JOURNAL of the Turkish Chemical Society
Section: A
Chemistry

TURKISH CHEMICAL SOCIETY

Studies of the Properties of New Electrospun Based on PVA-PEG Polymer Systems Electrolytes for Energy Storage Devices

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Abstract: Solid polymer electrolytes (SPEs) have been considerably investigated due to various electrochemical device applications. Most of the SPEs comprise polymer as a host material to provide strength and good mechanical stability and salt that transfers charge carriers to cause conductivity. Nanocomposite solid polymer electrolyte membranes based on poly(vinyl alcohol) (PVA)-poly(ethylene glycol) (PEG) blend complexed with $LiClO_4$ and nanofillers Al_2O_3 at different weight percent ratios have been obtained by using electrospinning method. The conductivity and structural properties of the different systems have been characterized by using various experimental approaches such as X-ray diffraction (XRD) and Fourier transform infrared FTIR spectroscopy. The ionic conductivity of the systems has been measured by using an LCR meter in a temperature ranging from 298 to 353 K. Maximum ionic conductivity of 1.58×10^{-4} S cm^{-1} at room temperature has been observed for the system of PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10) with 15 wt% weight percent of $LiClO_4$ salt in PVA-PEG blend matrix. The ac conductivity report indicates that the ionic conductivity of the PVA-PEG-$LiClO_4$-Al_2O_3 complex is influenced by the concentration of $LiClO_4$. The effect of temperature on the ionic conductivity of polymer electrolyte complexes has been estimated by changing the temperature ranging from 298 to 353 K. However, the conductivity of the nanofiber polymer electrolyte systems increases with the rise of temperature, and the maximum conductivity of 1.58×10^{-2} S cm^{-1} has been recorded at 353 K. The temperature-dependent conductivity follows the Arrhenius behavior.

Keywords: Complex polymer electrolytes, ionic conductivity, PVA-PEG, polymer blend, $LiClO_4$, nanocomposite.

Submitted: March 9, 2023. **Accepted:** November 17, 2023.

Cite this: Gul Khattak R, Mahmood WAK. Studies of the Properties of New Electrospun Based on PVA-PEG Polymer Systems Electrolytes for Energy Storage Devices. JOTCSA. 2024;11(1):313-20.

DOI: https://doi.org/10.18596/jotcsa.1262560

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1. INTRODUCTION

Solid polymer electrolytes (SPEs) possessing high ionic conductivity have been a subject of considerable attention in recent years because of their wide potential applications in various electrochemical devices such as solid-state rechargeable batteries, chemical sensors, fuel cells, super-capacitors, computers, electrochromic display devices, smart windows and other portable electrochemical devices applications (1-4). These polymer electrolytes have become prominent materials because they can satisfy several requirements due to their special properties including lightweight, high ionic conductivity, good electrode-electrolyte contact, excellent electrochemical stability, flexibility, good mechanical properties, and easy mouldability (5-6). The low cost of SPEs and their easy processibility make them a promising candidate for a wide variety of applications. However, the low ionic conductivity of the polymer-based materials at room temperature has limited their applications in electrochemical devices. The major efforts in this field have been focused on developing new advanced materials based on polymers having high ionic conductivity values with high mechanical, thermal, and electrochemical stability (7-9). Various experimental techniques, such as copolymerization, plasticization, blending, and addition of ceramic fillers have been used to enhance the ionic conductivity values of the SPEs. It has been shown that the ionic conductivity of the aforementioned polymer-based materials can be increased by the addition of a plasticizer into the polymer matrix due to an increase in the amorphous region (6, 10-15). Though the addition of plasticizers in the SPEs has caused a significant decrease in crystallinity and thus improved ionic conductivity, the increase of ionic conductivity by the addition of plasticizers has not been acceptable for use in various electrochemical devices. Among the above-mentioned routes, polymer blending is one of the most significant and useful tools for the development of new advanced polymer-based materials with improved high ionic conductivity values and mechanical stability. The significant advantages of the polymer blend systems are ease of preparation and control of physical properties by compositional variations that are superior as compared to the properties of each component polymer (16, 17). Materials with a wide variety of important and useful properties can be designed by using this technique (18). To become mechanically stable thin membranes, complexation formation by dispersion of metal oxides fillers such as Al_2O_3, ZnO, SiO_2, and TiO_2 to the aforesaid materials have become popular recently, as the obtained materials containing metal oxides attain high ionic conductivity value approaching that of liquid electrolytes along with improved stability (19-21). It is noted that the addition of metal oxide fillers not only improves the mechanical stability of the electrolyte membranes but also causes high ionic conductivity. Thus, the incorporation of inorganic metal oxide fillers into the SPEs would be very useful and significant. Various SPEs based on polymer blend matrixes containing inorganic salts and metal oxides have been proposed, but only very little work has been done and a limited number of literature is available on current research interest (22, 23).

An attempt has been made in the present research work to develop, nanofiber composite polymer blend electrolyte thin films based on polyvinyl alcohol and polyethylene glycol complexed with $LiClO_4$ and dispersed with Al_2O_3 nanofillers, which have been prepared by electrospinning technique and the results are discussed. These polymeric systems have been selected and studied because both the above-mentioned polymers are separately reported to complex formations with salts and metal oxides which are highly amorphous. In addition, the above-mentioned polymers possess properties to form good miscible blends (24). The search for Li^+ ion-containing nanofiber SPE membranes can be attractive because of their low cost, ease of fabrication, and application as thin film membranes. Based on properties like high ionic conductivity, and good thermal and electrochemical stabilities, composite polymer electrolytes containing metal oxides have become popular and attained greater importance over conventional polymer electrolytes. Comparative research studies of the structural, and electrical characteristics of the electrolyte systems have been carried out employing conductivity measurement and XRD, FTIR analysis. Thus in the present research, the effect of $LiClO_4$ salt and metal oxide on the ionic conductivity of the system has been investigated through the above-mentioned techniques. The influence of temperature variation on the ionic conductivity of the polymer electrolyte systems has been discussed.

2. EXPERIMENTAL SECTION

2.1. Preparation

Polyvinyl alcohol (PVA) (Mw = 5×10^5, Sigma Aldrich) and polyethylene glycol (PEG) (Mw = 6×10^3, Sigma Aldrich) were vacuum dried at 50°C for 12 hours before use. An equal amount of PVA and PEG (16 wt% weight percent) by weight have been dissolved in doubly distilled water and then stirred for 12 hours at room temperature until the homogeneous solution of polymers is obtained after the complete dissolution of the polymer. 16 wt% weight percent solution of varying content (wt%) of PVA and PEG have been mixed to obtain a uniformly blended solution. The required quantity (5, 10, 15, 20 wt%) of $LiClO_4$ is further added to the polymeric solution with uninterrupted steady stirring for about 12 hours. The concentrations of Al_2O_3 in the solutions are 0, 5, 10, and 15 wt% based on the weight of PVA. The subsequent homogeneous polymeric solutions are degassed for 20 minutes to obtain the bubble-free transparent solution. The nanofibrous membranes are obtained by the ordinary electrospinning technique at room temperature. The electrospinning system practiced in this research study to prepare nanofiber polymer electrolyte membranes consisted of a syringe and needle (ID = 0.35 mm), a syringe pump, a ground electrode, and a huge voltage supply. A stainless steel needle (24 G, outer diameter: 0.50 mm, inner diameter: 0.35 mm) is associated with an electrode of a high voltage power supply (Spellman, model CZE1000R) which could set up voltages up to 30 kV. Appropriate quantity of polymer blend solutions are electrospun at room temperature adapting a syringe withdrawal pump (New Era Pump Systems Inc., Model NE-300) with a stainless steel needle. The electrospun fibers in the form of untwisted membranes have been accumulated on a grounded, polished stainless steel sheet wrapped with a thin aluminum foil, where the ground collector plate and the spinneret needle tip distance have been fixed at 10 cm. Electrospinning of the homogeneous solutions of different samples is accomplished at a flow rate of 3 mL/hour with a huge voltage of 20 kV at room temperature. The polymer blend membrane thickness has been precisely and thoroughly controlled during electrospinning. The electrospun nanofibrous membranes are then accumulated on the collector plate. Electrospun nanofibrous membranes are collected and desiccated at room temperature for 12 hours to inhibit the contraction of fibers and then finally dried under vacuum at 50 °C for 24 hours before further use. After complete drying, the films were peeled off from the aluminum and kept in vacuum desiccators before use.

3. RESULTS AND DISCUSSION

3.1. Conductivity Analysis

The ionic conductivity of the electrospun nanofiber polymer blend electrolyte systems has been studied by a. c. conductivity analysis from 298 to 353 K. Fig. 1 shows the variation of ionic conductivity as a function of inverse temperature for the electrospun nanofiber composite polymer blend electrolyte systems prepared by electrospinning technique and containing various concentrations of $LiClO_4$ salt and aluminum oxide filler. Similar linear relationships can be seen in all complex polymer electrolyte system curves at all temperatures and this means that there is no phase transition occurs in the polymer blend matrix by the incorporation of lithium salt in the temperature range studied. The variation of ionic conductivity with temperature has been examined to follow Arrhenius-type behavior described by the relation

$$\sigma = \sigma_\circ \exp(-E_a/KT)$$

where $\sigma_\circ$ is the pre-exponential factor while E_a, K, and T are the activation energy, Boltzmann constant, and absolute temperature, respectively.

The ionic conductivity results indicate that the conductivity of all the systems increases with a step up in temperature. The reason may be because, with the rise in the temperature of the samples, the motion of the polymer chain improved, thereby increasing in fraction of free volume in the polymer systems due to the expansion of the polymer. The increase in free volume results in improved translational motion of ions and segmental mobility of polymers. Thus starts easy movement of charge carriers into the available free volume that will cause enhanced conductivity. The polymer segmental motion contributes a pathway for ion transportation. Hence, the ionic mobility in the polymer electrolytes may be due to the mobility of ions between different sites and polymeric segmental motion, which causes improved ionic conductivity of the systems. Therefore the resulting ionic conductivity can be expressed by the around mobility of ion and polymer segmental motion.

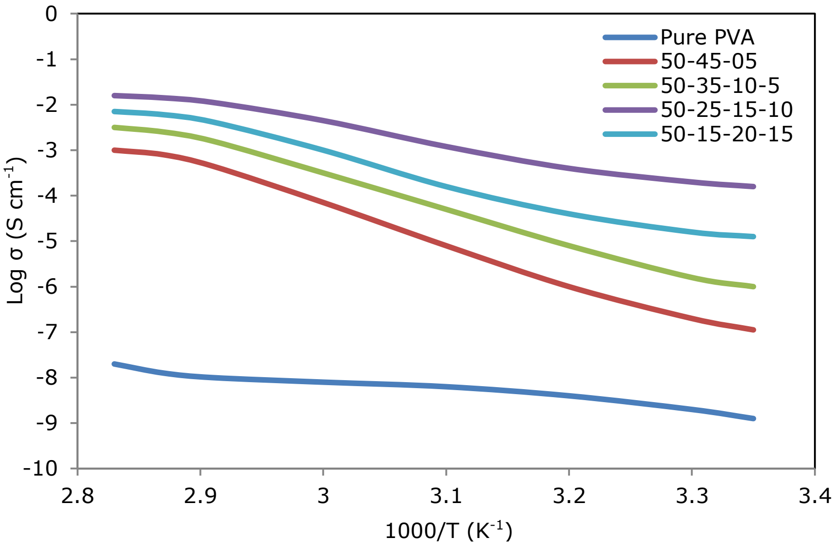

Figure 1: Arrhenius plot of log conductivity versus reciprocal temperature for PVA-PEG-$LiClO_4$-Al_2O_3 Polymer electrolyte complexes.

The activation energy (E_a) values of electrospun nanofiber-based polymer electrolytes systems containing various concentrations of $LiClO_4$ salt and Al_2O_3 filler have been obtained from the slopes of log σ versus 1/T plots at common temperature by using the Arrhenius relation and are presented in Table 1. From the table, it can be noted that E_a values markdown with the increase in the concentration of $LiClO_4$ salt up to 15 wt% beyond which it exhibits an increase, whereas ionic conductivity values increase up to 15 wt% beyond which these values decrease due to high activation energy. The variation in the ionic conductivity and E_a values of polymer electrolyte films can be elucidated on the basis that the nano-fibrous membranes may be a hybrid association of amorphous and crystalline zones.

Table 1: Activation energies (E_a) of pure PVA and PVA-based nanofiber composite polymer electrolyte films.

Polymer electrolyte	Activation energy (eV)
Pure PVA	0.60
PVA-PEG-$LiClO_4$-Al_2O_3 (50-45-05-0)	0.49
PVA-PEG-$LiClO_4$-Al_2O_3 (50-35-10-05)	0.42
PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10)	0.39
PVA-PEG-$LiClO_4$-Al_2O_3 (50-15-20-15)	0.43

Fig. 2 shows the ionic conductivity values of electrospun nanofiber polymer-based electrolyte systems (PVA-PEG-$LiClO_4$-Al_2O_3) as a function of $LiClO_4$ salt concentration 0, 5, 10, and 20 wt% in the temperature range 298-353 K. Ionic conductivity of electrospun nanofiber polymer-based electrolyte systems increases with increasing salt concentration up to 15 wt%, but any further incorporation of the salt beyond 15 wt% causes decrease in ionic conductivity. It can be noted that at lower concentrations of $LiClO_4$ salt, the ionic conductivity steps up initially with an increase in salt concentration while at higher salt concentrations, the conductivity declines. The high value of ionic conductivity at low salt concentrations may be due to the buildup of charge carriers while at higher salt concentrations, the lower value of ionic conductivity may be due to the reason of ion pairs effect and ion aggregations which may influence the overall mobility of ions by the retarding effect and the number of effective charge carriers (25). The maximum ionic conductivity of 1.58 × 10^{-4} S cm^{-1} has been obtained for the nanofiber polymer blend electrolyte system PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10) containing 15 wt% $LiClO_4$ salt at 298 K. The eminent value of ionic conductivity in an electrolyte may be due to improved ionic mobility and high concentration of ionic charge carrier.

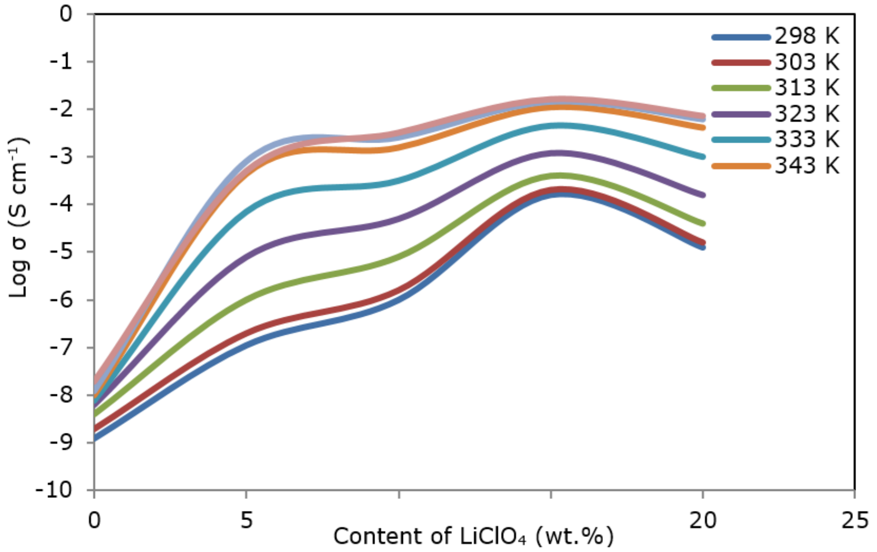

Figure 2: Plot of log conductivity vs. composition of PVA-PEG-$LiClO_4$-Al_2O_3 at various temperatures.

3.2. XRD Analysis

Fig. 3 illustrates the XRD patterns of pristine materials PVA, $LiClO_4$ salt together with that of pure PVA-PEG polymer blend (50-50) weight percent ratio. The figure also displays the XRD pattern of electrospun composite polymer electrolyte systems (PVA-PEG-$LiClO_4$-Al_2O_3) containing various concentrations of $LiClO_4$ salt and nanoparticles of aluminum oxide (Al_2O_3) in the complex polymer matrix. XRD pattern of pure PVA film shows a characteristic peak at 2θ = 19.6° (curve a) which indicates its semicrystalline nature (12, 26, 27). When the two-component polymers are added to form a blend (50-50 ratio), a relatively less intense broad amorphous peak at 2θ = 17.6° and 33.8° (curve b) is observed indicating interaction amongst component polymers of blend leading to the occurrence of the observed peak which different from its any component polymers. Therefore, the appearance of broad amorphous peaks around 17.6° and 33.8° can be attributed to the development of polymer blend. The occurrence of the less intense peak at a lower 2θ value can also indicate the compatibility of PEG with PVA which makes it an acceptable plasticizer for PVA (28, 29). A broad amorphous peak centered at 2θ = 18.5° can be observed in the PVA-PEG film containing 5 wt% $LiClO_4$ salt (curve c). However, no such peak appears in the PVA-PEG-$LiClO_4$-Al_2O_3 films containing a higher concentration of $LiClO_4$ salt (curve d-f) which indicates an increase in the complexation between polymer matrix and salt (30, 31). The absence of a peak corresponding to $LiClO_4$ salt shows the thorough dissolution of salt in the polymer complexes. When $LiClO_4$ is added to the PVA-PEG-Al_2O_3 blend electrolyte to form a complex system, the observed peaks corresponding to the salt appear to decrease, signifying an increase in the amorphous nature of membranes due to complexation between polymer blend matrix and salt (curve d-f). The decrease in the peaks and shift in 2θ values indicates interaction among various components leading to the development of the PVA-$LiClO_4$ complex. A new peak has appeared at 2θ = 45°, 50.4° and 75.1° for polymer electrolyte films (PVA-PEG-$LiClO_4$-Al_2O_3) containing 10, 15, and 20 wt% of $LiClO_4$ salt (curve d-f), which may be due to the interaction of $LiClO_4$ salt and aluminum oxide fillers with the polymer matrix. This indicates the structural changes of the polymer electrolyte systems by the addition of aluminum oxide fillers and $LiClO_4$ salt. It is obvious from Fig. 3 that the amorphicity is retained at lower concentrations of $LiClO_4$ salt while at higher concentrations of salt, the amorphicity of the complex is found to decrease, which may be the basis for the decreased conductivity of the films containing higher than 15 wt% concentration of $LiClO_4$ salt.

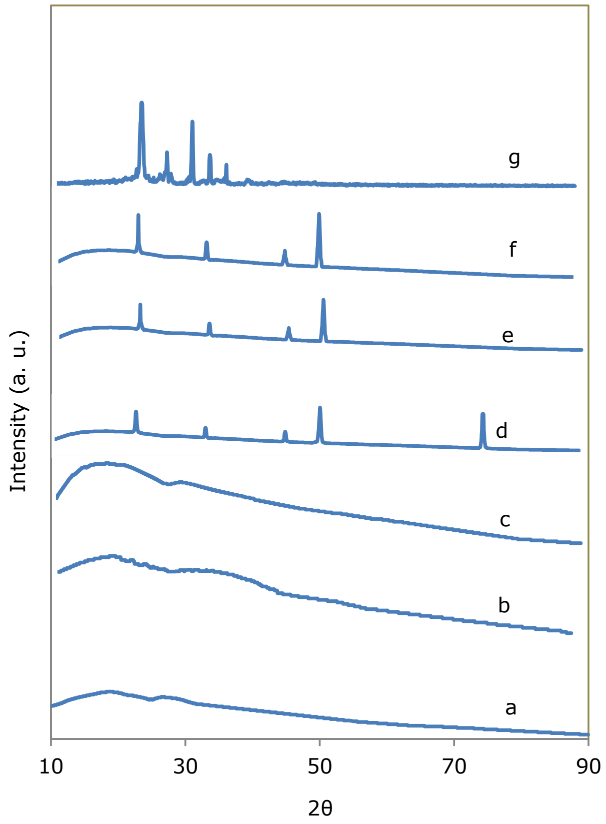

Figure 3: X-ray diffraction spectrum of (a) Pure PVA (b) PVA-PEG (50-50) (c) PVA-PEG-$LiClO_4$-Al_2O_3 (50-45-05-0) (d) PVA-PEG-$LiClO_4$-Al_2O_3 (50-35-10-05) (e) PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10) (f) PVA-PEG-$LiClO_4$-Al_2O_3 (50-15-20-15) (g) pure $LiClO_4$

3.3. FTIR Analysis

The addition of salts and inorganic metal oxide fillers to the host polymer matrix causes interactions and complexation of the host polymer matrix. Such interactions and complexation may affect significantly the sections of polymer backbones and their flexibility. FTIR analysis studies will provide the confirmation of complexation and interactions that occur in polymer matrix after the addition of salt and metal oxide. Fig. 4(a-e) shows the FTIR spectra of pure PVA, PVA-PEG, and PVA-PEG-$LiClO_4$-Al_2O_3 complex polymer electrolyte films containing 5-20 wt% of $LiClO_4$ salt. The most significant structural changes have been noticed after incorporation of $LiClO_4$ salt and aluminum oxide nanoparticles which causes shifts in the vibrational approaches of PVA. These changes have been observed through FTIR spectra recorded in the range 650-4000 cm^{-1}. Figure 4 shows that the enlarged band in the range of 3000-3624 cm^{-1} associated with the Intermolecular hydrogen-bonded O-H stretching vibration frequency of PVA has been shifted to 2942-3517, 2778-3374, 2455-3125, and 2416-3000 cm^{-1} in the PVA-PEG-$LiClO_4$-Al_2O_3 films containing 5, 10, 15 and 20 wt% of $LiClO_4$ salt respectively. This changeover in vibration frequency may be due to the complexation of dopants with the host polymer. The peak found at 2881-2982 cm^{-1} associated with C-H stretching frequency of PVA has been displaced to 2794-2889, 2622-2733, 2317-2924, and 2320-2412 cm^{-1} in PVA-PEG-$LiClO_4$-Al_2O_3 electrolyte systems containing 5, 10, 15 and 20 wt% of $LiClO_4$ salt, respectively. Furthermore, the peak occurred at 1758-1818 cm^{-1} associated with carbonyl stretching vibration frequency of pure PVA film and shifted to 1573-1687,1461-1532, 1088-1117 and 1188-1235 cm^{-1} for PVA-PEG-$LiClO_4$-Al_2O_3 polymer electrolyte films containing 5, 10, 15 and 20 wt% of $LiClO_4$ salt, respectively. Similarly, frequency corresponding to C-H bending mode is found at 1568 cm^{-1} in pure PVA film and displaced to 1449, 1235, 914, and 962 cm^{-1} for PVA-PEG-$LiClO_4$-Al_2O_3 polymer electrolyte films containing 5, 10, 15 and 20 wt% of $LiClO_4$ salt, respectively. The C-O stretching and O-H bending vibrations frequency are found to be observed at 1180-1310 cm^{-1} for pure PVA film and has been displaced to 1080-1176, 930-998, 701, and 669 cm^{-1} for PVA-PEG-$LiClO_4$-Al_2O_3 films containing 5, 10, 15 and 20 wt% of $LiClO_4$ salt, respectively. It can be confirmed that there is no characteristic frequency peak corresponding to $LiClO_4$ salt found in the FTIR spectra of the polymer electrolyte systems. However, it has been noticed that vibration frequency bands either displaced or disappeared with the addition of $LiClO_4$ salt in the pure PVA film. The shifting of frequency peaks and the appearance of new peaks in the polymer electrolyte systems indicates the occurrence of interaction and complexation of $LiClO_4$ salt with the host polymer matrix in the PVA-based blend polymer electrolyte system. The characteristic vibration frequency bands of aluminum oxide nanoparticles are not observed in the spectra which may be due to the reason that the size of the particles is very small and therefore only the local structural changes may occur (32).

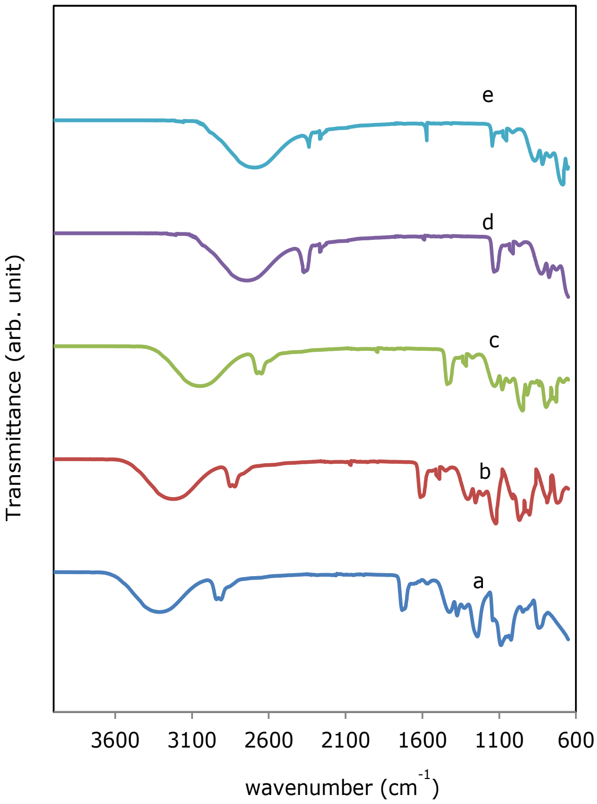

Figure 4: FTIR spectrum of pure PVA and PVA-PEG-$LiClO_4$-Al_2O_3 complex solid polymer electrolytes (a) Pure PVA (b) PVA-PEG-$LiClO_4$-Al_2O_3 (50-45-05-0) (c) PVA-PEG-$LiClO_4$-Al_2O_3 (50-35-10-05) (d) PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10)(e) PVA-PEG-$LiClO_4$-Al_2O_3 (50-15-20-15)

3.4. Scanning Electron Microscopy

The morphology of the electrospun nanofibers-based PVA-PEG blend polymer electrolyte membranes containing $LiClO_4$ salt and inorganic metal oxide (Al_2O_3) prepared with various compositional ratios wt% by weight percent have been examined by using scanning electron microscopy (SEM) and are presented in Fig. 5. SEM images reveal the influence of composition on morphology of the fibers of the membranes. It can be observed that the membranes on aluminum foil are composed of well interconnected network of interlaid, random, and ultrafine straight fibers which arises due to the operating processing parameters. SEM images also show that the electrospun membranes consist of multi-fibrous layers and fully interconnected porous fibrous structures. The exceptionally porous structure of the electrospun membranes may be due to the interlaying of the fibers. SEM micrographs indicate that the fiber morphology of the electrospun PVA-PEG blend polymer electrolyte membranes mainly depends on the number of component ratios (33). When the amount of $LiClO_4$ salt is increased, the average spacing between the fibers is observed to decrease. It can also be noted that the cross-linking of the fibers depends on the content of lithium perchlorate. In the PVA-$LiClO_4$ sample, the highest cross-linking of the fibers has been achieved in the presence of a high concentration of salt in the polymer matrix. No phase separation can be observed in electrospun fiber polymer electrolyte membranes which indicates compatibility of PVA and PEG blend polymer. It can be observed that the surface of the fibrous membranes is partially amorphous which may be due to the amount of water that remains on the surface of membranes after electrospinning which possibly dissolves the fibers. The residual water molecules may also cause the development of a considerable number of cross-links resulting in smaller pore size (34). SEM micrographs also reveal that the diameters of the fibers are not homogeneous and uniformly distributed throughout. It can be observed that all the polymer blend electrolyte membranes show long and straight fibrous morphology.

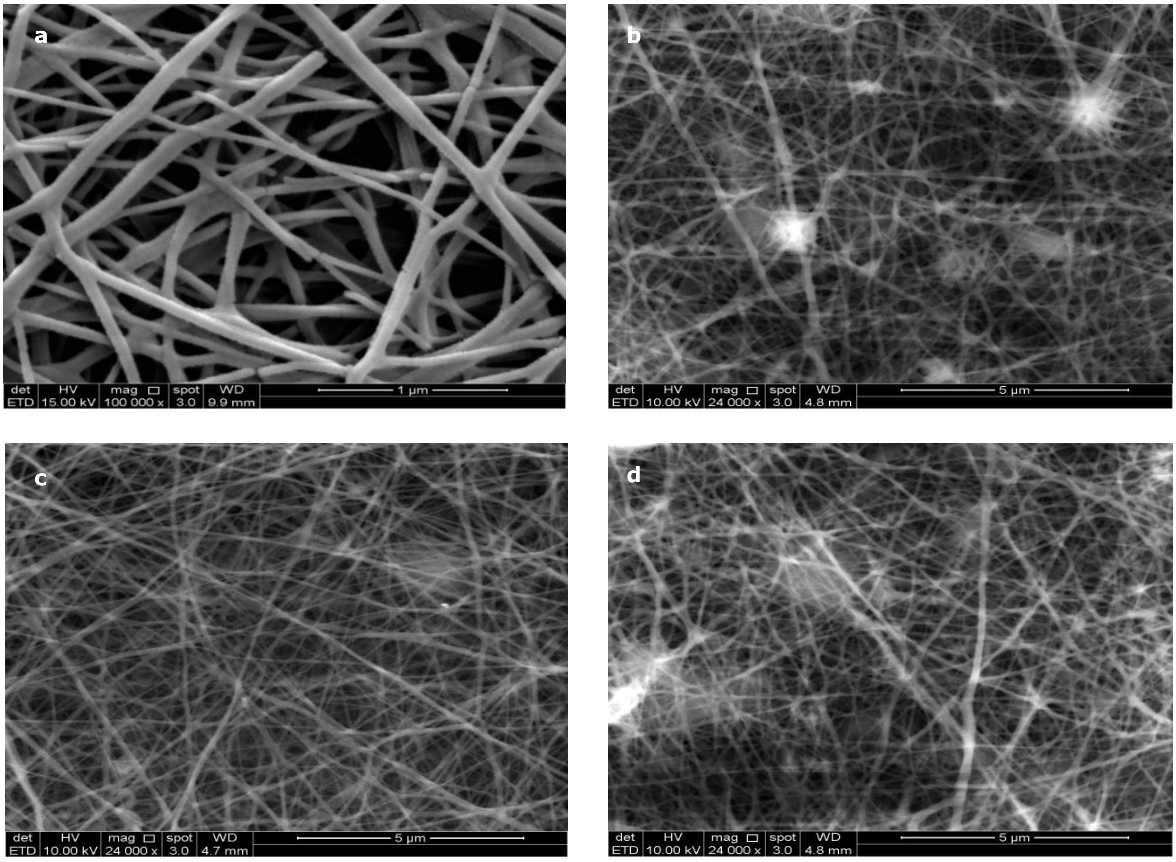

Figure 5: SEM images of (a) Pure PVA (b) PVA-PEG-$LiClO_4$-Al_2O_3 (50-35-10-05) (c) PVA-PEG-$LiClO_4$-Al_2O_3 (50-25-15-10) (d) PVA-PEG-$LiClO_4$-Al_2O_3 (50-15-20-15)

4. CONCLUSIONS

The new type of fibrous electrospun polymer blend (PVA-PEG) electrolyte systems are prepared successfully by the electrospinning technique and reported in the present research work. The highest ionic conductivity has been obtained to be $1{\cdot}58 \times 10^{-4}$ S cm^{-1} at 298 K when the polymer blend is complexed with 15 wt% of lithium perchlorate. The temperature-dependent ionic conductivity of polymer-based electrolyte systems shows the Arrhenius behavior with two distinct regions. The dependence of the ionic conductivity on temperature is found to be linear on a logarithmic scale. It follows the Arrhenius behavior of ionic conductivity. The XRD and FTIR research studies indicate the complex formation of salt with the polymer with a decrease in crystallinity upon the addition of salt and the results also indicate that the polymers are completely miscible with each other. The results based on high ionic conductivity and low crystallinity show that electrospun PVA-PEG blend polymer electrolyte membranes (PVA-PEG-$LiClO_4$-Al_2O_3) are a possible candidate for solid-state batteries and can be used in the forthcoming imminent lithium-ion batteries.

5. CONFLICT OF INTEREST

All authors have shared in (a) planning, designing, and explanation of the data; (b) drafting of the manuscript; and (c) recommendation of the final interpretation. This article has not been submitted to, nor is it under consideration at any other journal or publishing venue.

6. ACKNOWLEDGEMENT

The authors gratefully acknowledge the financial support and assistance to complete this work by the University Sains Malaysia (USM), Malaysia. The authors are also grateful to The World Academy of Sciences (TWAS), Italy for the financial support. The authors are thankful to the School of Electrical Engineering, USM, Malaysia for providing a conductivity measurement facility.

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