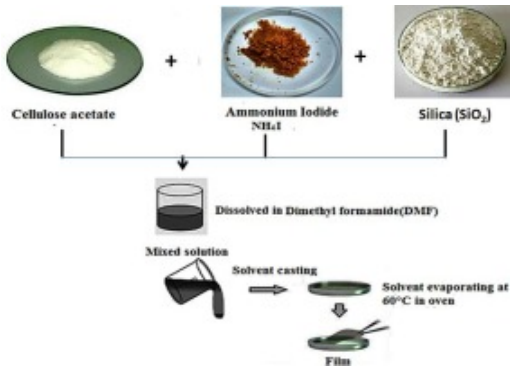
Abstract

Cellulose acetate (CA): ammonium iodide (NH_4I) solid biopolymer electrolyte and cellulose acetate (CA): ammonium iodide (NH_4I): silica (SiO_2) particle composite-based biopolymer electrolytes were developed via solution casting route for single proton-exchange membrane fuel cells (PEMFC) applications. X-ray diffraction (XRD) analysis was used to identify the diffraction peaks and their corresponding angles, which led to the confirmation of the aforementioned biopolymer. The admixture of SiO_2 into the 50CA:50NH_4I improves the amorphous nature than 50CA:50NH_4I in the XRD pattern. Using electrochemical impedance spectroscopy (EIS) analysis, the CA:NH_4I:SiO_2 has found higher ionic conductivity, lower electrical resistance, and a lesser CPE value than the CA and CA:NH_4I. The highest-conducting CA:NH_4I:SiO_2 and CA:NH_4I developed were used in the construction of a single PEMFC, where the obtained power density values were 122.4 and 106.4 $mWcm^{-2}$. The introduction of SiO_2 admixtures with CA:NH_4I has found relatively higher power density value than CA:NH_4I.

Graphical abstract



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Introduction

Biodegradable polymer-based electrolytes have received more attention in the field of material science. It has the advantages of light weight, a low-temperature synthesis process, low cost, non-corrosive, and non-toxic nature [1], [2], [3]. In a growing economy, the demand for composite biomaterials efficient for solid-state batteries, PEM fuel cells (PEMFC's), supercapacitors, and sensors has increased. One such promising device is PEMFCs, which can directly convert chemical energy into electrical energy with almost no emissions [4]. PEMFCs are sustainable energy systems that can be used for both portable and stationary power generation because of their high energy density and dynamic response, lower operating temperature, and ease of operation [4].

Previous research has been conducted on a composite-based biopolymer membrane containing chitosan and cellulose for PEMFC's applications [5], [6]. Among all biopolymers, cellulose acetate (CA) stands out for its low cost, non-toxicity, biodegradability, and biocompatibility. In our previous work, CA admixtures on NH_4SCN and NH_4NO_3 were found to have higher ionic conductivity than CA [7]. To improve the ionic conductivity and electrochemical stability of CA admixtures with NH_4I, a good proton source has been introduced in this work. However, ionic dopant fails to improve the performance of samples to be used in device applications.

In order to further increase the ionic conductivity of the biopolymer electrolyte, modifications such as adding filler particles have been helpful to improve the device's performance [3]. Silica admixtures in biopolymer systems support polymer segmental motion [8]. Because of its unique properties like size and surface nature. Dobos et al. [9] reported that cellulose acetate/silica were synthesized using CA with tetraethyl orthosilicate via solution casting method to investigate their mechanical strength, antibacterial properties and microstructural characteristics. In our work, we have synthesized CA:SiO_2 via solution casting route by taking CA with silica powder directly purchased from Aldrich.

There has been no report on the synthesis of a CA:NH_4I:SiO_2 based-polymer system via the solution casting route to be introduced in PEMFC's applications. The preliminary performance analysis of a single PEMFC's built with the highest conducting sample CA:NH_4I:SiO_2 and CA:NH_4I results has been

discussed in detail. The crystalline state, vibrational bands, and dielectric properties of the polymer electrolyte are also discussed in detail.

Section snippets

Materials and methods

For the synthesis of 50CA:50NH_4I and 50CA:50NH_4I:18 mg SiO_2, CA powder (39. wt% acetyl content, DS2.5, and average MW ~ 30,000, Colloids Impex), NH_4I (MW-144.94, purity 99%-NICE), SiO_2 (MW:60.08, purity 99.9%-Sigma Aldrich, size: 5–15 nm), and dimethyl formamide (DMF,MW-73.09, purity 99.8%-Merck) as a solvent were used. To obtain a homogeneous solution, a 1:1 ratio (in M.wt%) of CA and NH_4I salt was added in 50 mL of DMF solvent, which has been stirred for 24 hrs. 18 mg of SiO_2 was added to…

Structural analysis

Fig. 1(a-g) represents XRD patterns of pure CA at various concentrations of CA:NH_4I salt and SiO_2-doped CA:NH_4I membrane. The XRD patterns of pure CA and their diffraction peak were identified at 2θ=17°, 18°, 23°, 26°, and 28°, which has a semicrystalline nature also included in Fig. 1(a) [7]. Fig. 1(b-d) displays the XRD patterns of 60CA:40NH_4I, 50CA:50NH_4I, and 40CA:60NH_4I-doped polymer electrolytes. The addition of 40 and 50 wt% NH_4I to pure CA reduces the semicrystalline nature and…

Conclusion

In this work, 50CA:50NH_4I and 50CA:50NH_4I:18 mg SiO_2-based polymer electrolytes were synthesized, followed by structural state and electrochemical performance studied. The XRD results indicate that the amorphous nature of the diffraction pattern increases after the addition of SiO_2 admixtures to CA:NH_4I. The EIS study's goal is to comprehend the movement of ionic conductivity and the relaxation behaviour of the synthesized electrolytes. 50CA:50NH_4I:18 mg SiO_2 has higher ionic conductivity…

CRediT authorship contribution statement

S. Monisha: Methodology, Investigation, Writing – original draft, Conceptualization, Writing – review & editing. **J. Gajendiran:** Investigation, Writing – original draft, Conceptualization, Writing – review & editing. **G. Boopathi:** Writing – review & editing. **S. Selvalakshmi:** Writing – review & editing. **S. Gnanam:** Investigation, Writing – review & editing, Conceptualization. **S. Gokul Raj:** Writing – review & editing. **G. Ramesh Kumar:** Writing – review & editing. **V. Karuppasamy Vikraman:** Writing –…

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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