Malaysian Journal of Chemistry, 2023, Vol. 25(1), 108-115

Tensile and Ionic Conductivity Behaviour of Different Molecular Weight Poly(Ethylene Oxide (PEO) / Polyurethane (PU) Blend Electrolyte System

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The tensile stability of materials used for battery fabrication must be given due consideration despite ionic conductivity being a crucial criterion. In this study, the tensile stability and ionic conductivity analysis of solid polymer electrolytes (SPE) of different molecular weights, i.e. poly(ethylene oxide) (PEO) (M_w = 100 and 4,000 kg mol^{-1}) in a polyurethane (PU) blend system, was conducted at ambient temperatures. The semi-crystalline PEO was blended with amorphous PU to reduce the crystallinity of the film, and the conductivity of the system was further enhanced with the addition of lithium perchlorate ($LiClO_4$). The PEO/PU/$LiClO_4$ films at different polymer (PEO_{100} and PEO_{4000}) and salt concentrations were prepared by the solvent casting method. Tensile tests, Fourier Transform Infrared Spectroscopy (FTIR) and Electrochemical Impedance Spectroscopy (EIS) were used to study the tensile and conductivity behaviours of the system. The optimum elongation at break for the PEO_{100}/PU and PEO_{4000}/PU films were 11.25% and 17.71%, obtained at ratios of 50:50 and 80:20, respectively. The elasticity of the PEO_{100}/PU and PEO_{4000}/PU films increased from 54.697 to 20.948 MPa and from 67.393 to 15.980 MPa, respectively. These results showed that the elasticity of the system was enhanced when PEO and PU were blended together. The optimum conductivity values for PEO_{100} and PEO_{4000} were recorded at 3.61 x 10^{-7} S cm^{-1} and 1.16 x 10^{-7} S cm^{-1}, respectively, when 10 wt% and 5 wt% of $LiClO_4$ was added. It can be concluded that the PEO/PU polymer blend electrolyte systems prepared in this study could be efficient polymer electrolytes, especially for battery applications.

Keywords: Polymer electrolyte; poly(ethylene oxide); polyurethane; tensile stability; ionic conductivity

Received: Ocotber 2022; Accepted: January 2023

In this era, the availability of sustainable, safe, and environmentally friendly energy resources is critical. Electrochemical energy storage has gained much attention over the last decade due to the depletion of fossil fuels, global warming, and high energy demand [1]. Special consideration has been given to the development of optimum energy storage materials and much research has been carried out to develop effective and ideal energy storage devices for secondary battery systems.

Since the first lithium-ion batteries were commercialized in 1991, they have been used in many applications [2-5]. The revolution of fabricating solid-state electrochemical energy storage appliances, which include supercapacitors, rechargeable batteries, and photo electrochemical cells, has been greatly influenced by the discovery of the ionic conductivity (σ) behaviour of polymer-salt complexes [6,7]. Thus, solid polymer electrolytes (SPEs) have been developed in order to overcome the drawbacks of previous secondary battery systems.

Materials based on poly(ethylene oxide) (PEO) are frequently regarded as viable possibilities for polymer hosts in solid state electrolytes for high energy density secondary batteries [8]. PEO has been commonly used due to its ability to solvate a broad range of inorganic and organic salts. PEO can solvate different types of salts due to the lone pair of electrons on its oxygen atom [9].

Despite its favourable properties, the semi-crystalline structure of PEO constrains its use for SPE applications. Between the two regions in a semi-crystalline polymer (the amorphous and crystalline regions), ions prefer to dissolve in the amorphous region [10,11], which is where ion mobility occurs. The crystalline regions in PEO obstruct salt ion mobility, thus generating low conductivity in PEO-based energy storage systems [12,13]. The relatively low conductivity of PEO (below $\sigma \approx 10^{-8}$ S cm^{-1}) at ambient temperatures limits the application of PEO as SPE in lithium batteries [14]. However, the addition of lithium inorganic salts such as $LiClO_4$, which dissociates in solution into ions, into the SPE system will help improve its conductivity.

Polyurethane (PU), which is more amorphous in nature, can be blended together with PEO to suppress its crystallinity. Thus, more amorphous regions will form and the ion mobility in the system will be enhanced [12]. As this procedure significantly improves electrochemical characteristics, the blending process of PEO and PU is known to promote the mechanical stability and ionic conductivity of SPE. The overall performance of the PEO/PU blend system is improved by the use of a second polymer (polymer blend) which balances the other out [12]. By increasing the amorphous phase of the polymer electrolyte and decreasing the crystallinity of the electrolyte system, polymer blends have demonstrated improved physical properties. [15].

Therefore, in this study, we focussed on achieving optimum conditions for both tensile stability and ionic conductivity. The tensile strength and elasticity of the PEO_x/PU blend systems were studied and discussed. The optimum ionic conductivity of the $PEO_x/PU/LiClO_4$ films at different polymer (PEO_{100} and PEO_{4000}) and salt concentrations at room temperature were also investigated. We hope that this study might give a better understanding of how improving tensile strength can assist in the fabrication of SPEs with optimum ionic conductivity values, specifically in battery applications.

EXPERIMENTAL

Chemicals and Materials

Polyethylene oxide (PEO) of different molecular weights (M_w: 100 and 4,000 kg mol^{-1}) with purity ≥ 99 % were purchased from Sigma-Aldrich Chemical Company and used as such. Polyurethane (PU) with purity ≥ 99 % was purchased from Sigma-Aldrich and used after purification. Anhydrous $LiClO_4$ with purity > 99 % was obtained from Acros Organic and dried in a vacuum oven at 100 °C for 48 h to eliminate any traces of water before dissolution. Tetrahydrofuran (THF) with purity ≥ 99 % was purchased from Sigma-Aldrich and used as a solvent to dissolve $LiClO_4$, PEO and PU.

Preparation and Characterization Methods

In this study, PEO with different molecular weights were used. PEO_{100} and PEO_{4000} represent PEO_x with M_w = 100 and 4,000 kg mol^{-1}, respectively. The PEO_x/PU polymer blend was prepared first with different composition ratios from 100/0 to 50/50 at 10 % intervals via a solvent casting technique. A 2 (w/w) % ratio of PEO_x/PU to solvent (THF) was applied. PEO and PU were dissolved in THF using a magnetic stirrer for 24 to 48 h at 50 °C to ensure complete dissolution. The mixture was then poured into a Teflon petri dish and left to evaporate at ambient conditions for approximately 2 days, and subsequently dried in an oven at 40 °C for 1 day to form a free-standing film [28].

The solvent casting method was implemented to prepare the $PEO/PU/LiClO_4$ polymer blend electrolyte system. A PEO/PU ratio that gave the optimum tensile stability was used in the preparation of the $PEO/PU/LiClO_4$ electrolyte system. The amount of $LiClO_4$ added was varied from 0-15 %. The dried film was then characterized in terms of its chemical interactions, structural properties and conductivity.

The tensile strength and elasticity of this series of polymer blends was tested using the Instron universal mechanical testing machine at ambient temperatures in accordance to ASTM D882. The PEO_{100} film was tested with a load cell of 10 N with a cross-head speed of 10 mm/min, while the PEO_{4000} film was tested with a load cell of 10 N and a 50 mm/min cross-head speed. Since the PEO_{4000} film was more elastic, a higher cross-head speed was used to minimized the time taken for the film to break.

Fourier transform infrared spectroscopy (FTIR) analysis was run using a Thermo Nicolet 6700 FTIR Spectrometer between 4000 cm^{-1} and 440 cm^{-1} with a scanning resolution of 4 cm^{-1} to study the chemical bonds and detect the functional groups in the polymer electrolyte system. The ionic conductivity of the polymer electrolyte membranes was measured by EIS (Electrochemical Impedance Spectroscopy) at room temperature using a Hioki 3532-50 LCR HiTester at a frequency range of 50 Hz to 1 MHz at 1.0 V. Figure 1 summarizes the methodology used in this study.

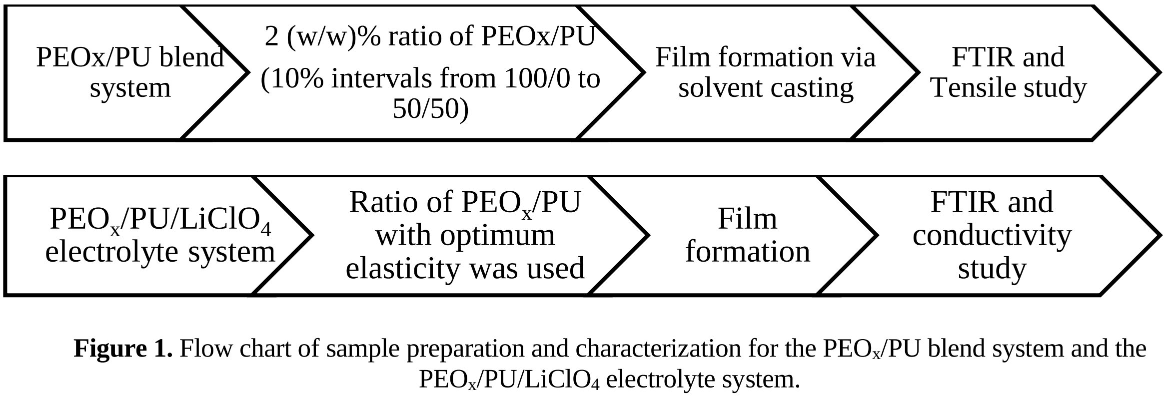

Figure 1. Flow chart of sample preparation and characterization for the PEO_x/PU blend system and the $PEO_x/PU/LiClO_4$ electrolyte system.

Table 1. Mechanical properties of PEO_{100}/PU and PEO_{4000}/PU polymer blends at different polymer ratios.

PEO/PU (ratio)	PEO_{100}/PU				PEO_{4000}/PU			
	Stress (MPa)	Strain	Young's Modulus	Elongation at break (%)	Stress (MPa)	Strain	Young's Modulus	Elongation at break (%)
100:0	0.432	0.008	54.697	0.79	5.055	0.075	67.393	7.50
90:10	0.970	0.022	43.784	2.22	3.169	0.140	22.595	14.03
80:20	0.662	0.018	37.979	1.75	4.690	0.294	15.980	17.71
70:30	0.961	0.022	43.753	2.21	4.196	0.117	35.966	11.67
60:40	2.086	0.095	22.197	9.50	3.121	0.125	24.977	12.50
50:50	2.336	0.125	20.948	11.25	3.229	0.177	18.230	14.73

RESULTS AND DISCUSSION

Mechanical Strength

The mechanical strength of the PEO_x/PU films produced in this study was tested via tensile testing. For the PEO_{100} film, the addition of PU improved its tensile stability. The optimum tensile stability was achieved when the ratio of PEO/PU was 50:50, while for PEO_{4000}, optimum tensile stability was achieved at a ratio of 80:20. Table 1 shows the values for stress, strain, Young's modulus and elongation at break of the polymer blend film.

From Table 1, the tensile properties of the films were notably improved after blending PEO and PU together. The PEO_{100}/PU and PEO_{4000}/PU blend films elongated at the optimum values of 11.25 % and 17.71 %, respectively. This proved that the blending process of PU with PEO decreased the crystallinity of the film and enhanced its elasticity. The notably high percentage of deformation before rupture also correlates with the significant strain value of the films. Figure 2 represents the elasticity properties of the PEO/PU blend. The PEO_{100}/PU exhibited its optimum elasticity at a ratio of 50:50 with 20.948 MPa. While for PEO_{4000}/PU, the optimum elasticity was 15.98 MPa, at a ratio of 80:20. It is noted that a smaller amount of PU was needed to enhance the tensile stability of PEO_{4000} compared to PEO_{100}. This shows that differences in molecular weight had an effect on the mechanical properties of PEO.

The rise in tensile strength was correlated to the decline in Young's modulus values. As the strength of the material improved, the Young's modulus value declined, as tensile strength is a reciprocal of Young's modulus, based on the stress-strain formula [16]. The low modulus value signifies that the film produced has higher elastic characteristics and vice versa. The enhancement of the film's mechanical behaviour suggests a reduction in the crystalline phase and the high number of chain entanglements in the PEO/PU SPE system [16]. However, the crystalline phase is still important for an electrolyte system as it maintains the shape of the film itself despite the increase in the amorphous phase, which enhances ion mobility.

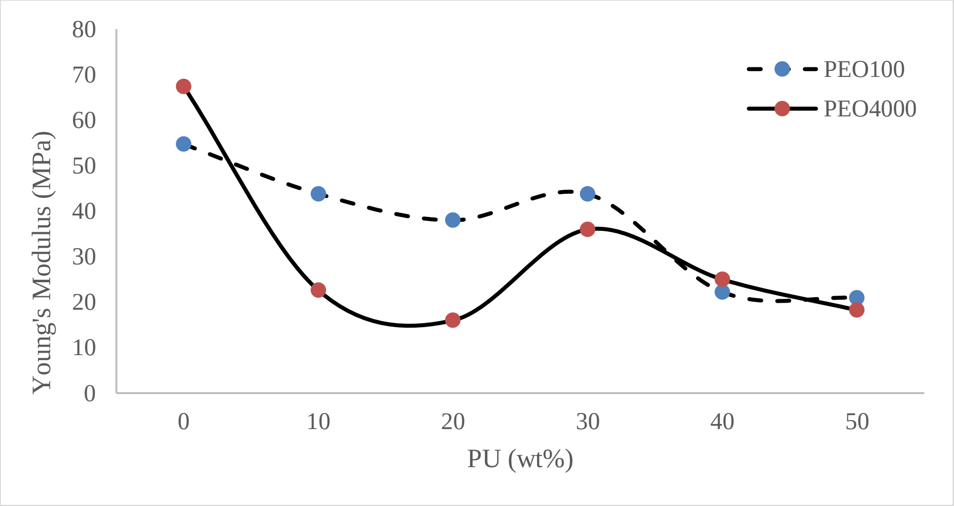

Figure 2. Young's modulus values for the PEO_{100}/PU and PEO_{4000}/PU polymer blends

Ionic Conductivity

The conductivity values at room and elevated temperatures are derived from impedance measurements via EIS. Ion mobility was studied to determine the optimum ionic conductivity (σ) for the PEO/PU/ $LiClO_4$ blend system and the optimum ratio for tensile strength at room temperature. The impedance data obtained from the EIS measurements have been described as the Nyquist or Cole-Cole plot [17]. In this plot, the imaginary impedance, Zi (capacitive) is plotted against the real impedance, Zr (resistive) at varying frequencies [18]. From the Cole-Cole plot, the ionic conductivity values of the PEO/PU/$LiClO_4$ electrolyte are calculated using equation (1),

$$\sigma = \frac{t}{R_b A} \tag{1}$$

where t is the thickness of the sample, A is the area of the blocking electrode and R_b is the bulk resistance of the SPE.

The addition of $LiClO_4$ salt to the PEO/PU polymer blend significantly improved the ionic conductivity of both PEO_x systems. This was mainly due to a strong interaction between the solvating group and the lithium salt cation as well as the polymer host segmental mobility [19]. The ionic conductivity values for PEO_{100}/PU/$LiClO_4$ and PEO_{4000}/PU/$LiClO_4$ with different $LiClO_4$ concentrations are shown in Figure 3. For the PEO_{100} polymer blend, the highest σ achieved was 3.61 x 10^{-7} S cm^{-1} with 10 wt% $LiClO_4$ salt.

The increase in the σ values was due to an increase in the total number of free ions as a result of the dissociation of the Li^+ ions from the $LiClO_4$ salt [20]. However, addition of $LiClO_4$ salt above 15 wt% resulted in the formation of a non-free-standing film (gel state). The optimum conductivity value of the PEO_{4000} polymer blend (1.16 x 10^{-5} S cm^{-1}) was achieved when 5 wt% of $LiClO_4$ salt was added. Further addition of $LiClO_4$ salt to 10 wt% caused the ionic conductivity to decrease and then increase again when 15 wt% salt was added.

This result could be attributed to charge transport in PEO, which involves the separation of the Li^+ cation from its coordinating oxygen to a contiguous site. In addition, the addition of PU to PEO decreased the crystallinity of PEO and stimulated lithium-ion movement in the polymer electrolyte. The high σ in an electrolyte is credited to the increased concentration of the ionic charge carrier. The movement of ions in SPE is a fluid mechanism, by which the development of ions through a polymer lattice is related to the substantial sufficiency of the segmental movement [21,22].

Fourier Transform Infrared Spectroscopy

The FTIR spectra of the PEO_{100}, PEO_{4000}, PU, PEO/PU blend and PEO/PU/$LiClO_4$ samples with optimum conductivity are shown in Figure 4. From the IR spectra, the presence of triple C-O-C stretching peaks proves the semi-crystalline phase of PEO [12]. The PEO/PU blends were proven to be miscible and able to form a homogeneous polymer blend film, based on the IR spectra obtained. The miscibility might be due to the formation of hydrogen bonding between both polymers [23,24]. The FTIR spectra showed that the peak at ~ 3291 cm^{-1} reduced in intensity after PEO/PU blending [6]. This behaviour is mainly due to the relationship between hydrogen interactions and the mechanical strength of the PEO/PU film [25].

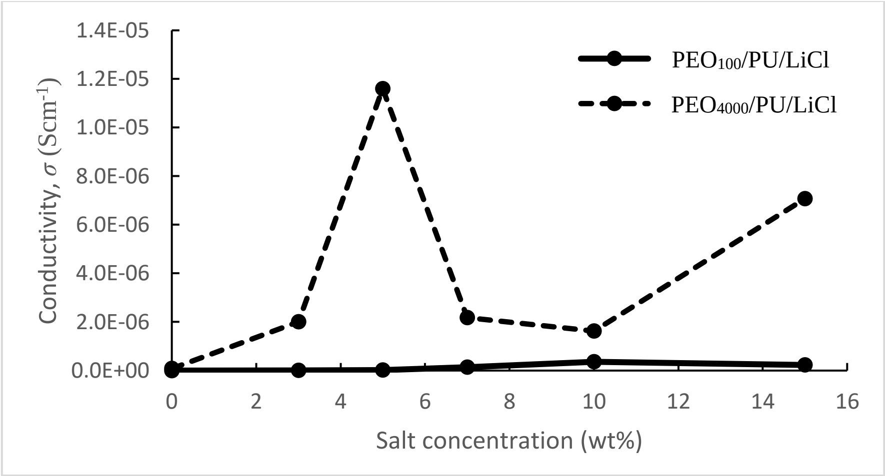

Figure 3. Ionic conductivity values of PEO_{100}/PU/$LiClO_4$ and PEO_{4000}/PU/$LiClO_4$ with different $LiClO_4$ concentrations.

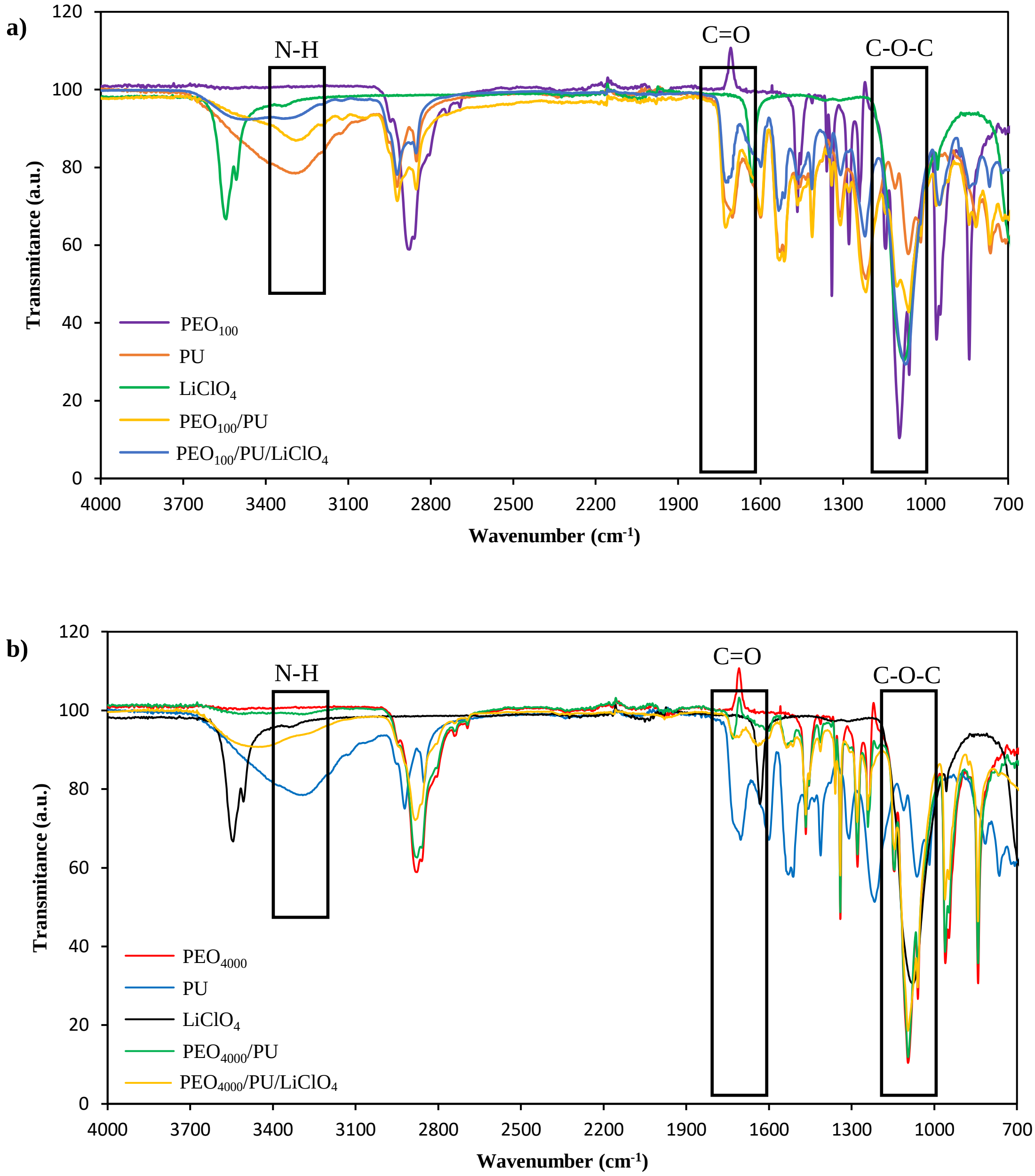

Figure 4. FTIR spectra of a) PEO_{100}, PU, $LiClO_4$, PEO_{100}/PU, PEO_{100}/PU/$LiClO_4$, and b) PEO_{4000}, PU, $LiClO_4$, PEO_{4000}/PU, PEO_{4000}/PU/$LiClO_4$

The formation of strong hydrogen bonds between PEO and the N–H groups of PU due to different electronegativities of the elements lowered the crystallinity intensity of the SPE, thus improving the mechanical performance of the PEO/PU blend system. The carbonyl group (C=O) of PEO_x/PU gave rise to sharp and intense peaks at 1727 and 1732 cm^{-1}. With the addition of $LiClO_4$, the C=O peak shifted to a lower wavenumber and its intensity also decreased. The C=O peak shifted from 1727 cm^{-1} [26] to 1710 cm^{-1}. Thus, this might indicate a complexation formation between the $LiClO_4$ salt and the PEO/PU polymer host due to the Li^+ ions' coordination capability [27]. However, the result was not significant enough to prove the interaction with the carbonyl group. The FTIR peaks and their corresponding functional groups are summarized in Table 2.

Table 2. Comparison of PEO, PU, PEO_{100}/PU, PEO_{4000}/PU and PEO_x/PU/$LiClO_4$ functional group

Functional Group	**Wavenumbers (cm^{-1})**				
	PEO	**PU**	**PEO_{100}/PU**	**PEO_{4000}/PU**	**PEO_x/PU/$LiClO_4$**
C-H Stretching	2878	2922, 2851	2922, 2852	2877	2923,2853
CH_2 Scissoring	1466	-	1466	1466	1464
CH_2 Wagging	1360	-	1341	1360	1348
CH_2 Twisting	1278	-	1279	1279	1281
C=O	-	1702	1727	1732	1710
N-H	-	3292, 1529	3291, 1532	1538	3290, 1533
C-O-C	1145, 1092, 1059	1062	1145, 1092, 1062	1145, 1095, 1060	1065

CONCLUSION

The use of a PEO/PU polymer blend enhanced the tensile stability of the electrolyte system. Analysis of PEO_{100}/PU and PEO_{4000}/PU showed that the optimum tensile stability was achieved at ratios of 50:50 and 80:20, respectively. For the PEO_{100}/PU blend system, the optimum tensile properties were observed where the stress, strain, Young's modulus and elongation at break were 2.336 MPa, 0.125, 20.948 MPa and 11.25 %, respectively. With the PEO_{4000}/PU blend system, the optimum results for stress, strain, Young's modulus and elongation at break were 4.69 MPa, 0.294, 15.98 MPa and 17.71 %, respectively. The FTIR results confirmed the interactions between PEO, PU and $LiClO_4$ where the N–H groups of PU lowered the crystallinity of the SPE, while the addition of $LiClO_4$ shifted the C=O peak to a lower wavenumber at 1710 cm^{-1}, and decreased its intensity. The presence of PU, which is more amorphous, reduced the crystallinity of PEO, thus increasing lithium ion mobility with the help of the segmental motion of the polymers in the system. However, the conductivity results for PEO_{100}/PU/$LiClO_4$ (3.61×10^{-7} S cm^{-1}) and PEO_{4000}/PU/$LiClO_4$ (1.16×10^{-5} S cm^{-1}) were better when 10 wt% and 5 wt% of $LiClO_4$ salt were added, respectively.

ACKNOWLEDGEMENTS

The authors are grateful to the Ministry of Higher Education Malaysia for the FRGS grant no. 600-IRMI/FRGS-RACER 5/3 (098/2019). We also would like to thank Universiti Teknologi MARA (UiTM) for their support ad for providing facilities for this research.

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