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Phase Behavior and Electrochemical Characterization of Blends of Perfluoropolyether, Poly(ethylene glycol), and a Lithium Salt

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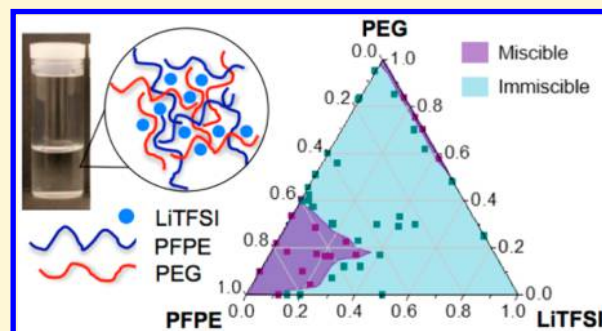
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Supporting Information

ABSTRACT: Electrolytes consisting of low molecular weight perfluoropolyether (PFPE), poly(ethylene glycol) (PEG), and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) blends were prepared and systematically studied for salt concentration and stoichiometry effects on the materials' thermal and electrochemical properties. Herein we report that the tunable ratios of PFPE and PEG allow for precise control of crystalline melting and glass transition temperature properties. These blended liquid polymer electrolytes are inherently nonflammable and remain stable in the amorphous phase from approximately 150 °C down to -85 °C. The ionic conductivity of the electrolytes are on the order of 10⁻⁴ S/cm at 30 °C, which makes them suitable for rechargeable lithium batteries.



INTRODUCTION

Rechargeable batteries are integral to technological development in our society.^{1,2} State-of-the-art lithium-ion (Li-ion) batteries are not only incorporated in zero-emission vehicles and aircraft but also have garnered interest for space and military applications.³ However, the thermal stability of Li-ion batteries, in particular that of the electrolyte, is a critical issue to address. Traditional alkyl carbonates, the most common class of electrolytes for Li-ion batteries, face a high risk of ignition under most operation conditions; this intrinsic instability is increased at high temperatures, and exothermic electrolyte breakdown can lead to thermal runaway.^{4,5} The relatively high freezing point of carbonate-based solvents limits performance and precludes their use at low temperatures required for military and aerospace missions.^{3,6,7}

Amorphous, low glass transition temperature (T_g) polymeric electrolytes are promising candidates for the development of thermally robust Li-ion batteries. Eliminating the use of

flammable solvents significantly enhances battery safety and improves high temperature performance. Poly(ethylene glycol) (PEG also known as poly(ethylene oxide) (PEO)) is the most commonly studied polymer in this regard. Although the ionic conductivity of PEG/lithium salt mixtures can be as high as 10⁻³ S/cm at temperatures above the PEG crystalline melting temperature (typically above 60 °C),⁸ crystalline behavior renders PEG impractical for use at ambient or low temperatures.^{9,10} Extensive studies dedicated to mitigating PEG crystallinity, including strategies such as the addition of copolymers^{11,12} and composite materials,^{13,14} as well as the development of star,¹¹ comb/brush,¹⁵⁻¹⁸ and dendritic¹⁹ polymeric architectures, have resulted in bulk conductive properties that are oftentimes drastically compromised. In

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addition, multistep sophisticated synthetic schemes can increase the cost and complexity of electrolyte preparation.¹⁰

Alternatively, miscible polymer blends could provide a facile and cost-effective method for tuning electrolyte properties. Physical, thermal, and electrochemical properties of miscible polymer blends can be controlled simply by varying the ratios of pure components without synthesis of new materials.²⁰ We have previously reported the surprising miscibility between low molecular weight PEG and perfluoropolyethers (PFPEs).²¹ Previous studies have established that PFPEs belong to a unique class of chemically resistant, noncrystalline, and nonflammable fluoropolymers that exhibit low T_g s and low toxicity.^{21,22} Interestingly, PFPEs can also solvate lithium bis(trifluoromethanesulfonyl)imide (LiTFSI),²³ a commonly used salt in lithium batteries.²⁴

Herein we report on the phase behavior and electrochemical properties of LiTFSI solvated in physical blends of PFPE₁₀₀₀-diol and PEG₄₀₀-diol electrolytes (the subscripts and hyphen after PFPE and PEG indicate the molar masses of the polymers in g/mol and their end-groups, respectively). The number of repeating units in PFPE₁₀₀₀-diol and PEG₄₀₀-diol are similar (approximately 10 and 8 repeating units, respectively). To our knowledge, there is little precedent for investigating the potential use of miscible polymer blends as lithium battery electrolytes. In this article we identify the miscibility windows of ternary PFPE₁₀₀₀-diol/PEG₄₀₀-diol/LiTFSI mixtures, followed by use of Fourier transform infrared (FTIR) and nuclear magnetic resonance (NMR) spectroscopy to study the interactions between LiTFSI and the polymers. Crystallization is studied by differential scanning calorimetry (DSC). Ionic conductivity and transference number are determined by alternating current (AC) impedance and direct current (DC) measurements.

EXPERIMENTAL SECTION

Materials and Sample Preparation. Poly(ethylene glycol), average M_n 400, and lithium bis(trifluoromethanesulfonyl)imide were obtained from Sigma-Aldrich. Perfluoropolyether Fluorolink D10 was obtained from Solvay-Solexis and Santa Cruz Biotechnology. All materials were dried at 90 °C under vacuum in a glovebox antechamber or vacuum oven for at least 24 h prior to use. LiTFSI, PFPE, and PEG were mixed together and stirred at room temperature for at least 12 h. Argon glove boxes (MBraun and Vacuum Atmospheres Company) with low oxygen and water concentration at subppm levels were used for electrochemical sample preparation and characterization.

Polymer Electrolyte Characterization. Ultraviolet–visible spectroscopy (UV–vis) measurements were measured using a Varian Cary 50Bio UV–vis Spectrometer over the wavelength range of 200 to 800 nm. FTIR was performed using a Bruker ALPHA FT-IR instrument from 4000 to 500 cm^{-1} at 4 cm^{-1} resolution under ambient conditions using an attenuated total reflectance (ATR) attachment. DSC thermograms were recorded using a TA Instruments DSC Q200 on samples that were prepared in air in the temperatures ranging from –130 to 100 °C using a heat/cool/heat method at a heating and cooling scanning rate of 10 and 5 °C/min, respectively. Sustained burning data was determined per ASTM D4206, conducted by Kidde Fenwal Combustion Research Center.

¹⁹F NMR was carried out on a Bruker AVANCES00 spectrometer operating at a frequency of 470 MHz at 25 °C. All experiments were performed in the melt by inserting a sealed coaxial insert containing trifluoroacetic acid in D₂O for reference, locking, and shimming into neat electrolyte samples. Line widths reported correspond to the full width at half-maximum of spectra with line broadening set to 0 Hz.

AC impedance spectroscopy was obtained in a procedure similar to that previously reported by Teran et al.²⁴ In brief, the electrolytes were

placed in conductivity cells; sample thicknesses were determined by subtracting the thickness of the lower and upper electrodes from the overall assembled cell thickness. The thicknesses ranged from 1 to 2.5 mm. A custom-made temperature controlled box was used to house the cells during the electrochemical experiments. The AC impedance spectroscopy measurements, performed using a 16-channel Bio-Logic VMP3 potentiostat, were made across the frequency range of 1 MHz to 1 Hz at a peak-to-peak amplitude voltage of 20 mV. The electrolyte resistance was determined by the low-frequency minimum on a Nyquist impedance plot. Measurements were made at a series of temperatures with a minimum of 3 h calibration at each temperature. All data presented in this work are from an initial heating run from 30 to 120 °C, followed by a cooling period until room temperature and a final 10 h stabilization period at –0.5 °C. Standard 2325 coin cells comprising a Celgard 2500 separator impregnated with liquid electrolyte between two Li metal electrodes were assembled. A steady potential of 0.02 V was applied for 10 h after cell stabilization at –0.5 °C, and the cell resistance that includes electrolyte and interface contributions was measured every hour by AC impedance spectroscopy using a DC signal of 0.02 V and an AC signal of 10 mV. The approximate transference number was calculated using methods previously proposed in the literature.²⁵

RESULTS AND DISCUSSION

PFPE/PEG/LiTFSI mixtures (since all of our experiments were conducted on PFPE₁₀₀₀-diol and PEG₄₀₀-diol, we do not mention the molar masses and end-groups of the polymer in the remainder of the article) were quantitatively evaluated by measuring the percent light transmittance and T_g using UV–vis spectroscopy and DSC, respectively. Mixtures were considered miscible if they showed high transmittance of at least 90% in the range of 400 to 800 nm and a single T_g . In contrast, immiscible mixtures were opaque and exhibited two broad T_g s, indicating coexistence of phase separated domains (Figure 1A). The UV–vis transmittance of phase separated mixtures was less than 25%. Immiscible solutions frequently separated into two distinct phases in a few hours. In contrast, miscible solutions were stable on the time scale examined (hours to weeks).

Figure 1B shows the miscibility windows of PFPE/PEG/LiTFSI solutions on a ternary phase diagram. The regions shaded in purple (darker regions) represent the miscible mixtures that were transparent. The region shaded in blue (lighter regions) represents the immiscible mixtures, which were opaque. Concentrations of the three components are specified in terms of weight fractions.

The phase behavior of the PFPE/PEG-diol systems is consistent with previously reported binary mixtures of dimethacryloxy-terminated PFPE (1000 g/mol) (PFPE-DMA) and dimethacryloxy-terminated PEG (700 g/mol) (PEG-DMA) without LiTFSI, where multiple T_g s and low optical transparencies were also observed in immiscible blends. Immiscibility was also shown to occur in PFPE-DMA and 700 g/mol PEG-DMA blends that consisted of high weight fractions of PEG-DMA.²⁶

Binary mixtures of PFPE and PEG are miscible when the weight fraction of PFPE exceeds 0.6, consistent with previous studies.²⁶ The large region of miscibility in Figure 1B comprising ternary mixtures rich in PFPE can be anticipated from the phase behavior of PFPE/PEG and PFPE/LiTFSI binary mixtures. Binary PEG/LiTFSI mixtures are also miscible when the PEG weight fraction exceeds 0.5. Interestingly, adding exceedingly small quantities of PFPE to these mixtures renders them immiscible indicating that the polymer/polymer interactions govern the overall phase behavior of the ternary

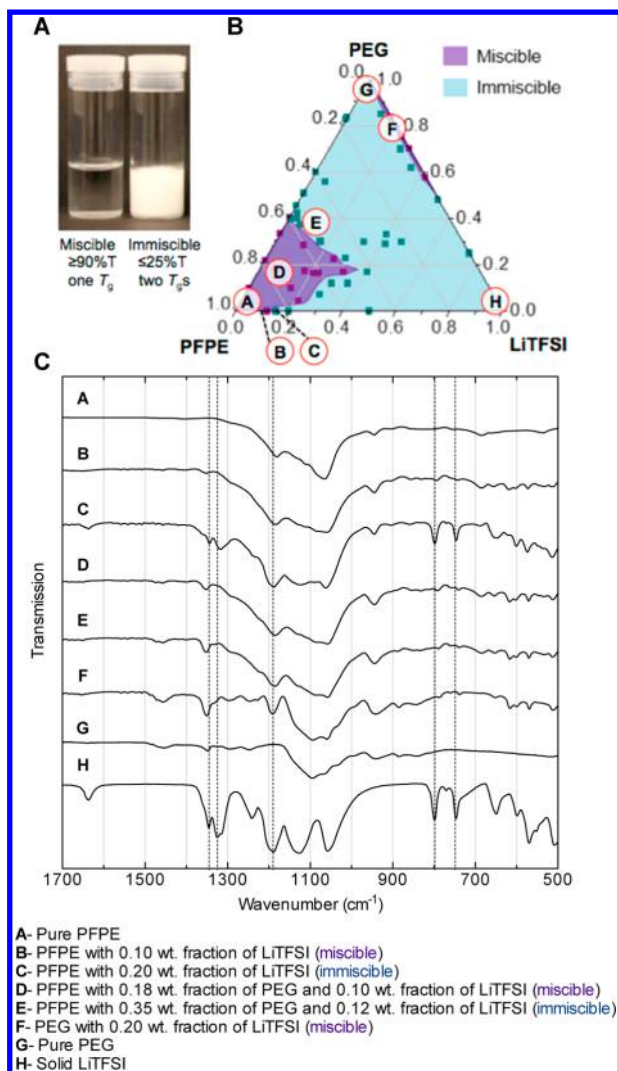


Figure 1. (A) Photograph of miscible and immiscible ternary blends PFPE/PEG/LiTFSI. (B) Ternary phase diagram of PFPE, PEG, and LiTFSI physical blends expressed in weight (wt.) fractions. (C) FTIR spectra of PFPE, PEG, LiTFSI, and blended solutions at various molar ratios. Highlighted guidelines are shown at 748, 798, 1190, 1325, and 1345 cm⁻¹, respectively.

mixture. The fundamental chemical interactions describing this unexpected behavior have not yet been established.

Fourier transform infrared spectroscopy was conducted in order to gain a better understanding of the solubility and phase behavior of these blends. Figure 1C shows the FTIR spectra between 1700 and 500 cm⁻¹ wavenumbers of several binary and ternary systems compared to the spectra of pure LiTFSI, PFPE, and PEG. The compositions of the systems of interest are identified by the letters A through H in Figure 1B.

Pure solid LiTFSI (H) exhibits characteristic bands at 1190 cm⁻¹, from SO₂ asymmetric stretching, at 1325 and 1345 cm⁻¹, from CF₃ asymmetric stretching, and at 748 and 798 cm⁻¹, which originate from symmetric S–N–S stretching. The presence of these peaks in mixtures is commonly taken as an indication of the presence of LiTFSI aggregates.^{27–32} All of the miscible solutions (B, D, and F) show a substantial reduction in peak intensity at 748, 798, 1190, 1325, and 1345 cm⁻¹, indicating the absence of LiTFSI aggregates. These observations are thus consistent with the presence of dissociated TFSI⁻ ions.^{23,33}

Interestingly, signatures of dissociated TFSI⁻ were also observed in the FTIR spectra of some immiscible ternary mixtures (e.g., E). This suggests that immiscibility in this mixture is mainly driven by the limited miscibility of the two polymers rather than the limit of solubility of LiTFSI in the polymers. We conclude that blend E contains PEG-rich and PFPE-rich phases with LiTFSI dissolved in both phases. In contrast, the immiscible binary PFPE/LiTFSI mixture (C) shows bands at 748 and 798 cm⁻¹ indicating the presence of LiTFSI aggregates in this system. Increased absorption at 1190, 1325, and 1345 cm⁻¹ is also observed in mixture C. However, interpretation of FTIR spectra in these regions is affected by the interference of peaks from pure PFPE (A).

The crystalline melting behavior of binary PFPE/PEG blends are shown in Figure 2. Increasing amounts of PFPE reduce the

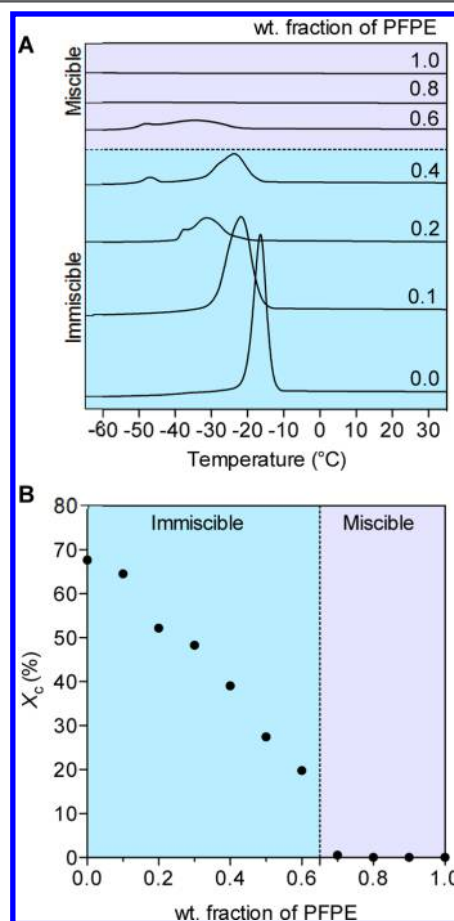


Figure 2. (A) DSC cooling traces of PFPE and PEG blends at various PFPE weight fractions. (B) Percent crystallinity, expressed in X_c , as a function of PFPE weight fraction.

enthalpy of crystallization (Figure 2A). Complex crystallization behavior is observed in immiscible PFPE/PEG blends where PFPE weight fractions are below 0.6. No crystallization is observed in miscible PFPE/PEG blends with PFPE weight fraction greater than 0.8. The degree of crystallinity, X_c , is given by

$$X_c = \left(\frac{\Delta H}{\Delta H_0} \right) \times 100$$

where ΔH is the crystallization enthalpy of the blend and ΔH_0 is that of a 100% crystalline PEO, reported as 166 J/g.³⁴ As

shown in Figure 2B, crystallinity decreases as the weight fraction of PFPE increases and reduces to zero when the PFPE weight fraction exceeds 0.7.

It is generally assumed that amorphous polymer/salt mixtures are ideally suited for battery electrolyte applications. We thus chose to study ternary mixtures wherein the weight fraction of PFPE in the liquid components is 0.8. In other words, the weight ratio of PFPE to PEG remains consistently at 80 to 20 even as LiTFSI is added; this blend is henceforth referred to as PFPE/PEG. This blend is noncrystalline (no detection of T_m or T_c), exhibits a single low T_g of -86°C , and shows good thermal stability (T_d (5%) = 198°C in nitrogen, 207°C in air). Moreover, PFPE/PEG is nonflammable even with dissolved LiTFSI (see Supporting Information). In the discussion below, we compare the properties of this PFPE/PEG blend with added LiTFSI to that of PFPE/LiTFSI and PEG/LiTFSI blends. For all three systems, the salt concentration is quantified by r , defined as the ratio of Li^+ ions to oxygen atoms. This is because of the well-established fact that the solvation shells surrounding dissociated ions are rich in oxygen.

In order to study the effect of LiTFSI on the thermal properties of the PFPE/PEG blends, DSC experiments were performed on binary and ternary blends containing LiTFSI. Figure 3 shows the linear increase of T_g with increasing salt

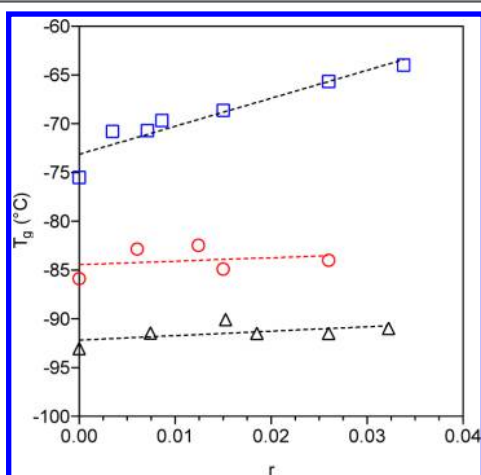


Figure 3. T_g of PFPE (Δ), PEG (\square), and PFPE/PEG blend (\circ) electrolytes as a function of salt concentration r ; dotted lines represent fitted linear regressions.

concentration r in all three systems. This is expected, as molecular dynamics simulations and neutron diffraction data indicate that oxygen atoms in the PEG backbone coordinate with lithium ions to form temporary cross-links,^{16,35–37} limiting polymer chain mobility and resulting in a higher T_g . Interestingly, the slope of T_g versus r is larger for PEG than PFPE and PFPE/PEG blend, indicating that the T_g of PEG is more sensitive to LiTFSI concentration. The effect of r on T_g is less pronounced in PFPE-containing electrolytes, as the ether oxygens are less nucleophilic due to the strong electron withdrawing fluorines in the PFPE backbone, which, in turn, weakens interactions between Li^+ and O. The PFPE/PEG blend demonstrates T_g vs r behavior similar to PFPE. More importantly, it is clear that blending PFPE with PEG reduces T_g relative to pure PEG. In the absence of LiTFSI, the measured T_g of pure PFPE/PEG blend is within 3°C of the calculated T_g as predicted by the Fox equation³⁸ for miscible binary systems:

$$\frac{1}{T_g} = \frac{x_1}{T_{g,1}} + \frac{x_2}{T_{g,2}}$$

where $T_{g,1}$ and $T_{g,2}$ pertain to the pure components, and x_1 and x_2 are the weight fractions of each component in the blend. Agreement with the Fox equation further confirms the miscibility of the two polymers. At constant $r = 0.026$, the T_g of the blended electrolyte is also almost 20°C lower than PEG, successfully extending the temperature range in which the electrolyte remains rubbery.

Interactions between LiTFSI and polymer chains were also investigated via ^{19}F NMR. Figure 4 depicts the ^{19}F NMR spectra of PFPE, PEG, and PFPE/PEG blend electrolytes all with $r = 0.026$ compared to a 0.05 M solution of LiTFSI in deuterium oxide (D_2O) and pure PFPE. Each sample contains a coaxial insert spiked with a 5 wt % solution of trifluoroacetic acid in D_2O whose signal was calibrated to -76.5 ppm for referencing.

Since PFPEs are random linear copolymers: $\text{X}-\text{O}-[(\text{CF}_2\text{CF}_2\text{O})_m-(\text{CF}_2\text{O})_n]-\text{X}$ where $m/n \approx 7/3$ and $\text{X} = -\text{CF}_2\text{CH}_2\text{OH}$, chemical shifts in the ^{19}F NMR spectra of the PFPEs are sensitive to the distinct location of the fluorinated atoms along the chain and can be categorized into three distinct regions (i) interior CF_2 groups give rise to peaks between -55 and -60 ppm, (ii) terminal CF_2 groups give rise to peaks between -82 and -87 ppm, and (iii) interior CF_2-CF_2 groups give rise to peaks between -90 and -95 ppm (Figure 4). The multiple peaks in each region arise from the different combinations of neighboring repeating units.^{39,40} For example, the most downfield peak in region (i) (-55 to -60 ppm) is assigned to a CF_2O unit neighboring two $\text{CF}_2\text{CF}_2\text{O}$ units on each side. Likewise, the middle peak corresponds to the CF_2O unit in between a CF_2O and $\text{CF}_2\text{CF}_2\text{O}$ group, and the most upfield peak in that region corresponds to a CF_2O unit in between two CF_2O groups.

The chemical shifts and full widths at half-maximum (FWHM) of NMR peaks in the LiTFSI region and region (ii) of PFPE are listed in Table 1. The FWHM values of peaks associated with LiTFSI in both PEG and D_2O are 3 Hz. This suggests the presence of mobile TFSI $^-$ ions in these systems. In contrast, the FWHM of the same peak in PFPE and PFPE/PEG blend is 11 and 6 Hz, respectively. There are two possible reasons for this observation: (1) a substantial decrease in the mobility of TFSI $^-$ ions and (2) an increase in the number of microenvironments surrounding the fluorinate fluorine atoms. A decrease in the mobility of the anion dissolved in the PEG, PFPE/PEG, and PFPE, respectively, should effectively increase the Li^+ transference number, t^+ , of the electrolyte, defined as the fraction of the overall current carried by the Li ions; this is consistent with our results discussed later.

The FWHM of the fluorine peaks in region (ii) of PFPE is 38 Hz for pure PFPE, 39 Hz for PFPE/LiTFSI, and 72 Hz for PFPE/PEG/LiTFSI. There are two possible explanations for these observations: (1) the addition of high T_g PEG slows down segmental motion of PFPE and (2) an increase in the number of microenvironments surrounding the fluorinate atoms. Note that the addition of PEG to PFPE/LiTFSI decreases FWHM in the LiTFSI fluorine region but increases the FWHM in the PFPE fluorine end group region (ii). The addition of PEG to PFPE/LiTFSI results in an upfield shift of the peak position in both LiTFSI and PFPE regions (see first three entries in Table 1). This is primarily due to changes in solvent polarity. The TFSI $^-$ ions are more deshielded in the

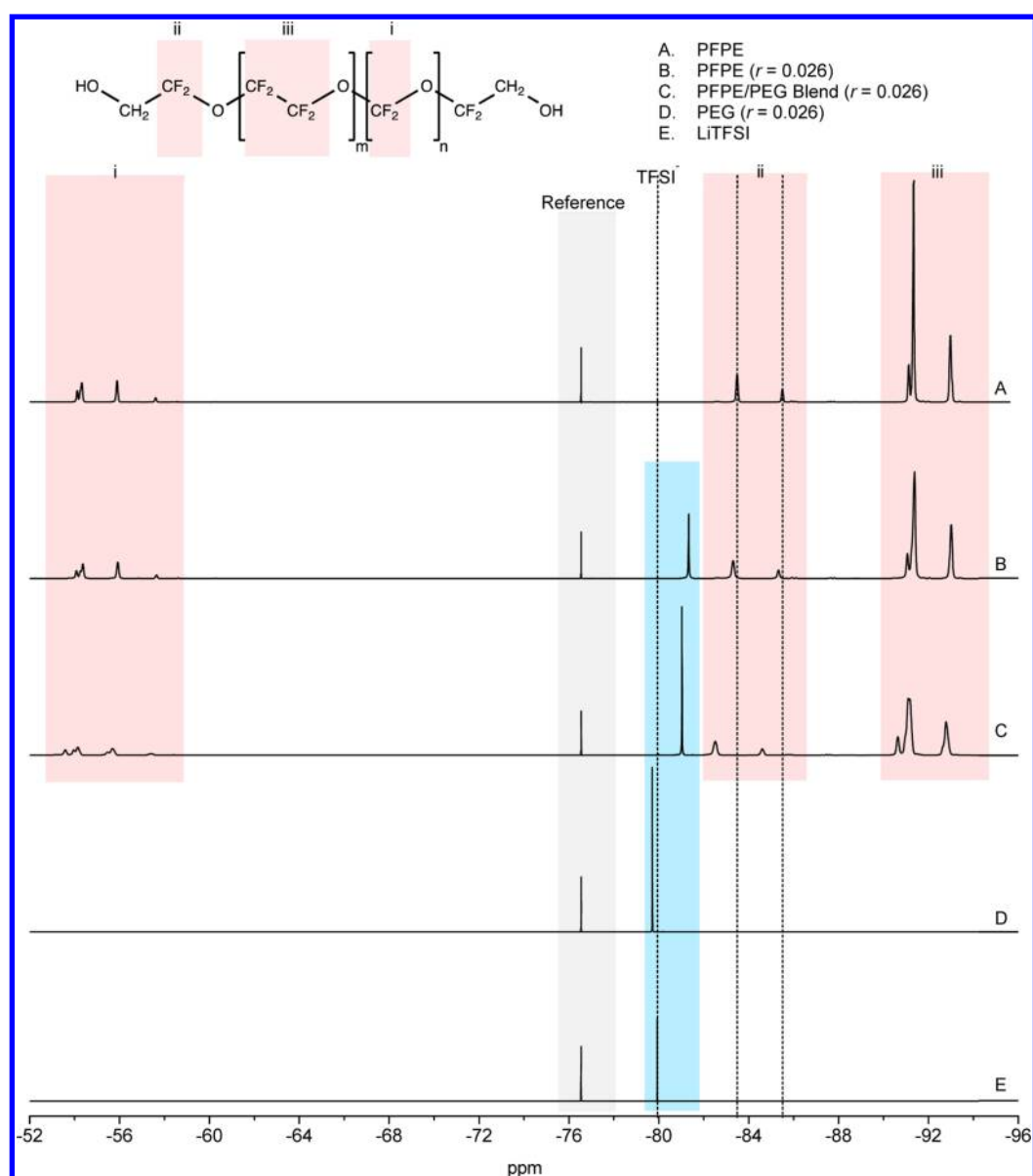


Figure 4. ^{19}F NMR spectra of PFPE, PEG, and PFPE/PEG electrolytes in reference to 0.05 M LiTFSI in D_2O and pure PFPE. Highlighted guidelines are shown at -79.94 , -83.49 , and -85.51 ppm.

Table 1. Selected Peaks and Full Widths at Half-Maximum of ^{19}F NMR for PEG, PFPE/PEG, and PFPE Electrolytes

sample	LiTFSI		PFPE (region (ii))	
	chemical shift (ppm)	FWHM (Hz)	chemical shift (ppm)	FWHM (Hz)
PFPE	n.a.	n.a.	$-83.5, -85.5$	46, 38
PFPE ($r = 0.026$)	-81.4	11	$-83.3, -85.3$	59, 49
PFPE/PEG blend ($r = 0.026$)	-81.0	6	$-82.5, -84.6$	82, 72
PEG ($r = 0.026$)	-79.7	3	n.a.	n.a.
LiTFSI in D_2O	-79.9	3	n.a.	n.a.

presence of PEG, as it is more polar than PFPE. Further upfield shifts in the TFSI region are seen in PEG and D_2O samples due to this polarity effect.

In the PFPE/PEG blend, the TFSI $^-$ fluorine signal arises as a single peak, with a chemical shift in between that of TFSI $^-$ in PFPE and PEG. This indicates, on the time scale faster than

experimental NMR acquisition, the average electronic environment experienced by TFSI $^-$ is homogeneous. This also crucially implies that miscible solutions of PFPE/PEG blends do not simply offer a combination of ionic environments from pure PFPE and PEG (which would have likely resulted in TFSI peak splitting or two separate peaks), but produces a uniform environment intermediate of both materials.

The ionic conductivity of PFPE, PEG, and PFPE/PEG blend electrolytes was measured at a salt concentration of $r = 0.026$ at temperatures between 0 and 120 $^\circ\text{C}$, as shown in Figure 5.

Pure PEG exhibits conductivity of between 3×10^{-5} to 4×10^{-3} S/cm in this temperature range, similar to values previously reported for low molar mass PEGs.^{12,24} However, the PFPE/PEG blend exhibits conductivity between 1×10^{-5} to 2×10^{-3} S/cm, lower than pure PEG but drastically higher than pure PFPE. The conductivities of these electrolytes are lower than conventional carbonate electrolytes ($\sim 7 \times 10^{-3}$ S/cm at room temperature)⁴¹ but are comparable to other nonflammable electrolyte systems with low T_g .⁴² In each case,

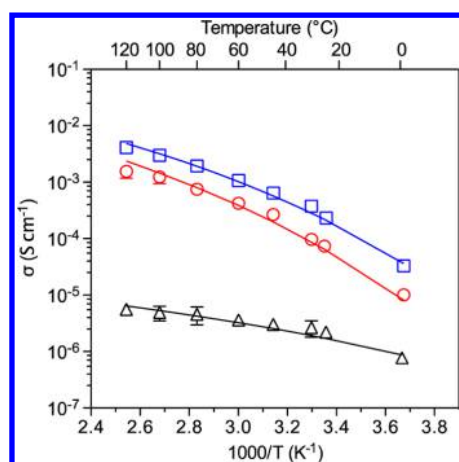


Figure 5. Temperature-dependent ionic conductivities of PFPE (Δ), PEG (\square), and PFPE/PEG blend (\circ) electrolytes ($r = 0.026$); each sample follows VTF behavior.

the ionic conductivity also increases with increasing temperature, typical of polymer electrolytes. The temperature dependence of the ionic conductivity of these materials was found to be well-described by the Vogel–Tamman–Fulcher (VTF) equation:^{43–45}

$$\sigma(T) = \frac{A}{\sqrt{T}} \exp\left(\frac{-B}{R(T - T_0)}\right)$$

where σ is the ionic conductivity, A is a constant proportional to the number of charge carriers, B is equivalent to the activation energy for ion motion, R is the gas constant, T is the experimental temperature, and T_0 is an empirical reference temperature taken as the idealized temperature corresponding to zero configurational entropy, typically chosen to be 50 K below the T_g of each sample.²⁷ Fits to this equation are shown in Figure 5 as solid lines for each data set, and the corresponding parameters are shown in Table 2.

Table 2. VTF Fitted Parameters for PEG, PFPE/PEG, and PFPE Electrolytes Containing LiTFSI ($r = 0.026$)

electrolyte	A ($S \text{ cm}^{-1} \text{ K}^{1/2}$)	B (kJ mol^{-1})	T_0 (K)
PEG	12.3	9.5	157
PFPE/PEG blend	12.3	10.9	157
PFPE	1.6×10^{-3}	5.5	132

By fitting measured conductivity using the VTF equation, it is clear that despite lower T_0 and activation energy parameters, the ionic conductivity of pure PFPE is low due to a significantly lower amount of charge carriers. The addition of PEG clearly enhances the number of charge carriers and conductivity for the overall electrolytes while retaining PFPE-like thermal properties (see Figure 3).

We have previously reported that pure PFPE-based electrolytes exhibit Li^+ transference numbers, t^+ , close to unity.²³ We use the same method described in ref 23 to estimate t^+ of PFPE, PEG, PFPE/PEG electrolytes, with $r = 0.026$, at -2 °C. We obtained t^+ values of 0.84 (PFPE), 0.17 (PEG), and 0.29 (PFPE/PEG). It is important to know that our method becomes increasingly inaccurate as the transference number decreases. The addition of PEG dramatically reduces the transference number of the PFPE electrolytes.

CONCLUSIONS

Physical blends of low molecular weight PFPE and PEG were prepared. Their miscibility, ability to solvate LiTFSI, thermal properties, and electrochemical properties were explored. An optimal ratio of PFPE to PEG was selected for electrochemical characterizations. Conductivity of this electrolyte at LiTFSI concentration $r = 0.026$ was measured to be $2 \times 10^{-4} \text{ S/cm}$ at 30 °C, slightly lower than that of pure PEG at the same salt concentration but approximately 2 orders of magnitude higher than pure PFPE electrolytes. In addition, blending PEG with PFPE reduces the T_g and prevents crystallization. The uniqueness of PFPE/PEG electrolytes arises from the fact that they enable control over physical, thermal, and electrochemical properties of a polymer electrolyte by simple blending rather than schemes that involve new chemical synthesis.

ASSOCIATED CONTENT

Supporting Information

Typical DSC curve of immiscible liquids, and thermogram and transference number data of the PFPE/PEG blend electrolyte. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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