

Influence of 1-Ethyl-3-methylimidazolium Bis(Fluorosulfonyl) Imide on the Conductivity, Thermal Properties and Molecular Structure of Poly (Vinylidene Fluoride-hexafluoropropylene) (PVDF-HFP) Polymer Electrolyte

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Ionic liquids are used in sodium batteries instead of organic solvents because the high flammability of organic solvents can cause serious safety issues, such as fires or explosions. In this study, ionic liquid polymer electrolyte (ILPE) films were prepared using a casting technique by adding 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide [C₂C₁im][FSI] ionic liquid in different ratios (10 wt.%, 20 wt.%, 30 wt.%, 40 wt.% and 50 wt.%) into the poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) solution as a host polymer. The shift in wavenumbers of the vibrational modes of S-N-S and SO₂ indicated the interactions between PVDF-HFP and [C₂C₁im][FSI] ionic liquid. The intensity of the crystalline X-ray diffraction (XRD) peaks decreased consistently with the addition of higher amounts of [C₂C₁im][FSI], suggesting that the presence of ionic liquid reduced the crystallinity of PVDF-HFP. The conductivity of ILPEs generally increased with increasing amounts of ionic liquid, and the highest conductivity at room temperature of $8.04 \times 10^{-5} \text{ S cm}^{-1}$ was recorded for the sample loaded with 50 wt.% [C₂C₁im][FSI]. Thermal gravimetric analysis (TGA) indicated that all ILPE samples could withstand temperatures of up to 300 °C, as no significant weight loss was observed. Differential scanning calorimetry (DSC) analysis indicated that the melting temperature (T_m) decreased with increasing amounts of ionic liquid. These findings imply that the ILPE containing 50 wt.% [C₂C₁im][FSI] may be taken into consideration for battery applications.

Keywords: 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide; ionic liquid polymer electrolytes; poly(vinylidene fluoride-co-hexafluoropropylene); ionic conductivity

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In 1800, Alessandro Volta discovered liquid electrolytes when he dipped two different electrodes into an electrolyte solution of brine (saltwater) and obtained a steady electric current, thus creating the first electrochemical cell [1]. Since then, a variety of liquid electrolytes have been developed and made available for use in electrochemical devices such as fuel cells and solar cells [2], as well as in rechargeable battery applications [3]. Liquid electrolytes often achieve high ionic conductivity values due to their high ion mobility, primarily in less viscous electrolytes. However, the usage of liquid electrolytes in a wide variety of electrochemical devices still faces several safety issues, as they are highly volatile and flammable, and risk leakage [4]. Thus, the solid polymer electrolyte (SPE) which comprises a polymer and a salt, was introduced as a safer alternative. SPEs offer distinct advantages over liquid electrolytes, such as a reduced risk of leakage as well as being less toxic, corrosive and flammable [2].

SPEs are electrolytes that offer great advantages in the field of high-energy electric propulsion batteries and fuel cells of electric vehicles. SPEs are primarily used in electrochemical devices such as batteries and supercapacitors [5]. SPEs may be used in energy devices to overcome the constant demand for high energy density [6]. However, most SPEs still possess poor ionic conductivity which makes them unsuitable for practical applications [7]. Gel polymer electrolytes (GPEs) have the advantage over SPEs as they have a much higher ionic conductivity due to the flexibility of the polymer backbone, which leads to better ion mobility as well as good electrode-electrolyte contact [8]. GPEs can be produced by adding an appropriate ratio of liquid electrolytes to a polymer matrix such as poly(ethylene oxide) (PEO) [9], poly(methyl methacrylate) PMMA [10], polyacrylonitrile (PAN) [11], poly(vinylidene fluoride) (PVDF) [12], or poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) [13]. PVDF-HFP is widely used due to its dielectric constant of $\epsilon = 8.4$ which allows good

dissolution of salts, and as a result, a high concentration of charge carriers. Besides having good hydrophobicity, PVDF-HFP also possesses both amorphous and crystalline phases; the amorphous phase gives better ionic conductivity whereas the crystalline phase provides mechanical support for the polymer electrolyte [1].

Although PVDF-HFP is considered a good polymer electrolyte, it still has certain limitations such as an average ionic conductivity value due to its high crystallinity [1]. As a result, some improvement is required. The introduction of an ionic liquid could improve ionic conductivity as well as chemical and thermal stability while overcoming volatility and flammability issues. Apart from acting as an ion source, an ionic liquid may also serve as a plasticizer. The plasticising nature of ionic liquids softens the polymer chain, allowing for better ion transport and thereby increasing ionic conductivity [14].

In this study, the PVDF-HFP film was doped with 1-ethyl-3-methylimidazolium bis(fluorosulfonyl) imide $[C_2C_1im][FSI]$ ionic liquid and the properties of the resulting ILPE films such as conductivity, thermal behaviour and molecular structure were studied. $[C_2C_1im][FSI]$ ionic liquid was used as $[FSI]$ anion-based ionic liquids have low viscosity and high ionic conductivity. Their great thermal stability and low flammability make them preferable to other types of ionic liquids. The combination of this host polymer and the ionic liquid was chosen as there have not been any studies on the combination of these two systems.

EXPERIMENTAL

Materials and Chemicals

Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) (purity >99.9 %) in pellet form was purchased from Sigma-Aldrich (St.Louis, MO, USA), and 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide

$[C_2C_1im][FSI]$ (purity >97.0 %) ($M_w=291.30$ g/mol) was purchased from Kanto Chemical Co., Inc. (Tokyo, Japan). Acetone solvent (purity >99.9 %) ($M_w=58.08$ g/mol) was obtained from Chemiz (M) Sdn. Bhd. (Selangor, Malaysia). Unless otherwise specified, reagents and chemicals were used as received without further purification. All the chemicals used in this experiment were of analytical grade.

Preparation of Ionic Liquid Polymer Electrolyte

The polymer PVDF-HFP films containing different weight percents (wt%) of ionic liquid were prepared by a casting technique. Initially, the PVDF-HFP pellets were dissolved in 30 mL of acetone. The solution was stirred until a clear homogenous solution was formed. Then, $[C_2C_1im][FSI]$ was added and the mixture was stirred again for 24 hours at room temperature to form a transparent and homogenous solution. The solution was cast onto a Teflon dish and left in the open for 48 hours. The dried polymer film was then peeled off and placed in an oven to remove moisture. Then, the film was stored in a desiccator. This process was repeated with different ratios of ionic liquid, as shown in Table 1.

Characterisation Methods

The functional groups in the films were investigated by attenuated total reflection FTIR analysis (FT-IR Spectrum One, Perkin Elmer). The spectra were recorded in transmission mode in the region of 4000 to 650 cm^{-1} , with a resolution of 4 cm^{-1} and averaging 15 scans.

X-ray diffraction (XRD) provided the chemical composition, physical properties and crystallographic structure of the ionic liquid polymer electrolyte. XRD was performed at room temperature using a Rigaku D = max-3B, X-ray diffractometer (40 kV, 20 mA) (Japan) with $Cu K\alpha$ ($\lambda= 1.540 \text{ \AA}$) irradiation at a scan rate of 2° min^{-1} in the range of 1.5-20°.

Table 1. Composition of PVDF-HFP- $[C_2C_1im][FSI]$ ILPE samples.

Polymer Film	Ratio (PVDF-HFP: $[C_2C_1im][FSI]$)	Weight (g)	
		PVDF-HFP	$[C_2C_1im][FSI]$
Pure PVDF-HFP	100:0	2.0	0.0
ILPE-1	90:10	1.8	0.2
ILPE-2	80:20	1.6	0.4
ILPE-3	70:30	1.4	0.6
ILPE-4	60:40	1.2	0.8
ILPE-5	50:50	1.0	1.0
ILPE-6	40:60	0.8	1.2

The ionic conductivity (σ) was measured by an impedance analyser (3520-80, Hioki E.E. Corp). The ILPE was placed between a pair of stainless-steel blocking electrodes. The thickness of the ILPE film was recorded using a digital thickness gauge. The data were collected in the frequency range between 1.0×10^6 and 50 Hz with the amplitude of 1.00 V at the open circuit potential. σ was calculated from the resistance of the bulk value (R_b). The ILPE was punched into a disc with an electrode area (A) of 3.14 cm^2 .

The ionic conductivity was calculated using the following equation:

$$\sigma = L / (R_b \times A) \quad \text{Eq(1)}$$

where σ is the ionic conductivity, L represents the thickness of the separators, R_b is the bulk resistance, and A is the effective area of the separator disc.

Thermal gravimetric analysis (TGA) was conducted at a heating rate of $20 \text{ }^\circ\text{C}/\text{min}$ under a nitrogen atmosphere from 30 to $750 \text{ }^\circ\text{C}$ by means of a Seiko Instruments TG / DTG 6200 thermal analyser (Seiko Instruments, Inc., Japan).

The melting and crystallisation temperatures, as well as the degree of crystallinity for each film were

measured by differential scanning calorimetry (DSC), with a Q100 (TA Instruments), in the temperature range of 30 to $200 \text{ }^\circ\text{C}$, at a heating rate of $10 \text{ }^\circ\text{C min}^{-1}$ under nitrogen flow. Samples of *ca.* 10 mg were wrapped in aluminium pans and subjected to heating/cooling/heating cycles. An empty aluminium pan was used as a reference.

RESULTS AND DISCUSSION

Physical Observation of ILPE Films

Figure 1 shows the physical condition of the ILPE films with different ratios of PVDF-HFP to $[\text{C}_2\text{C}_1\text{im}][\text{FSI}]$. The films displayed a relatively good homogeneity and appearance when the ionic liquid was $\leq 50 \text{ wt}\%$. This is reflected through good transparency, softness and flexibility, as shown in Fig. 1. However, ILPE-6 formed a jelly-like structure that was tightly glued to the PTFE dish, and the film was too thin, making it difficult to peel off. According to Dzulkipli et al (2019), higher amounts of $[\text{C}_4\text{C}_1\text{im}][\text{BF}_4]$ (about 70%) in PVDF-HFP would make the ILPE gel-like and more flexible. This proves that higher concentrations of ionic liquid could change the physical properties of ILPE due to the plasticising effect [14]. Thus, ILPE-6 was not considered for further examination and study.

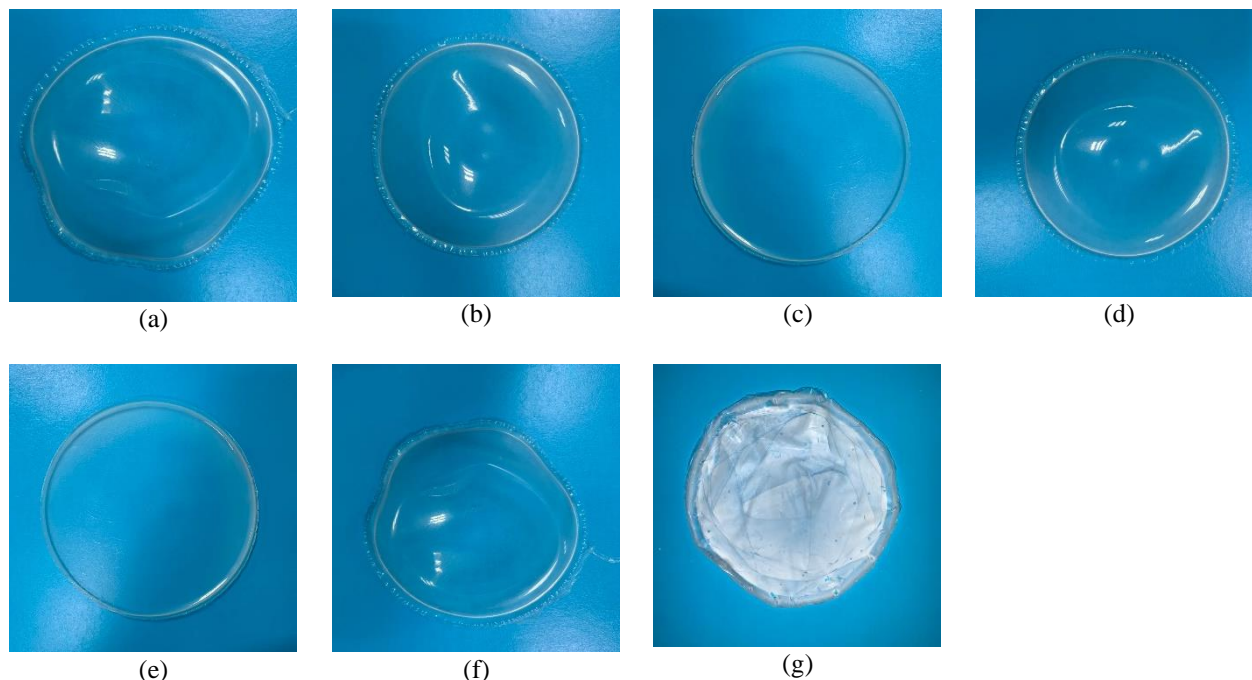


Figure 1. Physical appearance of the different ILPE films (a) Pure PVDF-HFP, (b) ILPE-1, (c) ILPE-2, (d) ILPE-3, (e) ILPE-4, (f) ILPE-5 and (g) ILPE-6.

Fourier Transform Infra-Red (FTIR)

Figure 2 shows that the band appearing for pure PVDF-HFP at 610 cm^{-1} was the mixed mode of CF_2 bending and C-C-C skeletal vibrations. The band at 760 cm^{-1} was related to the CH_2 rocking vibration. The band at 800 cm^{-1} was due to the CF_3 stretching vibration of PVDF-HFP, while the band at 872 cm^{-1} was due to the combined CF_2 and C-C symmetric stretching vibrations. The peak at 986 cm^{-1} was due to the non-polar trans-gauche-trans-gauche' (TGTG') conformation [15]. The CF_3 out-of-plane deformation and antisymmetric CF_2 stretch were visible at 1062 and 1178 cm^{-1} , while the CF_2 stretch appeared at 1400 cm^{-1} [16].

On adding $[\text{C}_2\text{C}_1\text{im}][\text{FSI}]$ to ILPE-1, two peaks were visible at 744 and 833 cm^{-1} that proved the presence of S-N-S symmetric stretching and S-N-S asymmetric stretching [17]. By adding the ionic liquid, these peaks shifted to lower wavenumbers at 736 and 829 cm^{-1} , respectively, for ILPE-5. Two peaks were visible at 1167 and 1071 cm^{-1} , which had shifted from 1178 and 1062 cm^{-1} , and these are related to SO_2 antisymmetric stretching and SO_2 symmetric stretching modes, respectively [18]. The characteristic absorption bands of the imidazole ring could be observed in all ILPEs between 3000 and 3200 cm^{-1} , which are attributed to C-H stretching modes [19].

In the PVDF-HFP structure, the C-F bonds are strongly polarised, with a permanent dipole moment. Therefore, the $[\text{C}_2\text{C}_1\text{im}]^+$ tends to be closed to fluorine atoms, while the $[\text{FSI}]^-$ rotates towards the C atom. On the other hand, hydrogen on the imidazolium ring can interact with fluorine atoms in PVDF-HFP [20]. This feature, combined with its larger molar volume, is responsible for weakening the interactions between the cation and anion in the IL, favouring better interactions with the polymer.

X-Ray Diffraction (XRD)

X-ray diffraction was performed to determine the crystalline state of each ILPE film. Figure 3 displays the XRD patterns of the ILPE films with different ratios. The semi-crystalline structure of pure PVDF-HFP was confirmed by the co-existence of crystalline peaks (42° to 45°) [21]. On adding the ionic liquid, the crystalline peaks began to diminish and gradually flatten. This is in agreement with the theory that an ionic liquid acts as an effective plasticiser for the PVDF-HFP polymer [22]. In ILPE-5, the peaks are not visible because the addition of $[\text{C}_2\text{C}_1\text{im}][\text{FSI}]$ increased the amorphous nature of ILPE, thus reducing crystallinity [23]. The increase in amorphicity was in good agreement with the increase in ionic conductivity. The low degree of crystallinity was due to a weakened polymer chain backbone [24]. From this data, the ILPE-5 sample showed a higher amorphicity compared to PVDF-HFP.

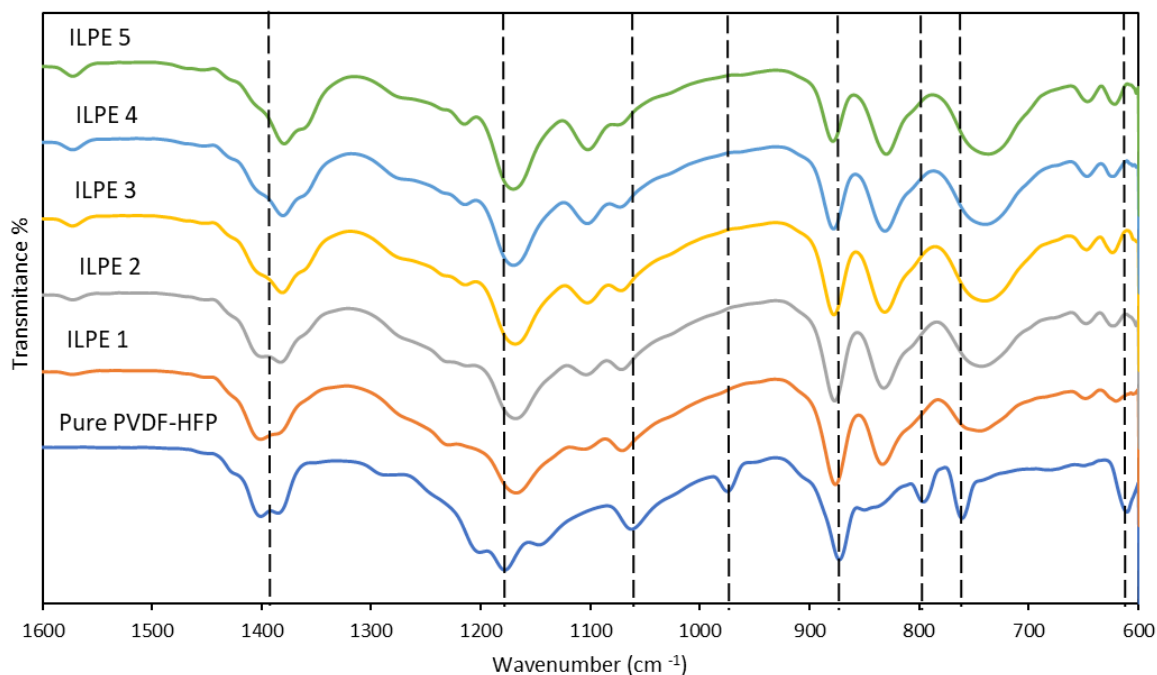


Figure 2. Extended region FTIR spectra.

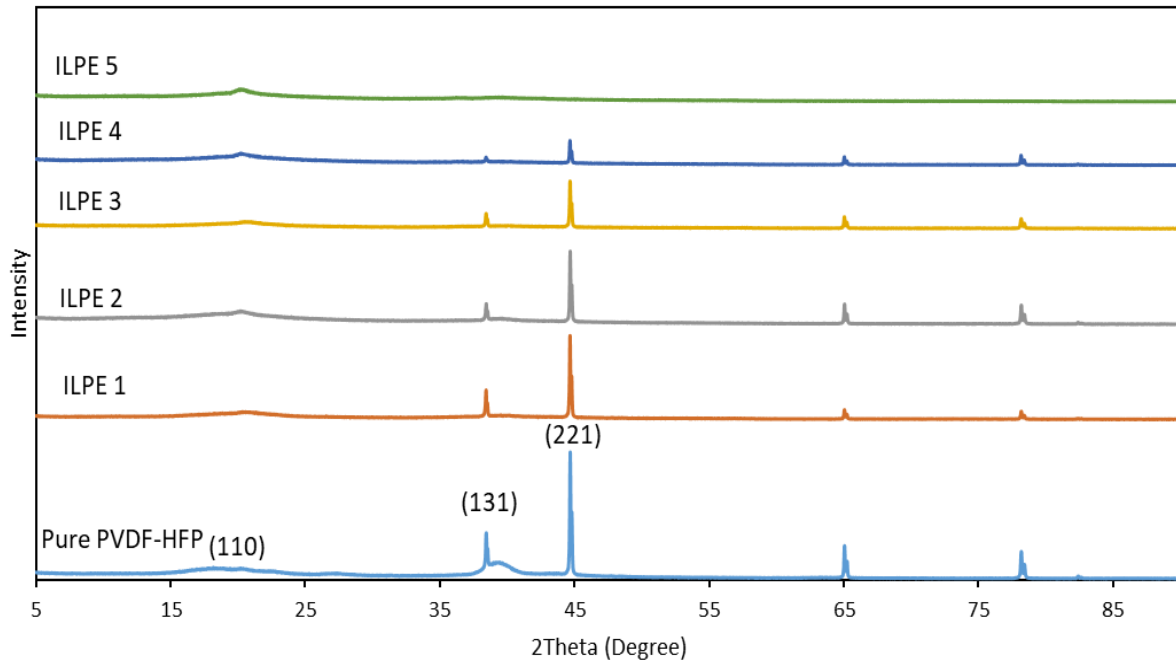


Figure 3. XRD patterns of different ILPE films.

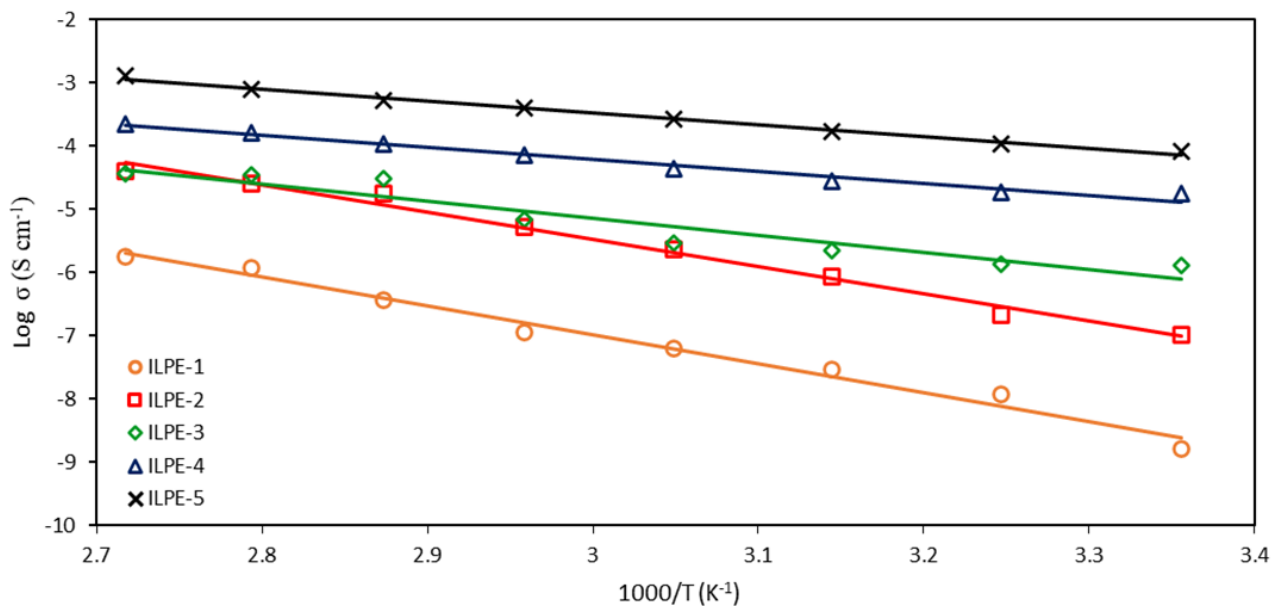


Figure 4. Conductivity of ILPEs with different ratios of ionic liquid.

Ionic Conductivity

Figure 4 shows that the higher the ratio of $[C_2C_1im][FSI]$ ionic liquid, the greater the conductivity. From Eq (1), the ionic conductivity of ILPE-5 was the highest at room temperature, at $8.04 \times 10^{-5} S\ cm^{-1}$. It should be noted here that blending the PVDF-HFP with $[C_2C_1im][FSI]$ would inevitably affect the ionic conductivity of the ILPEs. Rani et al. showed that ionic conductivity increased as PVDF-HFP content decreased, as the number of charge carriers in a membrane with a lower content of PVDF-HFP was

higher [25]. Furthermore, the ionic conductivity was also analysed at different temperatures. The ionic conductivity increased gradually with an increase in temperature. The conductivity increased with rising temperature due to the reduced viscosity and enhanced mobility of polymer chains. This can be explained on the basis of the free volume and the hopping of charge carriers between localised sites [26]. At low temperatures, the regular and ordered arrangement of polymer chains increases the crystallinity of the polymer and hinders the motion of ions, so that the overall ionic conductivity is low. As the temperature

increases, the amorphous nature of the polymer increases progressively; for example, the segmental mobility of the polymer chains increases. This increased segmental mobility can promote inter and intra-chain ion hopping movements, leading to an increase in ionic conductivity [27]. Therefore, ILPE-5 was the optimum ratio for use in battery applications at high temperatures.

Thermogravimetric Analysis (TGA)

The thermal stability of an electrolyte is vital to guarantee acceptable performance in any electrochemical device and has been a critical issue over the past decades. It is of great importance to develop an electrolyte with high thermal stability which can enhance the safety

properties of energy storage and conversion devices. The TGA curves for all ILPEs showed the high thermal stability of these films, as there was no significant weight loss up to 300 °C for all the samples. This is one of the major desirable properties for application in any electrochemical device. In addition, no weight loss was observed below 200 °C for all the ILPE films, which revealed that the samples were free from solvent and moisture. This is in good agreement with the hydrophobic nature of the fluoropolymer and moisture stability of imidazolium-based ionic liquids [28]. The first degradation step, in the range of 380 °C to 500 °C is believed to be due to the degradation of the ionic liquid. The following step, from 500 °C to 700 °C, may be due to the degradation of its polymer host [29].

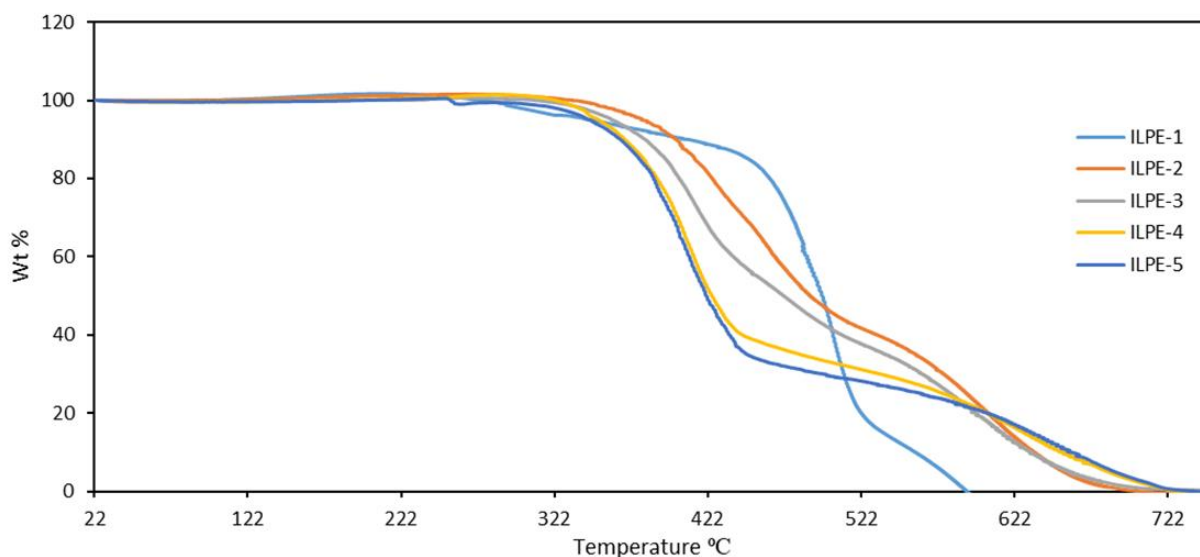


Figure 5. TGA analysis of ILPE films with different ratios.

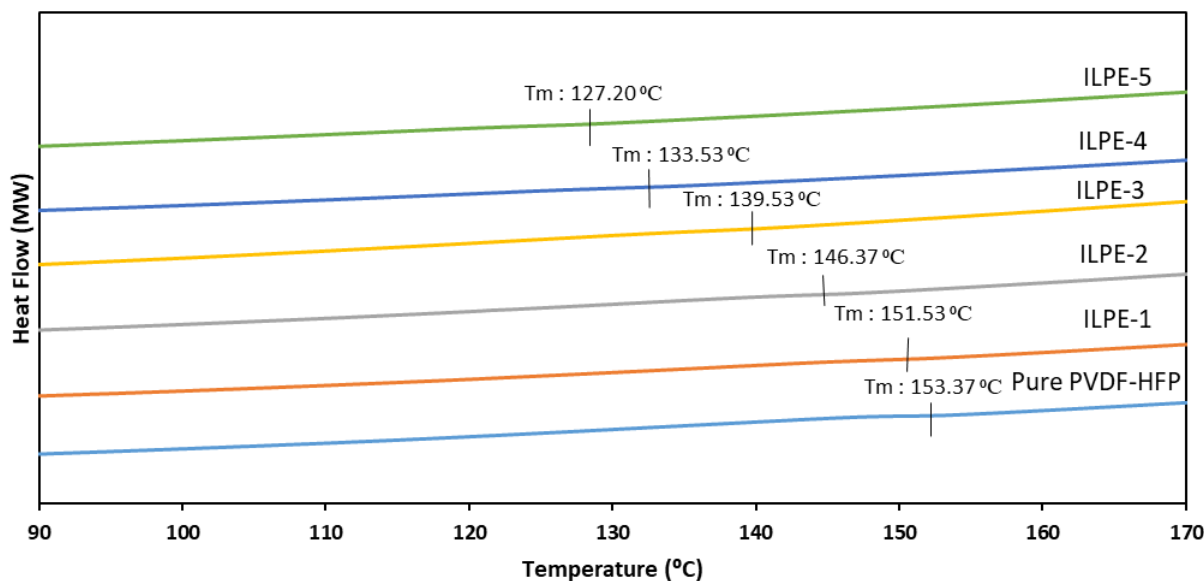


Figure 6. DSC analysis of ILPE films with different ratios.

Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry analysis presents essential information on the thermal characteristics of the electrolyte such as glass transition temperature (T_g) and melting temperature (T_m). Figure 6 shows the heat flow curves in the temperature range of 30 °C to 190 °C for ILPEs containing different ratios of ionic liquid. In ensuring the ILPE films were completely free from any traces of solvent and trapped moisture, all DSC traces were taken during the second heating step. Based on the graph shown in Figure 5, the T_m of the films shifted to a lower temperature on adding [C₂C₁im][FSI]. For example, the T_m value of pure PVDF-HFP was 150.2 °C which was close to the previously reported value of 143 °C [15], while the T_m value of ILPE-5 was 124.5 °C. The shifting of T_m to a lower temperature suggests a higher amorphicity present in the ILPE system. A broader T_m peak was attributed to the presence of liquid components that caused an increase in the amorphous proportion of the overall material [22]. The T_g value was not measured in this work due to instrumental limitations, but it has been reported that the T_g value of pure PVDF-HFP was -35 °C [1].

CONCLUSION

The addition of [C₂C₁im][FSI] ionic liquid into PVDF-HFP improved the physical and chemical properties of the ILPE. The shifting of FTIR peaks at two distinct coordination sites demonstrated the molecular interactions between the host PVDF-HFP polymer and the ionic liquid dopant. The degree of crystallinity of the polymer host decreased with the addition of ionic liquid, which was in a good agreement with the conductivity values. The conductivity values were higher when the percentage ratio of [C₂C₁im][FSI] ionic liquid to PVDF-HFP increased, due to the increase in the number of charge carriers supplied by the [C₂C₁im][FSI] ionic liquid. Thermal measurements showed that all ILPE films had high thermal stability up to 300 °C with minimum weight loss, which signifies an improvement in thermal stability. DSC analysis showed that the melting temperature (T_m) was depressed by the addition of ionic liquid. In general, this study shows that [C₂C₁im][FSI] could serve as a reinforcing agent to replace the traditional volatile and flammable organic solvent and formulate a safer electrolyte system of comparable performance.

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