World Journal of Advanced Research and Reviews
eISSN: 2581-9615 CODEN (USA): WJARAI
Cross Ref DOI: 10.30574/wjarr
Journal homepage: https://wjarr.com/

(REVIEW ARTICLE)

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Towards green energy: Choice of *Tamarind Gum* as an ecofriendly biopolymer electrolyte material

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World Journal of Advanced Research and Reviews, 2024, 24(02), 961–974

Publication history: Received on 29 September 2024; revised on 05 November 2024; accepted on 08 November 2024

Article DOI: https://doi.org/10.30574/wjarr.2024.24.2.3419

Abstract

The need for the preparation of eco-friendly and sustainable materials to be used as alternative sources for energy devices gained momentum in the mid-20th century. Several strategies and techniques have been deployed in the recent times to prepare and characterize biopolymer matrices for proton and ion conduction. Research studies conducted on properties of electrolytes were crucial in developing energy storage systems, such as batteries and supercapacitors. Research is progressing in order to develop new kinds and varieties of battery materials that are specifically designed to meet the demand for energy storage devices that are sustainable. Amongst the various types of electrolytes explored, polymer electrolytes are gaining significant attention to be used as better replacements for liquid electrolytes in Li-ion batteries, offering safety, flexibility and higher energy densities. Novel polymer electrolyte systems are being developed in order to achieve enhanced ionic conductivities, retaining few desirable properties. Synthetic polymer electrolytes (SPEs) mainly cause harm to the environmental pollution due to their toxic effect and non-biodegradable nature. Thus, research studies explored the possibility of usage of natural polymers as better substitute to synthetic polymers, while retaining the efficiency. Research studies on one such biopolymer, Tamarind Seed Polysaccharide are discussed in this paper, thereby proposing an eco-friendly and cost-effective biopolymer electrolyte, that can be used for the energy storage device application.

Keywords: TSP; Biopolymer electrolytes; Synthetic polymer electrolytes; Energy storage device

1. Introduction

Over the past few decades, there was a tremendous requirement for the production of energy. According to the scientific definition, energy, something which is defined as the ability to do work. Several ways and means of production of energy had evolved and progressed in the past century. Out of the various forms of energy existing on the planet, the energy received through the radiation from the cosmic rays is stored as carbon reserves, which otherwise are known as non-renewable sources of energy [1]. The exploitation of these nonrenewable resources had created avoid, and thus began a greater demand for the production of energy. Apart from production of energy, scientist have also felt the need for storage of energy since the time of industrial revolution [2]. There was a high need felt for development to be made on the energy storage systems. The global demand, driven up in both the developed and the developing nations, created a scarcity for the available sources of energy [3]. As a result, most of the non-renewable sources of energy, which are also otherwise known as the fossil fuels are being depleted, and thus, scientists have made significant progress in finding alternative materials to combat the energy crisis. The transition of moving from the conventional nonrenewable sources of energy to the renewable sources for energy storage needed the invention of technology that can store and provide energy as per the demand [4]. Of the several devices which have been used to store energy, batteries were preferred to

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be the most chosen energy storage systems due to several factors such as compact size, lighter, weight and minimal production costs [5–7].

2. Methodology

2.1. History of batteries

The oldest known battery of the world was called the 'Baghdad battery'. This was discovered in the town of Baghdad 2000 years ago [8]. The output of this battery was 1.5 to 2V. Later, an Italian scientist by name, *Alessandro Volta* discover and developed copper electrodes and zinc electrodes, which could be used in a battery [9]. He had immersed these electrodes in the solution, which was acidic or sometimes saline in nature. In his observations, he reported that while copper atoms had barely broken down, whereas zinc atoms had broken down and electrons had passed through the electrolyte. Based on these results, he proposed that copper atoms act as positive terminal, whereas zinc atom behave as the negative terminal and stated that electricity flowed from the copper to the zinc when these two electrodes were connected by a conducting material [10]. This was the first known battery called by name 'Volta battery' which became the foundation for all types of chemical batteries. Similar invention was made by the scientist *George Leclanche* in the year 1868 which lead to the origin of Leclanche cell, and this was considered to be the origin of the modern dry batteries [11]. In these batteries, zinc acts as the anode material and manganese dioxide acts as the cathode material. For the functioning of the battery, both were covered in a porous material and were immersed in the solution of ammonium chloride. As the circuit was connected, this battery generated voltage of magnitude 1.4 V. The stated disadvantage of this battery was that the ammonium chloride solution spilled over and there was inconvenient to use [12]. During the later stages of development, the acidic electrolyte was replaced with alkaline materials such as sodium hydroxide or potassium hydroxide and the batteries developed along these lines hand minimal internal resistance, longer life and higher open circuit voltage nearly 1.9 V [13]. With the passage of time, there was enhancement in the conductivity of the batteries. Eventually, several other batteries such as mercury, battery or button cells were introduced [14]. In the year 1899 by a Swedish scientist *Waldemar Jungner* devised the nickel-cadmium battery [15]. This was stated to be the first rechargeable battery, which enabled storage of the produced electricity. Further research studies have replaced nickel battery by lead acid battery post 1870's. The lead acid battery was fabricated by the scientist *Plante* and was further improved in terms of its efficiency by *Faure* [16]. With the passage of time, the traditional lead acid battery and nickel-cadmium batteries were replaced by lithium-ion batteries, due to the greater performance of latter. This invention took place due to the fact that the continuous demand of portable devices needed batteries with higher energy density, which led the researchers to find alternative power systems.

2.2. More on Li-ion batteries

The most important merits of the lithium-ion batteries are higher capacity, greater life cycle, greater energy density, lightweight and smaller in size [17]. Thus, the development of lithium-ion batteries improved the overall capacity and energy, which further improved the electrolyte and electrode materials with better properties and lower costs. Of the several advances that have been made in the production and storage systems, lithium battery served as promising materials for huge source of power output that had both industrial and domestic applications [18–22]. Compared to the several other batteries such as nickel cadmium battery, lead acid battery and nickel metal hydride battery, lithium-ion batteries, reported values of greater specific energy density. The primitive lithium battery consisted of anode and cathode, connected through an electrolyte rich in lithium ions [23]. The first rechargeable lithium-ion battery with the cathode material TiS_2 and a metal anode was prepared by Whittingham in the year 1976 [24]. Properties made on the discharge studies of lithium batteries showed that they had low self-discharge rate as compare to the nickel batteries. Apart from several advantages exhibited by lithium-ion batteries, there were also several demerits that had been reported which led to an eventual shift to alternative batteries compared to lithium batteries [25]. An incident of a Boeing airline catching fire due to the explosion of lithium battery, created a necessity to enhance the battery safety [26].

Few of the disadvantages exhibited by lithium batteries are

- The formation of dendrites in the lithium batteries, which led to short circuit upon extensive cycling.
- Hazardous nature rechargeable and susceptible nature, which lead to explosion of the battery
- Degradation of the performance of the battery at higher temperatures.
- Highly toxic nature
- lesser abundance and expensive
- Non-biodegradability.

Thus, the major drawback of liquid electrolytes is leakage problem [27]. In another class of electrolyte materials called as the gel electrolytes, there is a semi solid mass that shows more stability and equivalent conduction is similar to the conduction mechanism of liquid electrolytes [28]. In both these above stated types of electrolytes, the charge carriers that is ions conduct when they are moving in a polar solvent such as ethylene carbonate, butyrolactone, propylene carbonate etc. Both these stated electrolytes have their ionic conductivity lying in the range 10^{-2} to 10^{-3} S cm^{-1} [29]. For lithium batteries working at a room temperature, the ionic conductivity of these electrolytes is sufficient. As the ambient temperature rises, there are other factors that prevent the usage of liquid electrolytes in terms of safety and efficiency. Another setback in using these electrolytes is formation of insulating layer in battery application [30]. The decomposition products of these electrolytes get deposited on the electrodes, forming insulating layer at the electrolyte electrode interface, which hinder ionic movement, thereby reducing the lifespan.

2.3. Solid Electrolytes

Thus, the usage of liquid electrodes in batteries and other such storage devices has a limit on the operational temperature range. These drawbacks lead to the discovery of solid electrolytes. Solid electrolytes are chemically very strong and are used as self-standing membranes and can be considered as good alternatives to the liquid electrolytes. However, the limiting factor is that their ionic conductivity is low at room temperature. Generally, the conductivity values of the solid electrolyte lies below 10^{-5} S cm^{-1}.

Following our important features of the solid electrolytes.

- The structure of the materials to be used as solid electrolytes needs the presence of numerous empty sides which could be in the form of interestial sites or vacancies, which are easily accessible for the mobility of ions through the material. These empty sites are generally used for the moment of ions through the crystal lattice.
- The structure of the material should have solid framework with several channels that are open, probably in three-dimension. The structure of the material containing the mobile ions should be molten so as to allow their free movement.
- The occupied sites along with empty side should have nearly same potential energy and low activation energy for the hopping of ions among the neighboring sites. Higher values of energy are not preferred since they lead to reduction in the carrier mobility. Also, the presence of potential energy wells with greater depth or the availability of stable sites might lead to carrier localization.

Generally, the values of lower conductivity in solid polymer electrodes is due to the poor coordination of anions in the polymer matrix [31]. The dielectric constant of the system is also low. Thus, the materials which provide higher values of ionic conductivity or electronic conductivity along with greater transport numbers are important for researchers. Generally, cations are responsible for ionic conduction in solid electrolytes and thus small sized cations compare to that of anions are preferred in solid electrolytes. In the work of Kim et al. [32] it was shown that addition of larger sized or heavier anions in the polymer structure leads to reduction in the conductivity, at least by one order of magnitude.

Of the several characteristic features to be possessed by solid electrolyte, three are listed below:

- The charge carriers mainly should be union species only.
- The value of activation energy must be low.
- The value of ionic conductivity at room temperatures must be in the order of range of 10^{-1} to 10^{-4} S cm^{-1}.

2.4. Polymer Electrolytes

The usage of a special class of solid electrolytes, called as polymer electrolytes in the present-day industry increased in recent years [33]. Kucharski et al. have reported in the study that an electrolyte should have conductivity of the orders greater than 10^{-4} [12]. Electrolytes are reported to be existing in either in liquid gel or solid forms. Though liquid electrolytes have been used for several application over past few decades, these are reported to have certain drawbacks such as leakage, flammability, corrosion and reactivity with the electrodes [34]. All such drawbacks can be overcome through the usage of polymer electrolytes in the place of liquid electrolytes, since there is no leakage or corrosion in the former [35]. Polymer electrolytes are light in weight and are also reported to have wider electrochemical stability along with thermal stability. The process of preparation of solid materials such as crystalline and polymer electrolytes, glass, ceramics had started as early as 1970s [36].

The progress of solid-state materials like ceramic, glass, crystalline and polymer electrolytes was started in the early 1970s [37]. Polymer electrolytes are special kind of solid electrolytes, in which the membranes composed of the dissolution of salt in a polymer matrix of greater molecular weight. These are prepared and synthesized through the

dissolution of metallic salts in polar polymer hosts. The efficiency and the performance of these polymer electrolytes is evaluated and assessed based on their ionic conductivity, which mainly depends on two factors, degree of crystallinity and viscosity of the polymer electrolyte [38]. It is stated that polymer electrolytes having lower values of viscosity will have more sites and hence possess more ionic conductivity [39]. Also, crystalline polymer electrolytes have a greater degree of crystallinity and in the crystal phase, the ion possesses lower mobility and therefore they have lower ionic conductivity [40]. Several polymer electrolytes have gained immense attention in the areas of polymer science, inorganic, and organic chemistry and electrochemistry. Thus, large number of polymer electrolyte materials having various kinds of transporting ions such as H^+, K^+, Ag^+, Na^+, Li^+, Mg^{2+} were reported in the past [41].

Several attempts have been made by the researchers in order to enhance the value of conductivity of these polymer electrolytes, such as polymer blending, addition of a dopant (typically a compatible salt with the polymer), use of branched copolymers, incorporation of inorganic nano-fillers, addition of ionic liquids, plasticizers and nano materials etc. [25,42]. One of the main uses of these polymer electrolytes is that they can be prepared in desired shapes and sizes, with minimal cost apart from possessing higher flexibility [43]. With evolution, these have also been used in the development of fuel cells and battery applications.

Based on physical composition, these polymer electrolytes are segregated as

- Gel polymer electrolytes
- Composite polymer electrolytes
- Plasticized polymer electrolytes
- Blend polymer electrolytes

The very first research reported on the solid polymer electrolytes was by wright et al [44]. With the progress of time, the research on the development and preparation of the solid polymer electrolytes was enhanced due to their potential applications in several electrochemical devices. It was also reported that these had application in portable devices, such as mobile phones, laptops, credit, card cards, etc [45]. Generally, solid polymer electrolytes are prepared through the dissolution of inorganic salts in a polar host polymer. Though several attractive properties are possessed by solid polymer electrolytes, few of them are:

- Light weight
- Flexible
- Higher energy density.
- Easier to fabricate
- Solvent – free
- Highly thermal
- Low volatility
- Electrochemical stability
- Electrical and mechanical stability.

Therefore, the main advantage of solid polymer electrolytes in comparison to liquid electrolytes is prevention of the leakage of electrolyte along with a longer life span. Few drawbacks of the solid polymer electrolytes are lower ionic conductivity reported at ambient temperatures and also higher values of interfacial resistance [44]. In order to combat this problem, the preparation of ideal polymer electrolytes must have a good choice of host polymer along with a compatible dopant salt. It is also important to note that to ensure a good complexation of salt in the host polymer matrix, the polymer chosen should have lower values of glass transition temperature [46], as, the flexibility of the polymer ensures good complexation with the dopant salt. The glass transition temperature, denoted by T_g is taken as the turning point in studying the nature of amorphous polymers, and is defined as the temperature at which the amorphous polymer undergoes a transition from a glass state to a soft polymeric state. This is typically related to the strength of the material.

Lower values of T_g for polymer electrolytes can be achieved either through choice of polymers with T_g or through addition of a plasticizer (like like PC, EC etc.) to the host polymers. The condition that the total lattice energy of ionic salts introduced as dopants to the host polymer must be however less. This happens for salts having anions with larger radii like I^-, ClO_4^-, $CF_3SO_3^-$, Br^-, SCN^- etc.

2.5. Mechanism of ion conduction through polymer segments

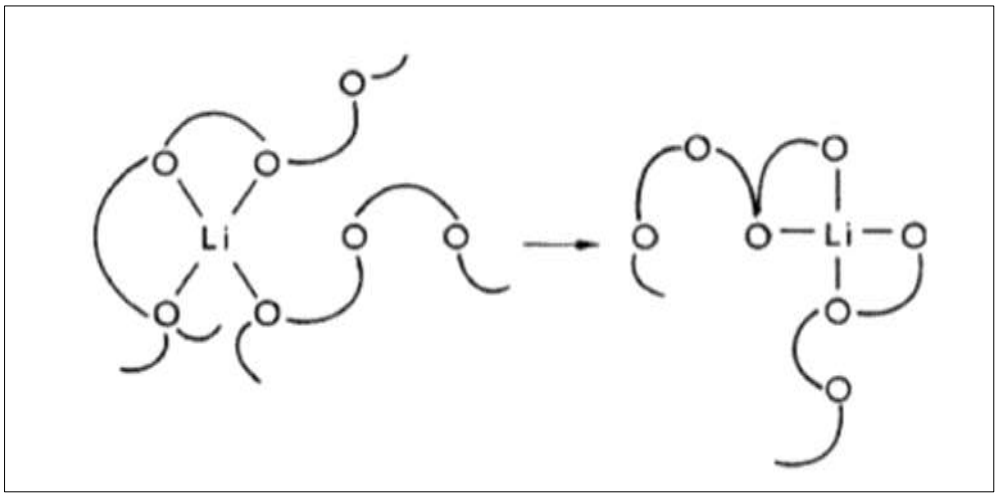

Figure 1 Hopping mechanism for ion transportation

(Source: Obtained from work of Shujahadeen B. Aziz, 2018)

Prior to moving to a special class of polymer electrolytes called as the biopolymer electrolytes, we discuss the mechanism of ionic conduction in the biopolymer segments. The conduction of ions in polymer electrolytes happens through the movement of ions through the polymer segments. The polymer salt complexed with amorphous nature are prepared based on the material composition. The conductivity exhibited by the amorphous complexes of polymer and salt is enhanced as compared to crystalline complexes of polymer and salt [47]. The enhancement of the ionic motion in the polymer segments is also enhanced by chain mobility mechanism [48]. The transportation of the mobile carriers through the polymer matrix happens in the available space and through the intermolecular interactions i.e hydrogen bonding, dispersion, ion dipoles etc. Compare to their counter parts, the solid polymers were preferred to be suitable choice as electrolyte materials due to their distinct features such as leakage free materials, flexibility, light weight, safety, longer life cycles and greater electro chemical stability.

Thus, it is stated that the movement of the cations of the ionic dopant salt between the coordination sites happens through inter–chain or intra–chain hopping mechanism [49].

When a graph of 1000/T vs. log σ is plotted, there is a non-linear nature observed in the curve instead of the linear nature, as proposed by Arrhenius [50]. VTF model is proposed to explain the non-linear nature of the graph, which shows that apart from hopping process, ion transportation mechanism also happens due to some other mechanism in the polymer electrolytes. Therefore, in order to understand the mechanism of ion transportation in polymer electrolytes and to study the changes occurring at the electrode/electrolyte interface, several theoretical models were reported.

Of the several models to explain the ion conduction mechanism, two of them have been reported below.

2.6. Vogel – Tamman – Fulcher (VTF) Model of ionic conduction

As per the free volume theory, the ionic movement in the polymer electrolytes is stated only due to the segmental motion, which enhances the transportation of ions through making and breaking of the sphere of coordination of the solvated ion [51]. In this process, a free volume space into which the ions diffuse into through the application of an external electric filed is created [52]. The Vogel – Tamman – Fulcher (VTF) relation is given by [53]:

$$\sigma = A\, e^{-\frac{E_a}{R(T-To)}} \quad \ldots\ldots\ldots\ldots\ldots\ldots(1)$$

where A is a pre-factor and assumes a constant value, E_a is the activation energy σ is the conductivity at a given temperature. T_0 here is stated as the Vogel temperature, which assumed the value equal to the glass transition temperatures for glasses but and in general, it is taken as 50 °C below the glass transition temperature for the polymer electrolytes [54]. The outcome of this model is that it states whenever the temperatures attain values below the ideal glass transition temperature (T_o), the free volume are assumed to disappear.

2.7. Arrhenius Theory of ionic conduction

From the research studies, it was stated that the ionic conductivity of the polymer electrolytes rises with an increase of temperature and is expressed by the Arrhenius relation [50]:

$$\sigma(T) = \sigma_0 e^{-\frac{E_a}{k_B T}} \quad \text{.....(2)}$$

where σ_0, E_a and k_B denote pre-exponential factor, energy of activation and Boltzmann constant respectively.

In order to exhibit the Arrhenius behaviour, the nature of the graph of 1000/T vs. log σ must be linear [55]. This model states that the ion motion through the host polymer segment is facilitated by defects and the ions move through the process of hopping. As shown in figure 2.2, the hopping of ions happens in two ways, i.e inter-chain and intra-chain hopping. It is a general observation that the anions and the cations of polymer electrolyte are always in motion, which in turn depends on the ionic radius of the counter ion (anion/cation) [56].

2.8. Biopolymer Electrolytes

Biodegradable polymer, also known as bio polymers are special kind of solid polymer electrolytes which are produced through partial or complete dissociation of the bio molecules or biomass through the enzymatic process [57]. Few of the merits of these of these biodegradable polymers, as compare to synthetic polymer are:

- Environmentally friendly
- Easily accessible
- Bio compatible nature
- Source of renewable energy
- Economically cheaper.

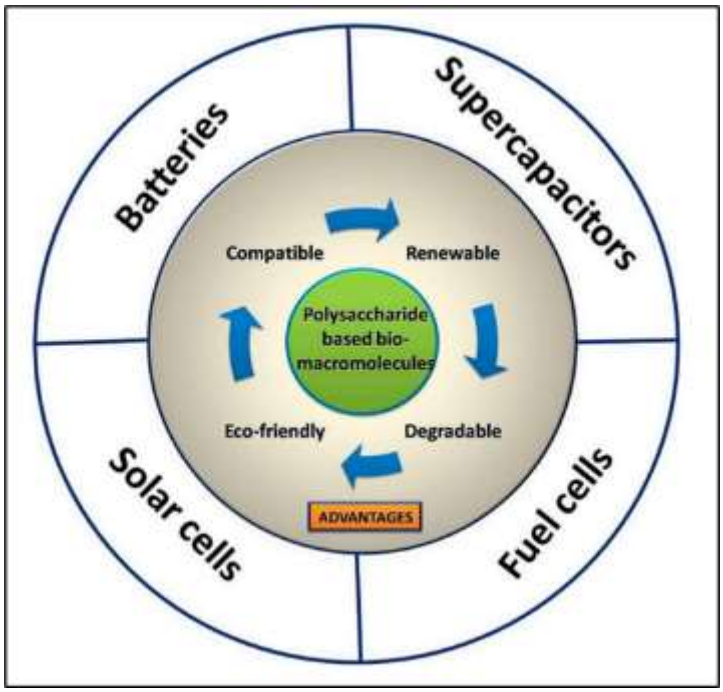

Figure 2 Application of polysaccharide-based bio molecules

(Source: Obtained from work of Pandurangan, Perumal. 2023)

Biopolymers were discovered in 1980 and variety of biopolymers were synthesized for obtaining stable biopolymeric systems that exhibit good electrical and mechanical properties [58]. Biopolymers are synthesized either through the usage of living organisms or using biological materials. The monomer units present in these materials is attached together through the covalent bonds for formation of biopolymers. Typically, biopolymers are readily degradable and are used in various industries like food industries, purification of water, biomedical applications, biomedical

engineering, packaging, agricultural, electronics etc [59]. Due to their abundance, biocompatibility and quality like nontoxicity, these are preferred over synthetic polymers. Few examples of biopolymers are cellulose, protein, lipids, starch, collagen, DNA, RNA, carbohydrates, alginate etc. For the purpose of energy storage devices, researchers have explored several biopolymers, like cellulose and its derivatives, cornstarch, agar-agar, κ – carrageenan, chitosan, and tamarind gum. Amongst the natural polymers, starch and chitosan were reported to be the most popular biopolymers, which are used as the host polymer for electrical double layer capacitor, as, these are capable of forming mechanically stable films with good solubility and electrolytic nature [60]. In 2016, work was reported that through fabricating a supercapacitor from the biodegradable blended polymer of chitosan and chitin sponge ensured 10,000 cycles capacity [61]. Perumal et al. [62] have presented in their work about the applications of the polysaccharide compounds (special class of biopolymers) for usage in electrical systems

- Biopolymers are broadly classified as Polypeptides, Polynucleotides and Polysaccharides.
  - *Polypeptides* – They are formed by chains of amino acids
  - *Polynucleotides* – Polynucleotides are formed by 13 or more nucleotides monomers covalently bonded in a chain. DNA and RNA are examples of polynucleotides
  - *Polysaccharides* – They are polymeric carbohydrate molecules comprised of long chains of monosaccharides units bound together by glycosidic linkages. Cellulose, starch and carrageenan are examples of polysaccharides.
- Based on origin, these are classified as
  - *Synthetic biopolymers – Which are made of biodegradable and renewable materials.*
  - *Natural biopolymers – Which are synthesized through living organisms.*
  - *Microbial biopolymers – These are produced from the microorganisms.*
- Based on Chemical Nature, these are classified as
  - *Biopolymers based on sugar – Made from sucrose unit from potatoes etc.*
  - *Biopolymers based on Starch – Made from glucose that is present in plant tissues.*
  - *Biopolymers based on Cellulose – Made up of glucose found in natural sources such as cotton.*
- Classification based on Monomeric Units
  - *Proteins – Made from amino acids like fibrin, collagen etc.*
  - *Polynucleotides – Made from nucleic acids such as DNA, RNA etc.*
  - *Polysaccharides – Made from linear or branched carbohydrates such as starch, cellulose etc.*

Several important and unique features of biopolymers like better ion conductivity, greater specific capacitance, and mechanical flexibility have made these as promising materials for enhancing the performance of electrochemical devices. Studies done several researchers [63–71] have demonstrated the successful integration of biopolymers in electrochemical cells, showing improved stability and efficiency. In his review, Perumal Pandurangan [62] had reported that Polysaccharides assisted bio-electrolyte membranes application in rechargeable charge storage and conversion devices. Thus, the current study aims at the application of one such novel biopolymer material called as Tamarind Seed Polysaccharide.

Amongst the several available natural polysaccharides, tamarind poly saccharide (TSP) was reported to have several applications in various industries. Also known by the name, tamarind gum, this bio polymer is available abundantly in the regions of Southeast Asian nations, African region and India [72].

2.9. Origin of TSP

Tamarind (generally known by the name *Imily* in India) is a very popular substance which is used as a substitute for starch in the Indian fabric market. This was commercially produced in 1940s. The most interesting properties of this bio polymer is that it has the ability to form gel like material in the presence of sugar or alcohol, which is useful in food items, such as jelly jams, and other preservatives [73]. TSP is a unique biopolymer, and is separated from the kernel seeds of *Tamarindus Indica Linn*, from the family of *Leguminosae*. TSP has branched carbohydrates and has a molecular weight of 1393 g/mol and mainly contains the monomer of three molecules Galactose, Xylose and glucose in the molar ratio 1:2.25:2.8 which make up to 65% of the seed component [74]. At times, this is also known by the name *galactoxyloglican*.

The chemical structure of TSP is of great importance while studying the electrochemical properties of this biopolymer. TSP has chains of β-(1,4)-D-glucan, substituted by the side chains of the molecule (1,6) linked [β-D-galactopyranosyl-(1,2)-α-Dxylopyranosyl] and α-(1,4)-D-xylopyranose to the glucose molecules [74]. TSP has gained attention for its film forming ability, biodegradability, transparency etc. When dissolved in water at 80 °C, TSP forms homogenous solution. TSP is insoluble in organic solvents such as methanol, ethanol, acetone [75].

3. Results and Discussion

3.1. TSP based biopolymer electrolytes for storage devices: Literature

Since past few decades, researcher proved in their studies that TSP based biopolymer electrolytes were used for lithium, sodium, magnesium and proton batteries. Through the deployment of solid biopolymer electrolytes, electrodes degradation in the solid-state batteries is monitored. The solid-state batteries that used TSP biopolymers are discussed below.

In 2016, Premalatha et al. [76] have reported maximum ionic conductivity value of 2.85×10^{-4} S cm^{-1} for proton conducting biopolymers prepared using TSP as the host polymer and Ammonium Thiocyanate as the dopant salt. Subsequently in the year 2017 [77], they have reported maximum ionic conductivity value of 1.58×10^{-3} S cm^{-1} & 4.83×10^{-4} S cm^{-1} for ionic conducting biopolymers prepared using TSP as the host polymer and ammonium bromide, lithium bromide as the dopant salts respectively.

In 2018, Sampathkumar et al. [78] have reported maximum ionic conductivity value of 6.7×10^{-3} S cm^{-1} for lithium-ion conducting biopolymers prepared using TSP as the host polymer and Lithium Chloride as the dopant salt. In the year 2019, they have also reported maximum ionic conductivity value of 8.77×10^{-4} S cm^{-1} for lithium-ion conducting biopolymers prepared using TSP as the host polymer and lithium perchlorate as the dopant salt [79].

In 2019, Perumal et al. [80] have reported maximum ionic conductivity value of 5.66×10^{-4} S cm^{-1} for magnesium-ion conducting biopolymers prepared using TSP as the host polymer and magnesium perchlorate as the dopant salt.

In the year 2020, Sampathkumar et al. have reported maximum ionic conductivity value of 8.37×10^{-4} S cm^{-1} for lithium-ion conducting biopolymers prepared using TSP as the host polymer and lithium triflate as the dopant salt [81].

In 2022, Maithilee et al. [82] have reported maximum ionic conductivity value of 1.7×10^{-3} S cm^{-1} for sodium-ion conducting biopolymers prepared using TSP as the host polymer and sodium perchlorate as the dopant salt. In the same year, Premalatha et al. [83] have reported maximum ionic conductivity value of 1.23×10^{-3} S cm^{-1} for proton conducting biopolymers prepared using TSP as the host polymer and ammonium formate as the dopant salt

In 2023, Saha et al. [84] have reported maximum ionic conductivity value of 1.94×10^{-4} S cm^{-1} for biopolymers prepared using TSP as the host polymer and sodium acetate as the dopant salt.

3.2. Addition of Plasticizers to improve efficiency of solid polymer electrolytes

In the year 2001, Chandrasekaran et al. [85] have reported the preparation of polymer electrolytes based on PEO and $NaClO_3$ with PEG as the plasticizer material, Na and MnO_2 as the anode and cathode materials respectively. The results proved that addition of plasticizer activated relaxations occurring in the polymer chain segments, that enabled ion hopping within the polymer. Highest conductivity of 9.47×10^{-4} S cm^{-1}was obtained at room temperature. Based on similar results, research studies made through incorporation of ethylene carbonate plasticizer into the biopolymer salt complex proved an enhanced conductivity of the TSP biopolymer membranes. The details are given below.

In 2019, Sampathkumar et al. [79] have reported maximum ionic conductivity value of 1.06×10^{-3} S cm^{-1} for lithium-ion conducting biopolymers prepared using TSP as the host polymer, lithium perchlorate as the dopant salt and ethylene carbonate (EC) as plasticizer.

In the year 2023, Maithilee et al. [86] have reported maximum ionic conductivity value of 1.49×10^{-3} S cm^{-1} for sodium-ion conducting biopolymers prepared using TSP as the host polymer and sodium nitrite as the dopant salt.

The summary of TSP based biopolymer electrolytes are tabulated in table below.

Table 1 Review on TSP based biopolymer electrolytes for electrochemical storage devices

Sl. No	Ionic Dopant	Highest conducting composition	Conductivity (in S cm^{-1}) exhibited by the highest conducting film	Date of Publishing	DOI
1	Ammonium thiocyanate (NH_4SCN)	1g TSP + 0.4 g NH_4SCN	2.85×10^{-4}	Dec-16	https://doi.org/10.1016/j.jnoncrysol.2016.10.008
2	Lithium bromide (LiBr)	1g TSP + 0.4 g LiBr	4.83×10^{-4}	Jan-17	https://doi.org/10.1007/s11581-017-1989-x
3	Ammonium bromide (NH_4Br)	1 g TSP + 0.4 g NH_4Br	1.58×10^{-3}	Nov-17	https://doi.org/10.1016/j.orgel.2017.08.017
4	Lithium chloride (LiCl)	1 g TSP + 0.4 g LiCl	6.7×10^{-3}	Apr-18	https://doi.org/10.1007/s11581-018-2541-3
5	Lithium perchlorate ($LiClO_4$)	1 g TSP + 0.45 g $LiClO_4$	8.77×10^{-4}	Jan-19	https://doi.org/10.1007/s11581-019-02857-1
6	Lithium perchlorate ($LiClO_4$) + ethylene carbonate (EC) plasticizer	1 g TSP + 0.45 g $LiClO_4$+ 0.3 wt.% EC	1.06×10^{-3}	Jan-19	https://doi.org/10.1007/s11581-019-02857-1
7	Magnesium perchlorate ($Mg(ClO_4)_2$)	1 g TSP + 0.25 (m.m.%) of $Mg(ClO_4)_2$	5.66×10^{-4}	Oct-19	https://doi.org/10.1016/j.materresbull.2019.05.015
8	Lithium triflate ($LiCF_3SO_3$)	1 g TSP: 0.45 g $LiCF_3SO_3$	8.37×10^{-4}	Apr-20	https://doi.org/10.1007/s00289-020-03185-5
9	Sodium perchlorate ($NaClO_4$)	1 g TSP + 0.8 g $NaClO_4$	1.70×10^{-3}	Jan-22	https://doi.org/10.1007/s11581-022-04440-7
10	Ammonium formate (NH_4HCO_2)	1 g TSP + 0.35 g NH_4HCO_2	1.23×10^{-3}	Aug-22	https://doi.org/10.1016/j.matlet.2022.132369
11	Sodium nitrite ($NaNO_2$) + Ethylene Carbonate (EC) plasticizer	1 g TSP + 0.7 wt.% $NaNO_2$ + 0.5 wt.% EC	1.49×10^{-3}	Jun-23	https://doi.org/10.1007/s12034-023-02948-w
12	Sodium acetate (CH3COONa)	(TSP: CH3COONa) = 80:20 wt.%	1.95×10^{-4}	Sep-23	https://doi.org/10.1016/j.jnoncrysol.2023.12246

4. Conclusion

Thus, the main objective of the present study is the choice of the biopolymer TSP for the electrochemical storage devices, as TSP offers wider stability and optimized performance as compared to other biopolymers. Also, in the work of Kumar et al. (2024), it was reported that sodium was a compatible material for several biopolymers. Hence, studies on the enhancement of conductivity of sodium doped TSP biopolymer electrolytes shall prove to exhibit better conductivity and ion conduction through the biopolymer segments.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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