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Structural, morphological, and electrical properties of safe PEO electrolyte

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ABSTRACT

This work aims to fabricate and characterize polymer composites as electrolyte materials. Using the solution casting process, lithium chloride (LiCl) salt concentrations of 5, 8, 10, 12, and 15% wt/wt were dissolved in a solution of acetonitrile and polyethylene oxide to create Polyethylene Oxide (PEO) electrolyte. After that, the electrolytes produced are characterized to examine their properties. A tension test was used to investigate the mechanical properties, X-ray diffraction (XRD) and scanning electron microscopy (SEM) were used to study the structural properties, and UV VIS and Fourier-transform infrared spectroscopy (FTIR) were used to study the optical properties. The DC and AC tests were used to examine the electrical properties. XRD results show the presence of two distinct Bragg peaks (120) and (112) at 19.15° and 23.35° respectively in the pure PEO XRD pattern suggesting that the polymer has a semi-crystalline structure. From the XRD studies, the electrolyte's crystallinity has deteriorated as a result of the rising concentration of lithium chloride. According to the SEM data, the micrographs demonstrate the crystalline nature and amorphous boundaries of the polymer electrolytes. The PEO electrolytes' surfaces show extensive pore architectures and interconnected network topologies. The results of the tension test indicated that the young modulus was 1.95 MPa at 8% LiCl concentration, the optimal ultimate strength was 22.5 MPa at 8% LiCl concentration, and the optimum elongation was 667% likewise at 8% LiCl concentration. The polymer and lithium chloride demonstrated good complexing, according to the FTIR data. According to the UV-VIS test, an increase in lithium chloride concentration causes the band gap to shrink by 3.7 eV for 12% LiCl, which is subsequently exacerbated by an increase in salt concentration. When the ionic conductivity was examined under temperature changes between 40 ˚C and 65˚C for PEO, the highest conductivity was 2.99×10^{-5} S/cm at 40˚C using 15% LiCl concentration.

Keywords: *Solid state composite electrolytes, Polyethylene oxide, Solid polymeric electrolytes (SPE), Ionic conductivity*

1. INTRODUCTION

Due to their potential use in electrochemical devices like batteries, polymer electrolytes have gained more and more attention [1–4]. Along with other attributes, they have good electrical conductivity and exceptional thermal stability [6]. To achieve high ionic conductivity, alkaline salts are commonly added to them [7–10]. Of all the polymer hosts, PEO has attracted the greatest attention due to its superior ability to solvate salt and form more stable complexes with inorganic salts than any other polymer [11]. A polymer electrolyte's ionic conductivity is strongly dependent on the amount of salt present, but it's unclear if all of the salt's ions participate in the conduction process. Despite the enormous application potential of composite polymer electrolytes, understanding the ion transport and relaxation mechanisms in these materials remains critical. PEO chains encircle Li ions in polymer electrolytes made from PEO-lithium salts. The O-atom in the PEO chain serves as a coordination site for Li ions via Lewis acid-base interaction [10], and Li ions alternate between coordination sites during the amorphous phase. Ion transport in polymer electrolytes is complicated by the polymer host chain mobility, which is required for ion transport. Numerous experimental findings, including modeling studies [12–17], using NMR, neutron scattering, positron lifetime, and other techniques, indicate that polymer segmental motions influence ion transport behavior in polymer electrolytes. Continued research into the dielectric relaxation behavior of ion-conducting polymer composite materials is required to understand their ionic transport properties. Conductivity relaxation spectroscopy can help us understand segmental relaxation as well as ion dynamics. It is also necessary to consider how the salt content influences the dynamics of relaxation [18].

We present an ac relaxation for different concentrations of lithium ions in the PEO/LiCl polymer electrolyte. The system we chose to investigate ac relaxation has the highest conductivity of any electrolyte. The analysis of the electric modulus spectra of the PEO-LiCl electrolyte reveals that the relaxation peak is broad, diverges from Debye-type relaxation, and has long relaxation periods. To comprehend the effects of composition and temperature on the ion dynamics of these electrolytes, scaling of the modulus spectra has also been performed.

2. EXPERIMENTAL PROCEDURE

2.1. Materials

Lithium chloride (LiCl) was purchased from Aldrich (Germany) to be used as the electrolyte solution, and acetonitrile (Germany) was used as a solvent for the PEO. Polyethylene oxide (PEO) with a molecular weight of 3 megamoles was used as the matrix phase for the electrolyte.

2.2. Sample Preparation

The preparation of PEO electrolyte electrolytes involves three stages, the polymer solution preparation, electrolyte solution preparation, and mixing of electrolyte solution with polymer solution.

2.2.1. Polymer Solution Preparation

Polyethylene oxide electrolyte membranes were synthesized using the solution casting method. To prepare the membrane, 3 g of polyethylene oxide powder in 150 ml of acetonitrile was used to dissolve, and a magnetic stirrer to mix at ambient temperature for 2 hours.

2.2.2. Electrolyte Preparation

Different weight ratios (5, 8, 10, 10, and 15%) of lithium chloride (LiCl) were used to prepare the electrolyte solution. Using a magnetic stirrer at room temperature, the salt was dissolved in 10 ml of highly concentrated ethanol and swirled for an hour.

2.2.3. Mixing of Electrolyte Solution with Polymer Solution

After producing the clear polymer solution and electrolyte solution, the two solutions must be mixed thoroughly for 1 hour to obtain the polymer electrolytes. After producing a homogenous solution, the polymer solution is cast into a 15 cm petri dish and left at room temperature for drying until a membrane is formed, dry out time is between 24–48 hours according to humidity. After drying out of the membrane, it can be easily removed from the petri dish.

2.3. Characterization

The X-ray device XRD-6000 model, manufactured by Shmiadzu in Japan, was used to study structural qualities. Below is a list of its requirements: Cu, NF type X-ray tube Radius of scanning: 185 mm Less than 2.5 Sv/h at maximum output for leakage X-rays. The morphological features of the PEO electrolytes utilized the SEM (MIRA 3 TESCAN) to conduct the investigations. The AC Tests were carried out using a homemade system for measuring the AC conductivity at different frequencies.

3. RESULTS AND DISCUSSION

This section shows the results of our prepared refractories.

3.1. X-ray diffraction (XRD) Analysis

Figure 1 shows the patterns of PEO's X-ray diffraction (XRD) films. The presence of two distinct Bragg peaks (120) and (112) at 19.15° and 23.35° in the pure PEO XRD pattern suggests that the polymer has a semi-crystalline structure [19]. Pure PEO is caused by high crystallinity, which is demonstrated by high-intensity peaks in the XRD pattern that represent pure PEO. Adding lithium chloride salt caused the PEO's crystallinity to deteriorate, which caused the ionic conductivity to rise. The sharpness of the semicrystalline PEO peaks is diminished, indicating an expansion of the amorphous phase, as a result of the disintegration of the ordered arrangements of the PEO chains. Since the polymer system PEO-LiCl has an amorphous improvement, the conductivity of the samples will change. The acquired amorphous phase lowers the energy barrier for the segmental motion of polymer chains. Consequently, as the amorphous region of the sample expands, the conductivity increases [20].

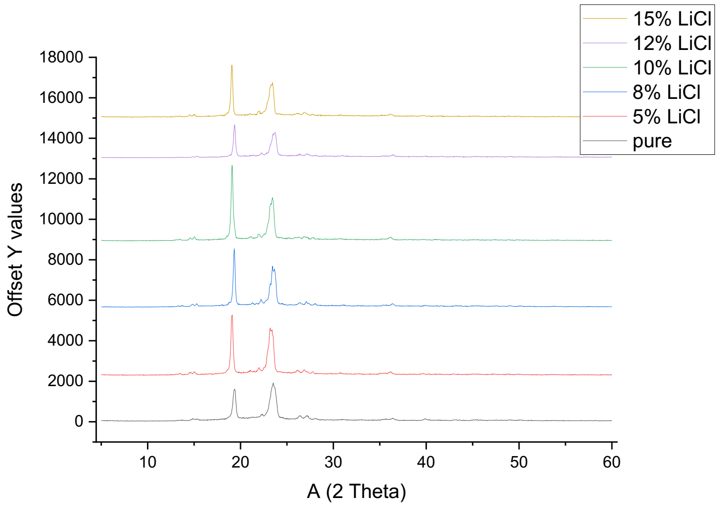

Figure 1. XRD graphs of the prepared PEO membranes at different lithium chloride weights

At ambient temperature, the specimen with the highest quantity of amorphous domain is expected to have the highest electrical conductivity [21, 22]. To demonstrate this improvement, an analysis of the sample's electrical conductivity was conducted.

3.2. SEM analysis

Scanning electron micrographs of PEO with different Lithium Chloride ratios (5, 8, 10, 12, and 15%) were shown respectively in Figure 2, the micrographs of the polymer-salt complexes clearly show the presence of unique spherulites with a distinctive lamellar microstructure. An amorphous phase is seen between spherulite borders. The micrographs demonstrate the crystalline nature and amorphous boundaries of the polymer electrolytes. With varied polymer-to-salt ratios, the surface shape does not vary consistently. Images clearly show that PEO electrolytes have a rich pore structure and a tangled network structure on their surface. This agrees with other studies [23].

3.3. Mechanical Properties

In this test polyethylene oxide electrolytes were made and diffused with lithium chloride at various concentrations typically, 5, 8, 10, 12, and 15% of weight ratios. The stress-strain behavior shows the visco-elastic behavior of the prepared membranes due to the hermetically low modulus of elasticity. From the graphs, we can observe the activity of the lithium chloride on the mechanical properties of the polyethylene oxide electrolytes. As seen, the tensile strength is at maximum when the membrane is lithiumchloride free, but then decreases dramatically when the salt is added, this is due to the plasticizing capabilities of the lithium chloride, we observed a slight increase at 8% weight ratio then extreme decrease was observed at higher concentrations. This behavior is spotted elsewhere [24].

The mechanical stability of the prepared polymeric electrolytes was tested and measured using the tensile machine to measure the ultimate strength, elongation, and Young's modulus, the results are listed in Table 1. The tensile stress-strain of the pure PEO is shown in Figure 3. As can be seen from Figure 4, the ultimate strength is 37.14 MPa, and Young's modulus was 4.48 MPa which is the standard value of the PEO polymer. The elongation was 54%. Upon addition of lithium chloride at a concentration of 5% as can be seen in Figure 4, the ultimate strength was decreased to 20 MPa, then in Figure 6, the elongation was increased to 155%, Young's modulus was also decreased 2.38 MPa as seen in Figure 5, and this is attributed to the plasticizing effect of the lithium salt. With a further increase of lithium chloride at 8% concentration, The PEO

Table 1. Mechanical properties of PEO/LiCl electrolytes

Polymer system	UT (MPa)	EI %	E (MPa)
0	37.230	39.640	3.422
5	20.200	144.433	2.481
8	25.544	667.100	1.953
10	10.730	644.300	0.962
12	7.720	181.400	1.052
15	6.510	585.300	0.630

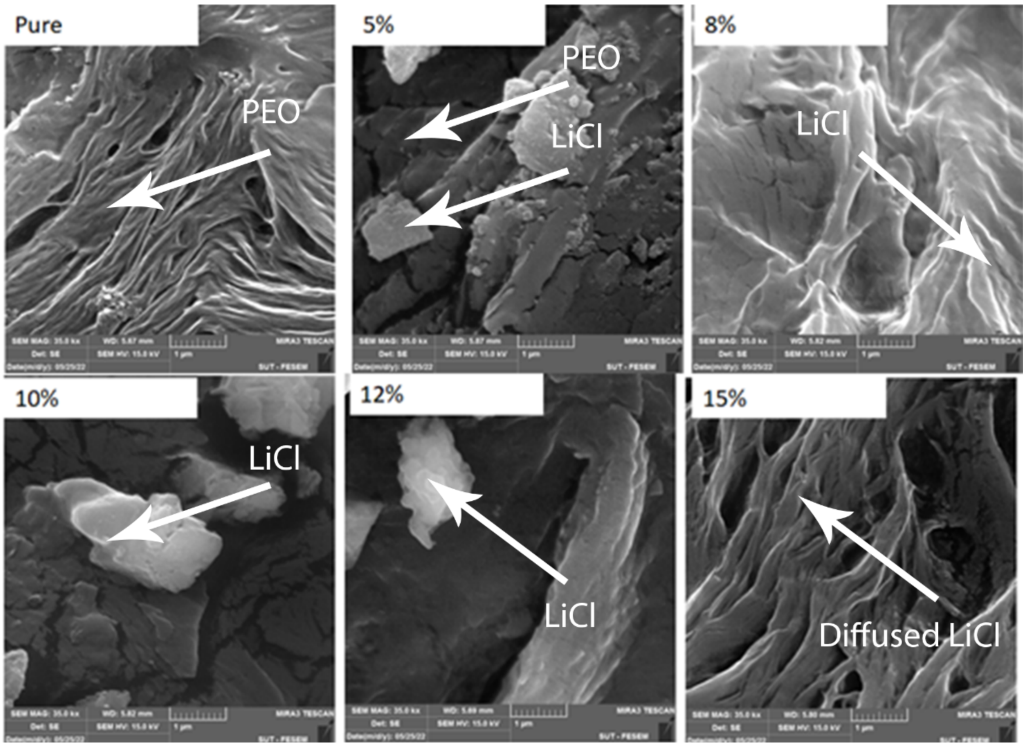

Figure 2. SEM images of the prepared PEO membranes at different lithium chloride weight ratios

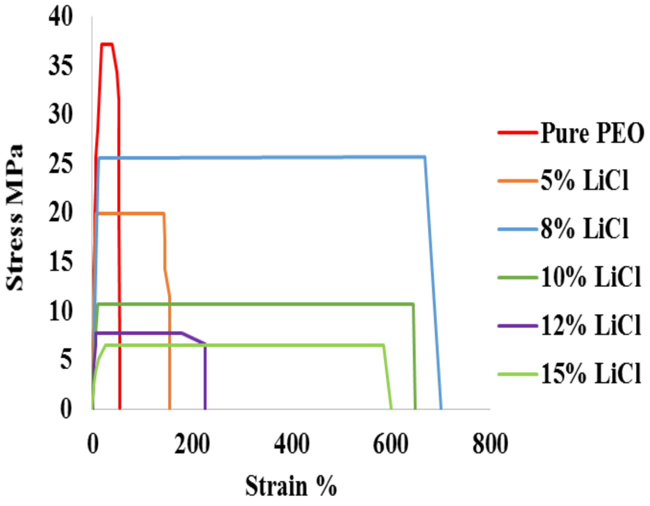

Figure 3. Stress-strain behavior of the PEO electrolytes

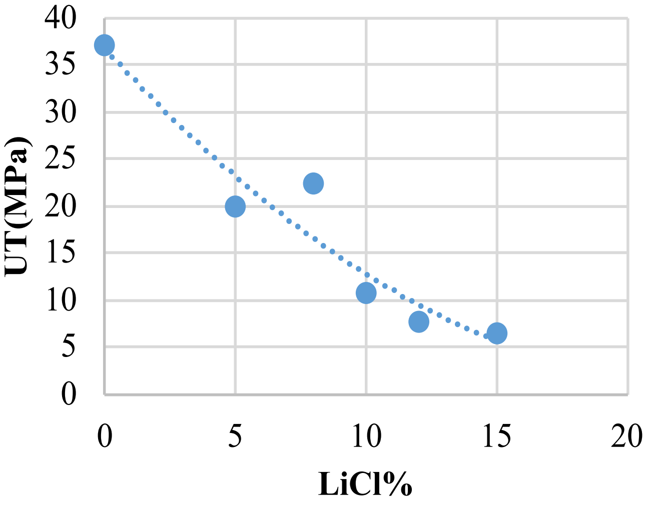

Figure 4. Ultimate strength of PEO electrolytes

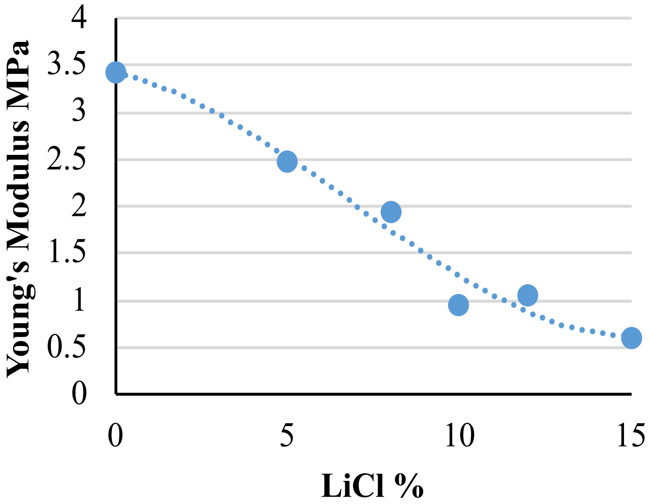

Figure 5. Young modulus of PEO electrolytes

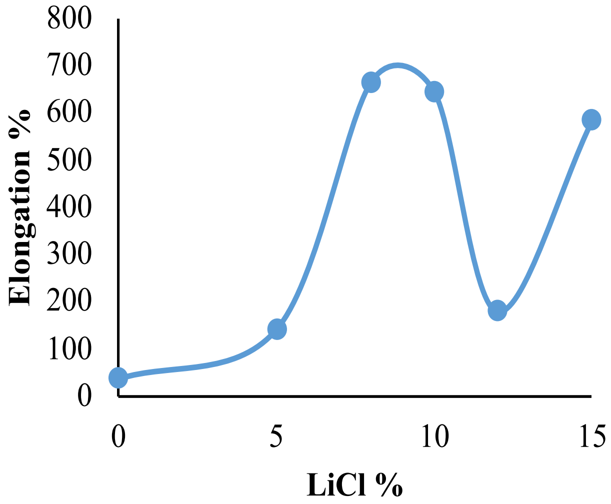

Figure 6. Elongation percentage of PEO electrolytes

electrolyte's mechanical characteristics slightly improved. As seen in Figure 4 and Figure 5, for example, the ultimate strength was increased to 25.7 MPa, Young's modulus was increased to 2.85 MPa, and elongation was hermetically increased to 669% as seen in Figure 6, this is explained by the good curling of the polymer chains made of PEO that lead to tight crosslinking between the salt and the polymeric chains, this lead to tight chain coils, and also due to good complexing [23].

Figures 4 and 5, the ultimate strength was decreased, as well as the Young's modulus, this was attributed to the destruction of the hydrogen bonds by the increase of the salt [23].

3.4. Fourier Transform

The FTIR spectrum of pure PEO is shown in Figure 7. These bands of PEO can be observed at 3564 cm^{-1} which can be traced to OH stretch, 2873 cm^{-1} can be attributed to CH stretching mode, 2164 cm^{-1} to C=C stretch, asymmetric stretching mode at 1963.54 cm^{-1}, 1465.90 cm^{-1} caused by C=C stretch, 1338.60 cm^{-1} by C–O–C stretch, 1276.88 cm^{-1} also due to C–O–C stretch, 1095.57 cm^{-1} due to C–O–C (stretching mode), CH2 twisting mode at 956.69 cm^{-1}, CH$_2$ wagging mode at 840.97 cm^{-1}, this agrees with other study [25].

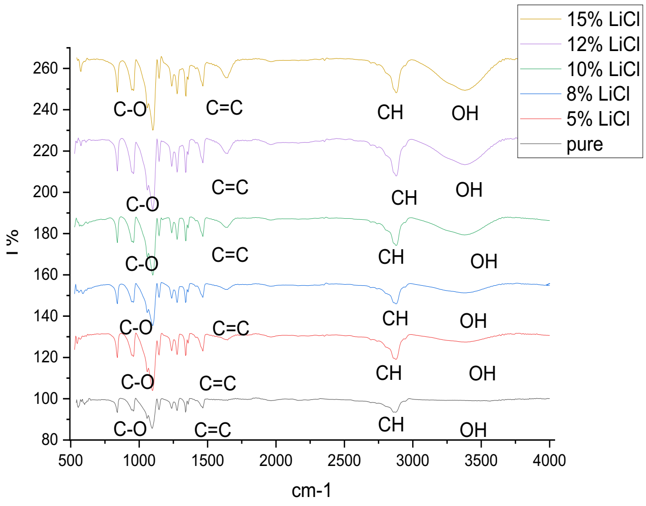

Figure 7. FTIR spectra for Pure PEO; and different PEO: LiCl weight ratio

The peak of 555.50 cm^{-1} was probably occurred by O–C–O bending. A 5% lithium chloride addition into the PEO matrix has led to a shift of the vibrational peaks as shown in Figure 7, and this agrees with another study [16]. As can be seen in Figure 7, the addition of the lithium salt has caused a shift in the vibrational peaks, typically from 2873.94 to 2877.79 cm^{-1}, also a peak at 2164 cm^{-1} was disappeared because of salt addition. Peaks at bands of 1963 cm^{-1} were also shifted by the action of asymmetric stretching by the action of lithium salt. A new peak was observed at 1647.21 cm^{-1} caused by lithium chloride, other peaks remained unchanged. At 8% lithium chloride, the intensity of the peaks was increased furthermore as shown in Figure 7. At 10% LiCl, the intensity of the peak of 3375.43 cm^{-1} was increased due to OH stretch, other peaks remained unchanged as seen in Figure 7. At a concentration of 12% and up to 15% of LiCl, the peak 3379.29 cm^{-1} was shifted from 3375.43 cm^{-1}, also the peak 1643.35 cm^{-1} was shifted to 1647.21 cm^{-1}, majority of other peaks remained unchanged, as shown in Figure 7.

3.5. UV-VIS Properties

Optical transmission and absorbance are essential in calculating the optical properties of polymeric electrolytes. Higher transparency in the visible wavelength is required in applications for optoelectronic devices. As a characterization method for the conductive materials placed onto transparent surfaces, the UV-Vis-NIR spectrophotometer is employed. The method is based on the interaction of UV, Vis, and NIR electromagnetic radiation with the substance.

Figure 8 shows the absorbance (A) as a function of wavelength in the range of 180 to 600 nm for PEO electrolytes synthesized by the solution casting method at different LiCl concentrations. From the figure, it's obvious that there are absorption peaks at wavelengths from 180 nm belonging to the ultraviolet region, then this absorption peak decreases as the wavelength increases in the wavelength region from 200 to 580 nm. This property in the UV range is considered to be valuable for optoelectronic applications therefore the PEO membranes proved to be a good UV absorber and that will make it a great choice for optoelectronic applications. The energy band gap was obtained by extrapolation of the $(\alpha h\nu)^2$ plot's straight line vs. the energy of the photon ($h\nu$). The direct band gap is indicated by the linear relationship of $(\alpha h\nu)^2$ and ($h\nu$). It was observed that the band gap of the pure PEO is high because of the dielectric nature of the PEO, then a slight decrease as observed upon LiCl addition, the band gap was increased at concentrations higher than 10% of LiCl as shown in Figure 10.

Table 2. Calculated bandgap of PEO membranes

Sample	Bandgap (EV)
Pure PEO	4.3
5% LiCl	4.6
8% LiCl	4.6
10% LiCl	4.2
12% LiCl	3.7
15% LiCl	4.3

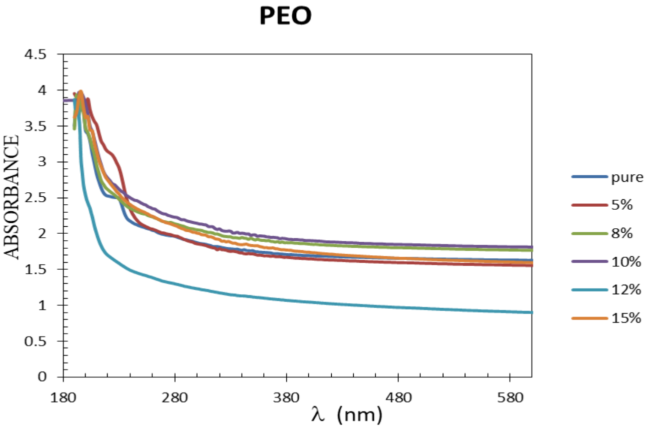

Figure 8. Absorbance of the PEO electrolyte at different LiCl concentrations

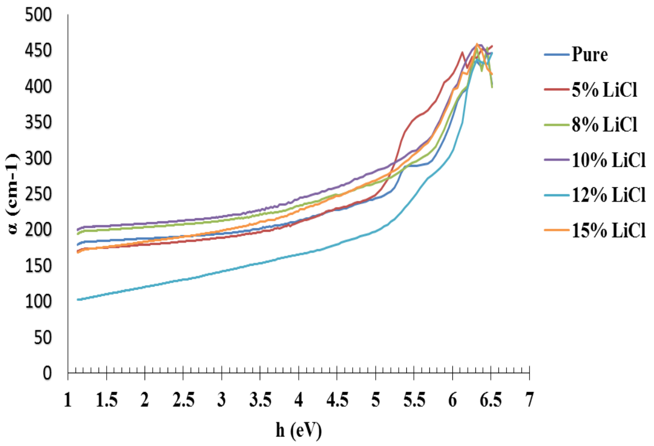

Figure 9. Optical absorption coefficient of the PEO electrolyte at different LiCl concentrations

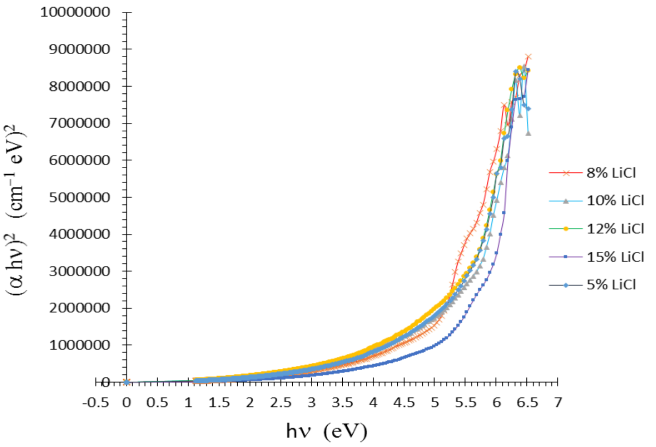

Figure 10. Variation of $(\alpha h\nu)^2$ versus hv of the PEO electrolyte at different LiCl concentrations

3.6. Electrical Properties

3.6.1. DC Conductivity

DC conductivity test was used to measure the DC conductivity of lithium chloride concentration increase. The ionic conductivities of the PEO electrolytes at various LiCl concentrations are shown in Table 3 and Figure 11. The ionic conductivity of each electrolyte increases as the temperature rises.

The major reason for this is that high temperatures encourage the movement of carrier ions. The electrolyte salt and the polymer matrix have the biggest effects on the ionic conductivity of PEO. Coordination of lithium ions with carbonyl groups' oxygen atoms in the PEO electrolyte, as well as the ions that are only loosely bound. The molecular chain will split due to this, as a result of an electric field. Ionic conductivity is the result of cations moving via the coordination sites of carbonyl oxygen atoms. PEO electrolytes have a high concentration of the action of carboxyl groups as sites of lithium ions with oxygen atoms to coordinate, improving the conductivity of ions. The lowest ionic conductivity is achieved by PEO when 5 wt% LiCl solution is added, due to the absence of Li+ ions. Due to the high ion concentration, conductivity increases with increasing lithium content, however, this results in a loss in mechanical strength. This is mostly caused by the abundance of salt that restricts the flow of polymer chains and lithium ions and damages the network structure of the polymer. The conductivity was higher than elsewhere [26].

Table 3. DC conductivity of the polyethylene oxide electrolytes at different LiCl weight ratios

Sample	DC Conductivity at 40°C (S/cm)
Pure PEO	4.75×10^{-8}
5% LiCl	2.72×10^{-6}
8% LiCl	1.81×10^{-6}
10% LiCl	9.72×10^{-7}
12% LiCl	7.29×10^{-6}
15% LiCl	2.99×10^{-5}

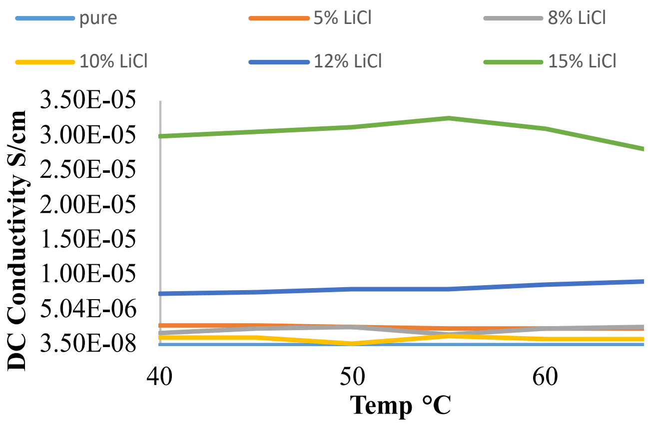

Figure 11. Ionic conductivity of PEO electrolytes at different salt concentrations

3.6.2. AC Conductivity

Using an impedance analyzer 429A, the electrical properties of PEO were compared before and after adding LiCl salt at various weight ratios (5, 8, 10, 12, and 15%). AC conductivity (AC) was measured at room temperature using a frequency range of 1 Hz to 1 KHz. Calculate electrical conductivity using the current's resistance, length, and area. The resistivity equation is p = RA/l, where p is resistivity, R is resistance, A is area, and l is length. The formula for conductivity is s = 1/p, where s represents conductivity. The electrical properties of PEO were investigated using an impedance analyzer 429A before and after the addition of LiCl salt at various weight ratios (5, 8, 10, 12, and 15%). The AC measurements were taken for fixed temperature and variable frequencies of a range of 1 Hz to 1 KHz. It included experimental data and Win Fit software for data processing and visualization. To calculate electrical conductivity, consider the resistance, length, and area of the current. The provided equation, p=RA/l, is a function of resistivity and can be used to calculate length, resistivity, resistance, and area. The formula for conductivity is s = 1/p, where s represents conductivity. Figure 12 shows the A.C. Conductivity fluctuations in PEO at various weight ratios (5. 8, 10, 12, and 15 Wt%) before and after LiCl addition. The conductivity of polar polymers varies with frequency [27–29]. According to [27, 28], the term "hopping term" refers to a charge carrier in a localized state that was excited by other charge carriers in the localized state and transmitted to the conduction band. We call this abrupt shift of charge carriers from one location to another, "hopping." It provides a way through mechanical quantum tunnels and potential barriers, and it is generally in good agreement with [30]. AC conductivity was attributed to leaping electrons within the polymer chain's redox sites as well as delocalized electron movements. The results showed that conductivity increased significantly as frequency increased. The increase in A.C. conductivity was caused by accelerating the hopping electron method to its maximum frequency and increasing the weight LiCl ratio, this is due to the reduction of ether bonds, which produces lithium alkoxides that are responsible for Li⁺ transport [28]. Figure 12 illustrates the

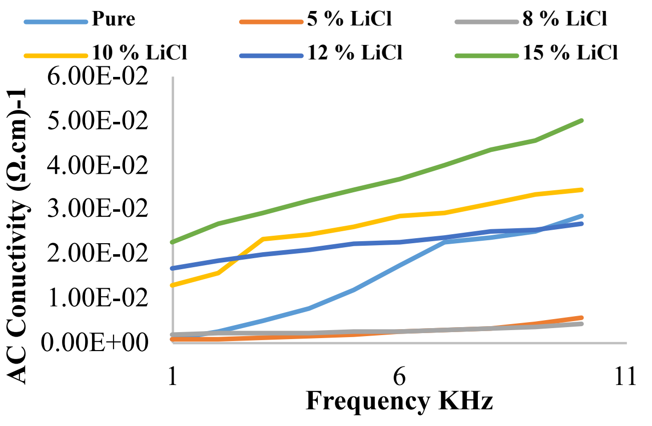

Figure 12. AC conductivity of the PEO electrolytes at different LiCl concentrations

variation in conductivity of alternating current (AC) with frequency. We discovered that these variations are determined by the two frequency-varying stages of relaxation. The first relaxation process called (-relaxation), and the second relaxation process, known as side motions in groups or changes within a collection of polymer matrices, were brought about by the polymer chain micro-Brownion motion in the crystalline and amorphous regions of the PEO [31–33].

4. CONCLUSION

Polyethylene oxide electrolyte films were successfully prepared using the solution casting method at room temperature without the use of high temperatures as other routes required. The prepared electrolytes were safe and fire hazard-free. The increase of lithium chloride and graphite weight ratios about the matrix weight has led to a decrease in the mechanical strength of the electrolyte. The increase in Lithium Chloride (LiCl) concentration proved to enhance the conductivity of the prepared electrolytes. It was concluded that to achieve high conductivity, the polyethylene oxide-based amorphous phase of the polymer matrix must be raised to a level that allows segmental motion near the polymer matrix, allowing the lithium ions to move freely. This can be done by increasing a dispersion phase like lithium salt. The prepared polyethylene oxide electrolytes can be employed for energy storage applications, it can also be deduced from the results. The prepared electrolytes can be used for the fabrication of highly flexible electronic devices such as flexible batteries and fuel cells due to the high elongation of the prepared samples, thanks to the visco-elastic behavior of the polyethylene oxide polymer.

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