Ion Conductors

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Single Lithium-Ion Conducting Polymer Electrolytes Based on a Super-Delocalized Polyanion

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Abstract: *A novel single lithium-ion (Li-ion) conducting polymer electrolyte is presented that is composed of the lithium salt of a polyanion, poly[(4-styrenesulfonyl)(trifluoromethyl(S-trifluoromethylsulfonylimino)sulfonyl)imide] (PSsTFSI$^-$), and high-molecular-weight poly(ethylene oxide) (PEO). The neat LiPSsTFSI ionomer displays a low glass-transition temperature (44.3°C; that is, strongly plasticizing effect). The complex of LiPSsTFSI/PEO exhibits a high Li-ion transference number ($t_{Li^+} = 0.91$) and is thermally stable up to 300°C. Meanwhile, it exhibits a Li-ion conductivity as high as 1.35×10^{-4} S cm^{-1} at 90°C, which is comparable to that for the classic ambipolar LiTFSI/PEO SPEs at the same temperature. These outstanding properties of the LiPSsTFSI/PEO blended polymer electrolyte would make it promising as solid polymer electrolytes for Li batteries.*

Solid polymer electrolytes (SPEs) for solvent-free lithium (Li) rechargeable batteries have captured much attention owing to their potential advantages, including safety, ease of packaging, excellent flexibility and containment, and their functioning as a separator compared with liquid or gel electrolytes.[1–8] To date, great progress has been made by utilizing LiTFSI (Li[$(CF_3SO_2)_2N$])/poly(ethylene oxide) (PEO) SPEs for Li/$LiFePO_4$ batteries, which is being used as power source for an electric car, Autolib.[9] However, conventional SPEs formed by dissolving Li salt with a small anion in a polymer host (usually high-molecular-weight PEO), are dual-ion conductors, in which both cations and anions are mobile.[10–14] This would generate the concentration gradients of the salt and cell polarization, which causes premature battery failure.[15,16]

One of the best solutions to the above-mentioned problem is to prepare single Li-ion conductors (SLICs), in which the anions are immobilized by anion trapping agents or by fixing the anions to the polymeric backbones,[17–20] so that the transference number (t_{Li^+}) of Li^+ cations is close to one. Until now, most of SLICs have been synthesized by grafting common alkyl carboxylate (RCO_2^-)[17] and/or sulfonate (RSO_3^-)[18] anions to the polymeric backbones. However, the ion dissociation in polyether media is very limited, owing to the low degrees of negative charge distribution in these anions.

Armand et al.[21] have reported a new type of SLICs by blending lithium poly[(4-styrenesulfonyl)(trifluoromethanesulfonyl)imide] (LiPSTFSI) with PEO. Owing to the enhanced flexibility and improved negative charge distribution of the $-SO_2-N^{(-)}-SO_2-CF_3$ vs. common $-CO_2^-$ and $-SO_3^-$ structures, the relatively high ionic conductivity of about 10^{-5} S cm^{-1} at 70°C has been obtained for the blended polymer electrolytes. Then, the conductivities were further improved by elegantly preparing random and triblock copolymers of LiPSTFSI through incorporating ethylene oxide (EO) units in side and main chains, respectively.[22,23] A prototype Li battery using this triblock polymer electrolyte exceeds a conventional battery based on a dual-ion polymer electrolyte, suggesting the importance of single Li-ion nature in improving the cycling performances. Inspired by these results, we envisaged that it would be beneficial to further enhance ionic conductivities of such imide anion-based SLICs by increasing its negative charge distribution.

Herein, we report a new type of single Li-ion conducting polymer electrolytes, which are prepared by simply dissolving lithium salt of a super-delocalized polyanion, namely poly[(4-styrenesulfonyl)(trifluoromethyl(S-trifluooromethylsulfonylimino)sulfonyl)imide] (PSsTFSI$^-$; Scheme 1) in PEO. We devised this kind of new anion structure on the base of following consideration. Replacing a =O group in above $-SO_2-N^{(-)}-SO_2-CF_3$ structure by a strong electron-withdrawing group, $=NSO_2CF_3$, can create much more delocalized anion structure ($-SO_2-N^{(-)}-SO(=NSO_2CF_3)-CF_3$, sTFSI$^-$), in which the Li^+ cations would be highly dissociated from this super-delocalized anion when mixed with a polymer media (that is, PEO), so that conductivities can be further improved.[24]

The procedures for preparing the monomeric and polymeric salts are detailed in the Supporting Information. The structure and composition of the monomer (KSsTFSI),

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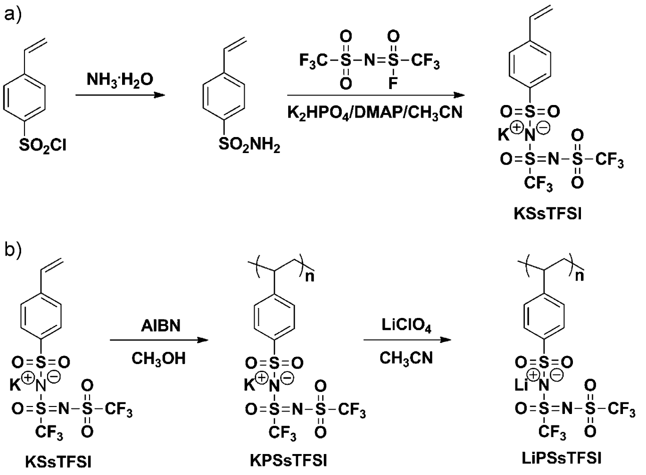

Scheme 1. The synthetic routes of a) monomer and b) polymers. DMAP = dimethylaminopyridine, AIBN = azodiisobutyronitrile.

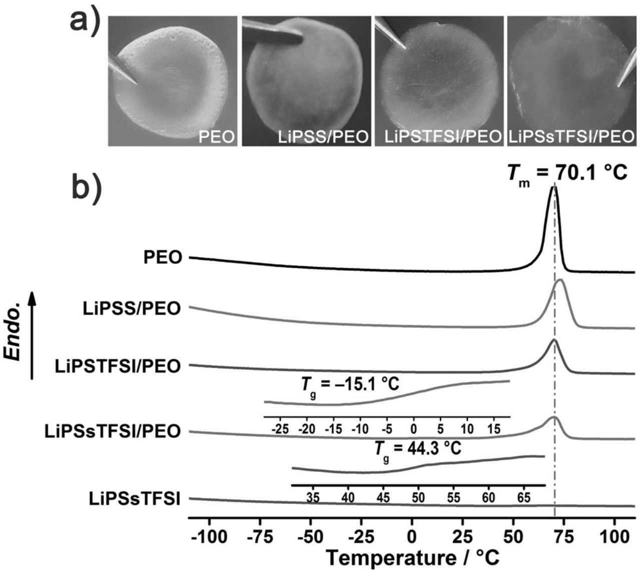

Figure 1. a) Photographs of membrane for the neat PEO and the LiX/PEO (X = PSS, PSTFSI, and PSsTFSI) blended polymer electrolytes (EO/Li^+ = 20). b) DSC traces of neat PEO, LiPSsTFSI, and the LiX/PEO (X = PSS, PSTFSI, PSsTFSI) blended polymer electrolytes (EO/Li^+ = 20) at the first heating scan.

intermediate (KPSsTFSI), and target salt (LiPSsTFSI) were characterized by ^{1}H and ^{19}F NMR (Supporting Information, Figure S1). The neat LiPSsTFSI salt has a number-average molecular weight (M_n) of about 2×10^5 g mol^{-1} with a polydispersity index (PDI) of 2.21 (Supporting Information, Table S1), and its complex of LiPSsTFSI/PEO exhibits an oxidation potential at ca. 4.0 V vs. Li^+/Li (Supporting Information, Figure S2), and is thermally stable up to 300 °C (Supporting Information, Figure S3).

To understand the impact of degree of negative charge distribution in anion on the ionic conductivity of SPEs reliably, the two related lithium polymer salts, lithium poly(4-styrenesulfonate) (LiPSS) and LiPSTFSI (Supporting Information, Figure S4), were also synthesized using the same methods as described previously.[22,25] The membranes of all of the blended polymer electrolytes were prepared by a solution-casting method (the procedures are detailed in Supporting Information).

Figure 1 a shows the photographs of membrane for the neat PEO and the LiX/PEO (X = PSS, PSTFSI, and PSsTFSI) blended polymer electrolytes at a molar ratio of EO/Li^+ = 20, as the concentration of lithium salt to EO unit in the classic LiTFSI/PEO SPEs around this ratio region has been found to afford relatively high ionic conductivities in medium high temperatures, and its ionic conduction is little affected by its heat history.[26] It can be seen that self-standing, translucent membranes are obtained. It is worthy to note that the membrane of LiPSsTFSI/PEO blended electrolyte has a better mechanical ductility than both the corresponding LiPSTFSI/PEO and LiPSS/PEO electrolytes, which is perceptible from appreciable manual bending or stretching. This would be attributed to the much better flexibility and greatest delocalized negative charge distribution of the $-SO_2-N^{(-)}-SO(=NSO_2CF_3)-CF_3$ structure (in PSsTFSI$^-$), among these three polyanions.

Figure 1 b comparatively shows the DSC traces of the neat PEO, LiPSsTFSI, and the three blended polymer electrolytes of LiX/PEO (X = PSS, PSTFSI, PSsTFSI) with the same molar ratio of EO/Li^+ = 20 at the first heating scan (all the measurement data are summarized in the Supporting Information, Table S2). The neat PEO shows a sharp melting transition peak at 70.1 °C, which is consistent well with the previous reports for neat PEO.[27,28] Important note is that the neat LiPSsTFSI salt displays a low glass transition temperature (T_g) at 44.3 °C (Figure 1 b; Supporting Information, Figure S5). This would be attributable to highly flexibility and super-delocalized negative charge distribution of the $-SO_2-N^{(-)}-SO(=NSO_2CF_3)-CF_3$ structure in PSsTFSI$^-$, which impedes the motion of both Li^+ cations and PSsTFSI$^-$ anions to rearrange orderly in space (that is, slow kinetics of crystallization), thus making LiPSsTFSI glassformer at low temperature. In contrast, glass transitions below 50 °C have not observed for any neat Li ionomer based on lesser delocalized anions (for example, $-CO_2^-$, $-SO_3^-$, and $-SO_2-N^{(-)}-SO_2-CF_3$) without PEO blending.[17,21,22,25] It is interesting to note that a glass transition at the low temperature of −15.1 °C is also observed for the LiPSsTFSI/PEO blended electrolyte (this glass transition at the low temperature of −15.3 °C is repeatedly observed at the second heating scan; Supporting Information, Figure S6), and is however not detected for both the LiPSS/PEO and LiPSTFSI/PEO electrolytes. This result clearly indicates the stronger plasticizing effect of the PSsTFSI$^-$ vs. PSS$^-$ and PSTFSI$^-$ anions, which is consistent with the glassformer nature of neat LiPSsTFSI, and is favorable for enhancing segmental motions of PEO in the blended electrolyte. The glass transition temperature is a qualitative signature of ion mobility in SPEs, and a low glass transition temperature for LiPSsTFSI/PEO is a sign of high ion mobility in the blended electrolyte. Moreover, the LiPSsTFSI/PEO blended electrolyte displays the lowest value of enthalpy of melting (that is, ΔH_m = 58.3 J g^{-1}; Supporting Information, Table S2), which is obviously lower than that for the LiPSTFSI/PEO electrolyte (that is, $\Delta H_m = 71.9\ \mathrm{J\,g^{-1}}$; Supporting Information, Table S2), and is significantly lower than that for the LiPSS/PEO electrolyte (that is, $\Delta H_m = 101.2\ \mathrm{J\,g^{-1}}$; Supporting Information, Table S2). This is a signature of a higher degree of amorphous phase in the former (LiPSsTFSI/PEO) than in the latter two (LiPSS/PEO and LiPSTFSI/PEO). All above results obtained from DSC characterization suggest that the LiPSsTFSI/PEO blended electrolyte has higher degrees of amorphous phases and larger segmental motions than both the LiPSS/PEO and LiPSTFSI/PEO electrolytes, which are beneficial for improving its ionic conductivities.

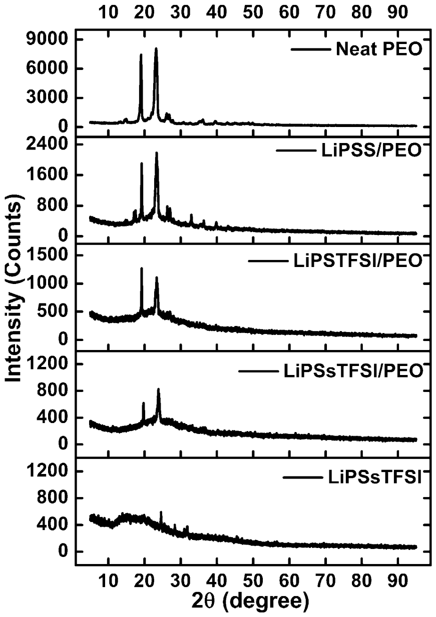

Figure 2. XRD patterns of the LiX/PEO (X = PSS, PSTFSI, PSsTFSI) blended polymer electrolytes ($\mathrm{EO/Li^+} = 20$), as well as the neat PEO and LiPSsTFSI.

Figure 2 comparatively displays the XRD patterns of the LiX/PEO (X = PSS, PSTFSI, PSsTFSI) blended polymer electrolytes with a molar ratio of $\mathrm{EO/Li^+} = 20$, as well as the neat PEO and LiPSsTFSI for comparison. As shown in Figure 2, two sharp diffraction peaks are observed at $2\theta = 19.2°$ and $23.3°$ for the neat PEO, suggesting the nature of highly crystalline phase.[14] In contrast, no characteristic diffraction peak is observed for the neat LiPSsTFSI salt, indicating a mainly amorphous phase. It is interesting to note that the intensities of these two representative XRD peaks for neat PEO decrease rapidly with addition of the polymer salts in the anion order of $\mathrm{PSS^-} < \mathrm{PSTFSI^-} < \mathrm{PSsTFSI^-}$. This is totally consistent with the degree of negative charge distribution and freedom (that is, structural flexibility) of the anion. This trend in XRD patterns is also well consistent with the DSC results in Figure 1b (Supporting Information, Table S2), wherein the complex of LiPSsTFSI/PEO shows low glass transition before melting (that is, low tendency to crystallize), while the latter two complexes of LiX/PEO (X = PSS, PSTFSI) only show melting with larger values of enthalpy (that is, a higher degree of crystalline phase) without glass transitions (Supporting Information, Table S2).

Figure 3a compares the temperature dependence of ionic conductivities (σ) for the LiX/PEO (X = PSS, PSTFSI, PSsTFSI) blended polymer electrolytes with a molar ratio of $\mathrm{EO/Li^+} = 20$, as well as the classic LiTFSI/PEO blended electrolyte for comparison, and some of the representative data are presented in the Supporting Information, Table S3. As seen from Figure 3a, our conductivity values for the classic LiTFSI/PEO blended electrolyte are consistent with the previous report,[26] indicating that our measurements are reliable. As a common feature for the PEO-based SPEs,[29] an increase in the ionic conductivity to a variable extent is observed with increasing the temperature up to about 70 °C for all the LiX/PEO (X = PSS, PSTFSI, PSsTFSI, TFSI) blended electrolytes (Figure 3a), owing to a crystalline melting transition of the PEO host and/or the free-volume activation. The ionic conductivities for all these four kinds of blended electrolytes decrease in the order of LiTFSI/PEO > LiPSsTFSI/PEO > LiPSTFSI/PEO > LiPSS/PEO in the measured temperature range from 25 to 90 °C. It is generally accepted that the ionic conductivity of SPEs is mainly governed by the polymer crystallinity and/or glass transition temperature, and the charge carrier concentration.[2,22,25,30] The highest ionic conductivities for the blended electrolyte containing LiTFSI are observed, which is expected from that both the $\mathrm{Li^+}$ cations and relatively small $\mathrm{TFSI^-}$ anions are mobile in the complex of LiTFSI/PEO. By contrast, the three blended electrolytes containing the polymer salts show obviously lower ionic conductivities, which is due to the huge volume of the polymeric anion.

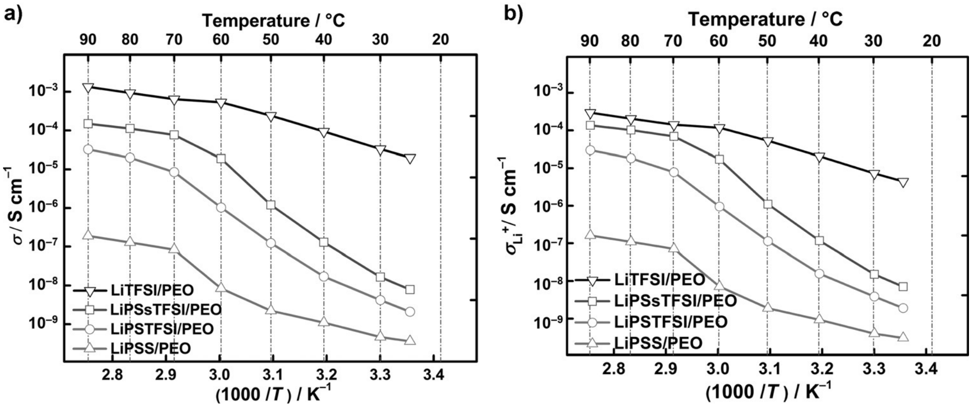

Figure 3. a) The temperature dependence of ionic conductivities for the LiX/PEO (X = PSS, PSTFSI, PSsTFSI, TFSI) blended polymer electrolytes ($\mathrm{EO/Li^+} = 20$). b) The temperature dependence of ionic conductivities of $\mathrm{Li^+}$ cations for the LiX/PEO (X = PSS, PSTFSI, PSsTFSI, TFSI) blended polymer electrolytes ($\mathrm{EO/Li^+} = 20$).

Among these three blended electrolytes containing polymeric anions, the complex of LiPSsTFSI/PEO displays the highest conductivities, and is higher by about one order in magnitude than those of the previous state-of-the-art LiPSTFSI/PEO electrolyte, and higher by 2–3 orders in magnitude than those of LiPSS/PEO electrolyte in the whole temperature range. This would be essentially attributed to more flexible and super-delocalized nature of the $-SO_2-N^{(-)}-SO(=NSO_2CF_3)-CF_3$ structure (in $PSsTFSI^-$) compared with both the $-SO_2-N^{(-)}-SO_2-CF_3$ (in $PSTFSI^-$) and $-SO_3^-$ structures (in PSS^-), which can not only further improve polymeric segmental motions responsible for promoting Li-ion motion through the chains of PEO in amorphous phase, but also increase degree of dissociation of the Li salts. This anion-dependence trend in conductivity concurs with the results in DSC and XRD measurements (Figures 1 b and 2), wherein a higher degree of amorphous phase is observed in the complex of LiPSsTFSI/PEO.

The lithium-ion transference number (t_{Li^+}) is a key parameter for evaluating the performance of SPEs. The value of t_{Li^+} for the classic ambipolar LiTFSI/PEO blended electrolyte is 0.22 at 60 °C, which is comparable to that of 0.29 reported previously.[30] The value of t_{Li^+} for the complex of LiPSsTFSI/PEO is as high as 0.91 at 60 °C, indicative of single Li-ion conducting behavior, which is expected from the huge volume of the polyanion (Supporting Information, Tables S4 and Figure S7).

The ionic conductivities of individual Li^+ cations in SPEs play a paramount role in dictating the performance of Li batteries, as Li^+ cations are only active species needed during the cycling process of Li batteries; however, few work have characterized the conduction behaviors of individual Li^+ cations. Figure 3 b comparatively displays the temperature dependence of ionic conductivities of individual Li^+ cations (σ_{Li^+}, $\sigma_{\mathrm{Li}^+} = \sigma \times t_{\mathrm{Li}^+}$) for the four LiX/PEO (X = PPS, PSTFSI, PSsTFSI, TFSI) blended polymer electrolytes. It is important to note that the values of σ_{Li^+} for the LiPSsTFSI/PEO blended electrolyte are comparable to those for the classic ambipolar LiTFSI/PEO electrolyte above the melting of PEO (70 °C; for example, at 70 °C, $\sigma_{\mathrm{Li}^+} = 6.92 \times 10^{-5}\,\mathrm{S\,cm^{-1}}$ (LiPSsTFSI/PEO) vs. $\sigma_{\mathrm{Li}^+} = 1.41 \times 10^{-4}\,\mathrm{S\,cm^{-1}}$ (LiTFSI/PEO); at 90 °C, $\sigma_{\mathrm{Li}^+} = 1.35 \times 10^{-4}\,\mathrm{S\,cm^{-1}}$ (LiPSsTFSI/PEO) vs. $\sigma_{\mathrm{Li}^+} = 2.94 \times 10^{-4}\,\mathrm{S\,cm^{-1}}$ (LiTFSI/PEO); Supporting Information, Table S5). This is inspiring as the PEO-based SPEs works above its melting point in practical applications, and indicates that LiPSsTFSI/PEO blended electrolyte would have better electrochemical performance than LiTFSI/PEO electrolyte above its melting point, as the concentration gradients of the salt and cell polarization, owing to the transference of small anions, could be removed in the SLICs of LiPSsTFSI/PEO. Among these SLICs, the highest the values of σ_{Li^+} for the complex of LiPSsTFSI/PEO are observed in the testing temperature range.

In summary, a new type of lithium polymer salt (LiPSsTFSI) has been synthesized by free radical polymerization. The neat LiPSsTFSI ionomer displays a low glass transition temperature of 44.3 °C. Its blended polymer electrolyte of the LiPSsTFSI/PEO type exhibits single Li-ion conducting behavior with a t_{Li^+} value as high as 0.91 and an oxidation potential at ca. 4.0 V vs. Li^+/Li, and is thermally stable up to 300 °C. The ionic conductivities of the LiPSsTFSI/PEO ($EO/Li^+ = 20$) blended electrolyte are higher by about one order in magnitude than those of the LiPSTFSI/PEO ($EO/Li^+ = 20$) electrolyte, and by 2–3 orders in magnitude than those of LiPSS/PEO ($EO/Li^+ = 20$) electrolyte over the whole temperature range. This is essentially attributed to the unique structure of $-SO_2-N^{(-)}-SO(=NSO_2CF_3)-CF_3$ in the $PSsTFSI^-$ anion with both super-delocalized negative charge distribution and highly flexible features, which not only makes highly dissociation of Li^+ ions from anions but also favorably enhances the degrees of amorphous phases and the segmental motions in SPEs required for ionic conduction. More importantly, the ionic conductivities of individual Li^+ cations for the LiPSsTFSI/PEO blended electrolyte are comparable to those for the classic ambipolar LiTFSI/PEO electrolyte above the melting point of PEO. All of these preliminary results suggest that the LiPSsTFSI would be a promising salt of polymer electrolyte and would open the door to design new highly conductive single lithium-ion conductors for lithium batteries.

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