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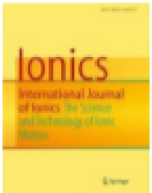

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ORIGINAL PAPER

Development of biopolymer electrolyte membrane using Gellan gum biopolymer incorporated with NH_4SCN for electro-chemical application

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Abstract

Based on the biopolymer Gellan gum with ammonium thiocyanate (NH_4SCN) salt, solid electrolyte has been prepared with distilled water as solvent, using solution casting technique. The prepared solid electrolytes are subjected to various characterization techniques such as XRD, FTIR, DSC, and Ac impedance technique. Amorphous/crystalline nature of biopolymer membrane is studied by XRD. The polymer–salt complex formation has been studied by FTIR technique. Biopolymer membrane of 1 g Gellan gum with 1.1 M wt% of NH_4SCN exhibits very high amorphous nature with a high proton conductivity of 1.41×10^{-2} S/cm and a glass transition temperature (T_g) of 42.98 °C. Using the highest ionic conducting biopolymer electrolyte, proton battery and fuel cell have been fabricated and their performance is studied. Proton battery constructed shows the open circuit voltage of *1.62 V*. A single fuel cell constructed using the highest conducting membrane gives the voltage of *580 mV*.

Keywords Biopolymer · Gellan gum · Ac impedance study · Proton battery · Fuel cell

Introduction

The human life is unpleasant without the use of electronic devices. The life span of the electronic device depends on its power sources such as battery and components of battery. The performance of the battery depends on its electrolyte. This electrolyte must fulfill the following requirements such as safety, consistent performance/accuracy, and stability there by making the process easier for the ion transportation [1, 2]. The electrolytes are of various types like liquid electrolyte, solid electrolyte, and polymer electrolyte. In the realm of polymer electrolyte, the proton conducting polymer electrolyte has received considerable attention in the view of their perspective applications in electrochemical devices such as solid state batteries, fuel cells, dye sensitized solar cell, super capacitors, humidity sensor, gas sensors, and electrochemical windows [3–13].

The main preference towards the solid polymer electrolyte in solid state ionic devices raises due to their film forming capacity of different size, flexibility, mechanical strength, processability, ability of ion transportation, electrochemical stability, proper electrode–electrolyte contact, safety, long life, no leakage, and light weight [14–16]. Blending of two polymers, adding of inorganic filters, use of grafts, block co-polymer, and adding plasticizers to the polymers are the methods that can improve the properties of electrolytes [14–18].

Various types of solid polymer electrolytes such as PVC [19], PAN [20–24], PVP [25], PEO [26], PVA [27–31], PGS [32], and PMMA [33] have been explored in the development of solid electrolyte system. But these polymers are synthetic-based polymers. They posses very high cost and are non-biodegradable. The renewable form of energy is essential in recent days to overcome the challenges like power crisis and environmental pollution. Keeping the caution of environment pollution and the cost efficiency in mind, the researchers recently work on the development of natural polymers as electrolyte, which will be a promising substitute to the synthetic polymers. The preference towards the natural polymers is due

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to their abundant, environment friendly and biodegradable nature, and very good mechanical and electrical properties. Among the natural polymers, polysaccharides have good potential as hosts for ionic conduction since they are cheap and eco-friendly. The polysaccharides such as cellulose acetate, starch, pectin, carbonyl methyl cellulose, carrageenan, tamarind seed, chitosan, agar-agar, and gums are used as polymer electrolytes in the electrochemical devices [34–46]. S. Selvalakshmi et al. has reported that the composition of 50 agar: 50 NH_4SCN gives the conductivity of about 1.03×10^{-3} S/cm [36]. S. Monisha et al. reported the conductivity of 1.02×10^{-3} S/cm for the sample 50CA:50NH_4NO_3 [38]. M. Premalatha et al. has reported the conductivity of 1.58×10^{-3} S/cm for the composition, 1 g TSP:0.4 g NH_4Br [40]. G. Nirmala devi et al. has reported for the composition, 60Dextrin:40NH_4SCN providing the conductivity of 4.05×10^{-4} S/cm [41]. M. Muthukrishanan et al. reported for the bio polymer composition 40 Pectin: 60 NH_4SCN having the conductivity, 1.5×10^{-3} S/cm [42]. S. Karthikeyan et al. has reported that 80% I-carrageenan: 20 wt% NH_4Br gives the highest conductivity 1.08×10^{-3} S/cm [44], etc.

In the present work, the Gellan gum polymer is taken as the host polymer. Gellan gum is a water-soluble anionic polysaccharide, produced by the bacterium, *Sphingomonous Elodea* [47, 48]. It is a multifunctional gelling agent. It can be used alone or can be used with other products to obtain a wide interesting character, including the thermo-reversible membranes (that shows the reversible properties under heating and cooling). From the view of chemical structure, the Gellan gum is a heteropolysaccharide composed of complex tetrasaccharide repeating unit consisting of α-L-rhamnose (Rha), β-D-glucose (Glc), and β-D-glucoronic acid (GlcA) in the ratio of 1:2:1. Figure 1 shows the structure of Gellan gum. It has been chosen because it contains good amount of –OH group, to which cation of any salt can be attached to increase the charge carrier. These are available as low acyl that forms hard, brittle gels and high acyl that forms soft, elastic, not brittle gels. This means that they can produce gel texture ranging from hard and brittle to fluid.

For the development of new contact lenses in the field of ophthalmology, the Gellan gum is very helpful. As the Gellan gum is edible, they are used in the food industry as a stabilizer and food thickener [49–51].

The unique property of Gellan gum is its ability of producing solutions with low viscosity [49, 51]. Another important property is that it has high thermal stability up to 120 °C and highly transparent, because of which they can be applied in electrochemical devices too. Due to abundance availability, low cost, and easier process ability, the Gellan gum can also be a best replacer in the biopolymer line for the non bio-degradable, toxic, and harmful materials used in the commercial electrochemical devices. Literature survey reveals that there are very few reports in Gellan gum. S.R. Majid et al. has studied lithium ion conductivity in Gellan gum blended with PVP using $LiClO_4$ [52]. I.S.M. Noor reported study of Gellan gum with $LiCF_3SO_4$ [53, 54].

Ammonium salts are reported as they are very good proton donors to the polymer matrix. The protonic transport in the polymer membrane generally involves the motion of H+ ion. Preparation of biopolymers with the ammonium salts has attracted many researchers for the development of high conducting biopolymer electrolyte. [29–32, 34–46, 55–57].

The objective of the present work is to synthesize the electrolyte with a higher conductivity value and to apply the polymer film for the electrochemical devices. The polymer electrolyte based on Gellan gum doped with NH_4SCN was prepared, and the characterization was done using XRD for its amorphous nature, FTIR for its chemical bonding, DSC for the determination of glass transition temperature, and Ac impedance spectroscopy for its conductivity measurement. Proton battery and fuel cell have been fabricated, and their performance are studied.

Experimental method

Materials and preparation

For the synthesis of the biopolymer electrolyte, the simplest technique known as Solution Casting Technique has been employed. In this work, the polymer, Gellan gum (Sisco Research Laboratories Pvt. Ltd, India), is used as the raw material. Double distilled pure water is used as the solvent to dissolve the polymer. As the gelling temperature of the Gellan gum is 35–40 °C (2% soln), to maintain the aqueous form and to dissolve, the distilled water is heated above 90 °C and 1 g Gellan gum was added pinch by pinch and stirred thoroughly

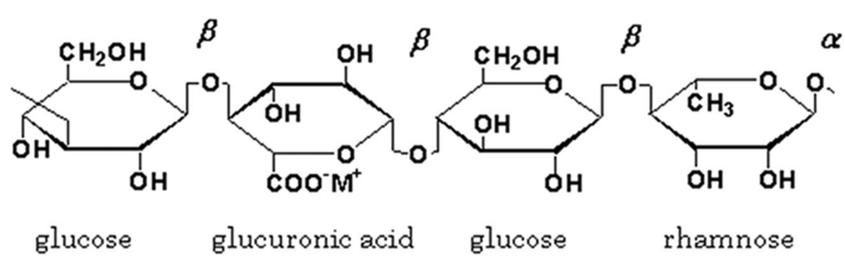

Fig. 1 Structure of Gellan gum