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Polyether based electrolyte for energy application

ABSTRACT

Electrolytes are the most significant parts or element of energy storage devices. Polymer electrolytes are known for their wide range of application but, the main problem with polymer electrolytes is their low ionic conductivity. This has attracted the researchers to developed different approaches to increase the ionic conductivity of polymers. Preparation of polymer matrix using solution casting techniques was explained. This review, report the various approaches as well as the result obtained by different studies using polyether-based electrolyte which include Polyethylene oxide (PEO), Polyethylene oxide/Polyethylene glycol (PEO/PEG), Polyphenylene oxide(PPO), Polyethylene Glycol Diacrylate (PEDGA), Polytetramethylene Ether Glycol (PTMEG) etc. by adding different quantity of various salts as a dopant. From the reviewed work, there is significant development of the ionic conductivity reported by different researchers this clearly shows a promising wide application in energy storage devices such as supercapacitor, batteries, fuel cells etc.

Keywords: *polymer electrolytes, polyether, supercapacitor, ionic conductivity*

1. INTRODUCTION

Supercapacitors which are also called ultracapacitors for their high energy storage ability, are energy storage device with high capacity[1]. They serve as bridge between capacitors and batteries with both average energy density and power density[2]. Any supercapacitor has the following main part viz: a material separator used to separate between two electrodes. The performance of a supercapacitor, depend upon the characteristic or the physical properties of the materials used to make both the electrode and electrolyte[2]. A supercapacitor stored charges electrostatically. A supercapacitor has the ability of storing great amount of energy as well as given that huge amount of energy within short period of time. That properties made it to have greater power density compared to batteries. Looking at the energy storage process of supercapacitor, and the movement charge carriers from the electrolyte to the surfaces of electrodes, they can be considered as pseudo capacitors or electric double layer capacitors (EDLCs) [3].

Electric double layer: for this type of supercapacitor, it can store charge either in a non-faradaic process or by forming layer of charges at the interface between the electrode and electrolytes. Initially the charge carriers were randomly distributed throughout the electrolyte. When electrical potential is applied to the high surface area carbon electrodes, the ions start to shift or move in to respective electrode. Positive ions move to cathode and negative ions move to anode where they form a layer charge at the electrode electrolyte interface. This will continue until the supercapacitor is fully charged [3].

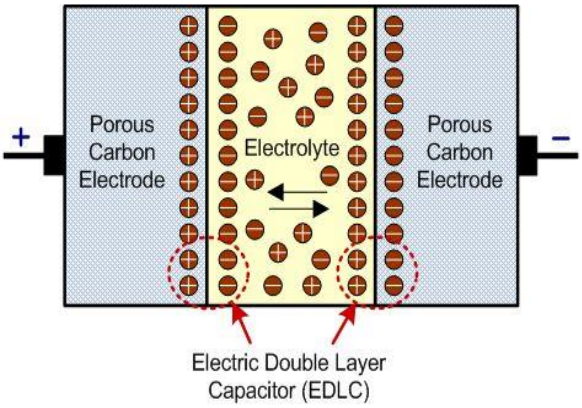

Figure 1. Schematic representation of EDLC [6]

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The capacitance of EDLC can be calculated by considering each double layers as individual conventional capacitor[4]. $C= (C_1 \times C_2)/(C_1+C_2)$ this is done by considering the double layers as series arrangement of capacitors C_1 and C_2. The highest power or maximum power and maximum energy can be calculated as $P_{max} = V^2/(4R)$, and $E_{max}=(CV^2)/2$ respectively where C is the capacitance, V is the voltage and R is equivalent series resistance [5].

Polyether-based electrolytes are really important because they're good at carrying ions and they have a many of good qualities. They have currently drawn muchof attention to be used to replace electrolytes that are liquid for various application in devices for storing energy [7].

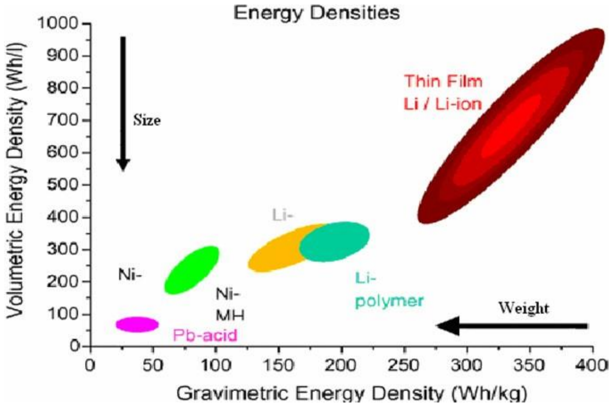

Figure2. Comparison of energy densitie sof different battery systems [8]

They can conduct ions well, are stable when it comes to chemicals, heat, and electricity, are easy to make into big sheets or thin layers, can bend without breaking, and work well with electrodes[8]. Synthesizing electrolytes based on polymers in the form of films required dimensions, given the additional importance of potentially decreasing the internal resistance of the device[9].Ionic conductivity is an essential feature that determines the efficiency of polymer electrolytes, especially in energy storage and conversion devices like supercapacitors, batteries, and fuel cells. It assesses the efficiency of ion movement across the electrolyte when subjected to an electric field, hence influencing the overall efficiency, energy density, and cycling stability of the device. Conductivity is generally elevated in the amorphous phase of the polymer, when chain mobility is enhanced. Consequently, augmenting the amorphous content by additives (such as plasticizers or ionic liquids) or by elevating temperature can substantially improve ionic conductivity.

$$\sigma = \frac{l}{R_b \times A} \qquad (1)$$

Ionic conductivity is represented by the symbol "σ" and is expressed in (S cm^{-1}). The thickness of the polymer film, expressed in centimeters, is represented by the variable "l". The bulk resistance of the polymer electrolytes, expressed in ohms (Ω), is denoted by "R_b". The Nyquist plot yields the value of "R_b" while "A" denotes the surface area of probe in square centimeters (cm^2), under which the sample is put. The values of ionic conductivity for various polyether based electrolyte systems are provided in Table 1.

Polyethylene oxide combined with Na and Ka salts, initially initiated in the year 1973, represented the beginning of ionic conduction of polymer development[10]. Since then, abundance of polymer-based electrolyte has been explored, demonstrating reasonably favorable electrochemical properties enough for electrochemical appliances like SCs. Polymer-based electrolytes are broadly categorized based on different preparation methods during film casting and under various physical conditions[11]. Polyethers are groups of organic substances that are formed by connecting or polymerizing monomers by introducing ether links in between them which provide a repeating unit in the polymer structure[12]. Specifically, polyethers are class of polymer with ether bond in their central chain, (C-O-C)[13]. These polyethers include the following: Polyoxylkylenes(e.g polyethylene glycol PEG, polypropylene glycol PPG, [14]. Poly(ethylene oxide) (PEO), Poly(ethylene oxide-co-propylene oxide) (PEO-PPO), Polytetrahydrofuran (PolyTHF) are also examples of polyethers.

2. METHOD OF PREPARATION

Polymers such as (PEO, PPG, PVA, PEDGMA, PVP, PMMA, PPO etc. are prepared by solution casting method. This involve dissolving them in a solvent such as Acetone, Chloroform, Dichloromethane (DCM), Ethanol and Methanol etc. at different weight ratios. The solution is continuously stirred for some hours at specific temperatures using a magnetic stirrer to ensure proper mixing. After stirring, it is then allowed for solvent used to escape at the room temperature while stirring continues. This process helps to concentrate the polymer solution. The concentrated polymer solution is then cast onto glass petri dishes to form thin films. This is likely done by pouring the solution onto the dishes and allowing it to spread evenly. The thin film specimens are subjected to vacuum drying at some specific temperatures for 24 hours. This step helps to remove any remaining solvent and ensures the formation of solid thin films.

Table 1. Different Polyether based electrolytes with salts and their corresponding conductivities

Sr. No.	System	Salt	Conductivity	Capacity	Ref.
1	Polyethylene oxide PEO	Lithium bis(trifluoromethanesulfonyl) imide (LiTFSI)	1.3×10^{-4} Scm^{-1} 60℃	157 mAh g^{-1} after 40 cycles at 0.2 C	[15]
2	Propylene oxide (PO)	$NaClO_4$	0.2×10^{-4}Scm^{-1} 25℃	-	[16]
3	Polyethylene oxide PEO	Lithium bis(trifluoromethane-sulfonyl)-imide (LiTFSI)	10^{-3} Scm^{-1}at 60 °C	-	[17]
4	Poly(ethylene glycol) (PEO-based networks),	Lithium 3-[(trifluoromethane) sulfonamidosulfonyl]propyl methacrylate (LiMTFSI)	2×10^{-7} Scm^{-1} at 70℃	-	[18]
5	Polyethylene oxide (PEO)-based	LiTFSI.	3.6×10^{-5} Scm^{-1} at 45℃	-	[19]
6	PEO-based electrolyte	Lithium salt LiTFSI	10^{-4} Scm^{-1} at 90 °C	-	[20]
7	PEO,	salt ($LiPF_6$)	2.75×10^{-5} Scm^{-1}	-	[21]
8	Poly(ethylene oxide) (PEO)	lithium triflate ($LiCF_3SO_3$) salt	10^{-8} to 10^{-6} Scm^{-1}	-	[22]
9	PEO-based	LiTFSI	2.4×10^{-4} Scm^{-1} at 60°C	120mAhg^{-1}	[23]
10	PEO/PMMA blend	$LiCF3SO_3$	1.25×10^{-4} Scm^{-1}	-	[24]
11	Blended poly(vinylidene fluoride) (PVDF) /poly(ethylene oxide)(PEO)	Lithium perchlorate ($LiClO_4$)	2.01×10^{-5} Scm^{-1} at 27°C	-	[25]
12	Poly (ethylene oxide) (PEO)-based	LiTFSI	1.24×10^{-4} Scm^{-1} at 40 °C	Discharge ability of 114.6 mAh at 0.2Cg after 200 cycles	[26]
13	PEO-based solid polymer electrolyte	LiTFSI.	1.26×10^{-4} Scm^{-1}	-	[27]
14	PEO_5 and PVDF	Lithium salt	6.44×10^{-4} Scm^{-1} at 303 K	-	[28]
15	(PEG)	KNO_3 salt	8.24×10^{-6} Scm^{-1}	-	[29]
16	PEO/PEG5	$LiClO_4$ salt	1.90×10^{-5} Scm^{-1} at 45 °C	-	[30]
17	POSS–PEG		2.78×10^{-1} Scm^{-1}	-	[31]
18	(PEG-b-PPG-b-PEG	LiTFSI	10^{-3} Scm^{-1} at RT	-	[32]
19	Poly(propylene glycol) (PPG)-based	$Zn(TFSI)_2$ + (LiTFSI)	-	121.7 mAh g^{-1}	[33]
20	PTMEG	LiTFSI	3.77×10^{-3} Smc^{-1} at 30 °C	-	[34]
21	Poly(tetrahydrofuran) (PTHF)	$NaClO_4$ salt	1.3×10^{-4} Scm^{-1}	-	[35]
22	Polytetrahydrofuran (PTHF)	Calcium nitrate salt	1.14×10^{-4} Scm^{-1} at RT	-	[36]
23	Polytetrahydrofuran (PTHF)	$LiBF_4$ salt	10^{-3} Scm^{-1}	-	[37]
25	PS-PEO-PPO-PEO-PS	LLZTO salt	3.10×10^{-4} Scm^{-1} at 28°C	-	[38]
26	PPO	LiFePO	6.67×10^{-4} Scm^{-1} at RT	-	[39]
27	PEGDA	Lithium salts	8.81×10^{-3} Scm^{-1} at 30°C	-	[40]
28	PEGDA	Calcium salts	10^{-5} and 10^{-4} Scm^{-1}	-	[41]
29	Poly(ethylene glycol) (PEG)	Lithium salts	2.39×10^{-3} Scm^{-1}	-	[42]
30	PEO	$LiFePO_4$	0.19×10^{-1} Scm^{-1}	141.5 mAh g^{-1} at 0.03 C.	[43]
31	PEO:MC	Ammonium iodide (NH4I)	7.62×10^{-5} Scm^{-1} at 303K	-	[44]

Different concentrations of salt in percentage by weight are break down into specific solutions involving the blended polymer. This step aims to introduce ions into the polymer matrix, thereby enhancing its ionic conductivity and electrolytic properties. Similar process of solution casting method as stated earlier is done again to prepare a new sequence of polymer blend electrolyte systems, now containing the dopant. The solution is cast onto glass petri dishes and subjected to vacuum drying to form a thin film sample.

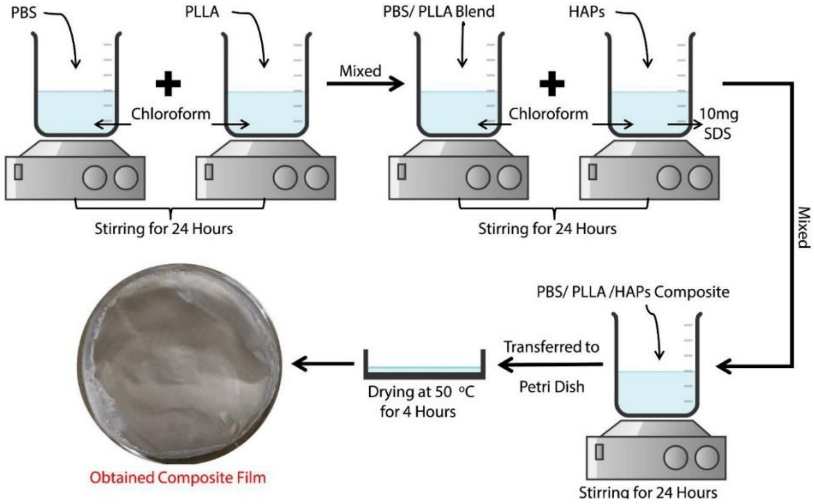

Figure 3. Illustration of solution casting technique [46]

3. RESULTS AND DISCUSSIONS

H chen et al [45], in their research they synthesized and characterized composite all-solid-state electrolytes which are flexible that can be applied in all-solid-state-batteries (ASSBs). They prepared the electrolytes by using oxygen-vacancy-rich Ca-doped CeO$_2$ nanotubes, lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) salt and polyethylene oxide PEO. This was done in order to achieve stable and seamless interface between solid parts of (ASSBs) which are very important to achieve high ionic conductivity as well as high-rate working of the device. According to them, the PEO contribute to the flexibility of the system and also the stable contact between the electrolyte and solid electrodes in the all-solid state-battery (ASSBs). In their research, the maximum conductivity was found to be 1.3×10^{-4}Scm^{-1} and the transference number for Li-ion to 0.453 with maximum voltage stability. The electrochemical characterization as well as density function theory computations shows that the system provide some free lithium ions which help in increasing the conductivity. According to them, by adding the doping level of the system, it corresponds to the increase in roughness of the surface. The scanning electron microscope (SEM) and transmission electron microscope (TEM) were used to study the structure morphology of the sample. With high resolution transmission electron microscope (TEM), the lattice point having a spacing of 0.3205 nanometers is equivalent to plane with indices (111) of the CeO$_2$ signifying Ca^{2+} doping, that substitutes the Ce^{4+} and expand the interplanar spacing in comparison with pure samples.

Y. Zang, et al [23], have conducted similar work and where they focused on finding the effect of different composition of LiTFSI salts on the diverse properties that solid polymer electrolytes have (SPEs) for (ASSBs). Most important parameters studied were interfacial properties and ionic conductivity which are very important for the working or performance of the battery ASSBs. According to their study the solid-state polymer electrolytes with LiTFSI show the maximum or highest conductivities with the values as 5.3 × 10^{-6} Scm^{-1} at 25 °C and 2.4 × 10^{-4} Scm^{-1} at 60 °C with high discharging ability which can be associated to the factors such as low crystallinity and large lithium-ion transference number.On the other hand, SPEs with LiFSI demonstrate cycling performance with large interfacial properties, attributed with the formed dense with flat lithium fluoride solid electrolyte interface (LiF SEI) layer subsequent to each cycling. Their results show that for polyethylene oxide-based all-solid-state batteries, LiTFSI help to provide high discharge capacity and LiFSI can be applied to produce high retention rate.

K.P.Sindhu et al [24], conducted a research on the nanocomposite solid polyelectrolytes consisting polyethylene/blended PMMA serving for root polymer system, and they used LiCF$_3$SO$_3$ as the salt for electrolytes. They also used Barium titanate

($BaTiO_3$) as nanofiller. Fourier Transform Infrared Spectroscopy showed the existing interaction in PEO/PMMA/$LiCF_3SO_3$ combined with $BaTiO_3$ nanoparticles. X-ray Diffraction (XRD) together with Differential Scanning Calorimetry (DSC) analysis have shown a clear decrease in the crystal nature of the main polymers in the solid nanocomposite polyelectrolytes. The dielectric constant of used nanocomposite polyelectrolytes is improved. This improvement can be associated to the incorporation of $BaTiO_3$ nanoparticles. The observed maximum dielectric and conductivity were found at 3 % by weight of the $BaTiO_3$.They obtained the room temperature ionic conductivity to be 1.25×10^{-4} Scm^{-1} which shows the hopeful implication of the improved system electronics that are flexible. The ASSLIBs based on polymer electrolyte, shows safety and great energy density, but the problems of low ionic conductivity for lithium ion terminate their application, Hui. Wang et al[19], suggest for a new way to develop the PEO conductivity based solid electrolyte. In their method, the initially prepared the system by solution casting method followed by exertion of ultra sound on the electrolyte to adjust its structure. By analyzing the performance of the electrolyte, they observed that treating the electrolyte by ultrasonic, an increment of 78% of the conductivity was observed. The electrolytes' impedance was found to reduced 1960 ohms to 1100 ohms as a result of treating with ultrasonic at 25℃ which was accompanied by 44% decrement. And there was an increment of the conductivity from (1.8 to 3.2) × $10^{-6}Scm^{-1}$ at 25℃, which was found to be developed significantly with 78 percent by treating with ultrasonic.by increasing temperature to 45degree Celsius, ionic conductivity was found to be maximum of 3.6×10^{-5} Scm^{-1} at 45℃ because of the ultrasonic treatment. Increasing the temperature caused the PEO to melt and all the chain were loosed and hence treating with ultrasonic had small effect on the electrolytes. When cooled to room temperature, the electrolytes lost the increment in conductivity totally.

Hawzhin T. Ahmed and Omed Gh. Abdullah [44], prepared a proton conducting polymer electrolytes using the mixture of polyethylene oxide, PEO and various composition of ammonium iodide salt as a dopant (NH_4I). they used solution casting techniques which involve dissolving various component into a suitable solvent such as acetol and then casting it on substrate or Petridish to form the film. The characterization of the samples were done and analyzed using X-ray diffraction, polarized optical microscope to study the crystallinity of the sample. According them, the XRD and POM result showed the sample that has 30% by weight of ammonium salt, has lowest crystallinity which can be considered as a result of the amorphous phase introduced by the dopant salt. Using Fourier Transform, the interactions between the molecules of the polymer and that of ammonium iodide salt were studied. The shift in the position of N-H stretching band shows the interaction between the polymer and the ammonium iodide salt. They observed the maximum conductivity at a temperature of 303k with 30% of the ammonium iodide salt to be 7.62×10^{-5} Scm^{-1} using electrical impedance spectroscopy (EIS).

4. CONCLUSION

Looking at different approaches to develop the ions conduction of the polymer electrolytes specifically polyether including PEO, PPG, PEGDA, PPO etc. researchers had developed different method of blending polymers, adding dopants (salts) to the host polymer as summarized in table one above. Different results obtained have shown the current development and potential improvement for polymer-based electrolytes for energy storage application. Although there is improvement, but still there are many gaps to further our study in such area. These gaps include, compatibility with electrode materials, optimization of electrolyte composition, long-term stability and durability optimization for specifications many studies have shown high-compatibility and voltage stability for ASSBs based on the developed composite electrolyte, however, optimizing electrolyte composition and structure for specification applications including portable electronics or electric vehicles, looking at factors like energy density cycling stability and safety will give area for further research. The maximum ionic conductivity can be achieved through doping with different salt compositions.

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Conflict of Interest

The authors declare no conflict of interest.

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IZVOD

ELEKTROLIT NA BAZI POLIETRA ZA ENERGETIKU

Elektroliti su najznačajniji delovi ili element uređaja za skladištenje energije. Polimerni elektroliti su poznati po širokom spektru primene, ali glavni problem sa polimernim elektrolitima je njihova niska jonska provodljivost. Ovo je privuklo istraživače da razviju različite pristupe za povećanje jonske provodljivosti polimera. Objašnjena je priprema polimerne matrice primenom tehnika livenja rastvora. Ovaj pregled prikazuje različSite pristupe kao i rezultate dobijene različitim studijama korišćenjem elektrolita na bazi polietra koji uključuju polietilen oksid (PEO), polietilen oksid/polietilen glikol (PEO/PEG), polifenilen oksid (PPO), polietilen glikol dijakrilat (PEDGA), politetrametilen etar glikol (PTMEG) itd. dodavanjem različite količine različitih soli kao dopanta. Iz pregledanog rada, postoji značajan razvoj jonske provodljivosti koji su prijavili različiti istraživači, što jasno pokazuje obećavajuću široku primenu u uređajima za skladištenje energije kao što su superkondenzatori, baterije, gorivne ćelije itd.

Ključne reči: *polimerni elektroliti, polietar, superkondenzator, jonska provodljivost*

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