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Linear and Crosslinked Ionic Liquid Polymers as Binders in Lithium-Sulfur Battery

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Abstract

A collection of different polymeric ionic liquids (PILs) were explored as cathode binders in lithium-sulfur batteries. The PIL molecular structure, polymer backbone, and polymer architecture were found to influence cell capacity, cyclability, and the morphology of the cathode itself. PILs with styrene backbones performed better than vinyl-based polymer, while crosslinked PILs imparted further improved capacities, cyclability, and reduced over-potentials. Unlike PVdF, PIL binders mixed with the sulfide species, resulting in more uniformly distributed sulfides in the cathode and better sulfide transport. These features helped to mitigate volume change-induced degradation that typically plagues Li-S batteries. The uptake of polysulfides by PILs also constrains the polysulfide shuttle during battery cycling, leading to better cycling stability. While traditionally binders are viewed only as a “glue” to hold active material together, PIL binders have additional functions and play an active role during Li-S working operation.
**Introduction**

High performance lithium-sulfur (Li-S) batteries that are lightweight and energy dense are contenders to replace current Li-ion technology.\(^1\) While the potential energy storage of Li-S batteries is quite high, low cyclability arising from polysulfide migration to the anode, and/or volume changes at the cathode, significantly reduces the lifetime of such devices. For these reasons, researchers have sought to develop new electrolytes,\(^2\) membranes,\(^5,6\) cathodes,\(^7,8\) and sulfur impregnation techniques\(^8\) to overcome these persistent issues. In general, the role of binders in the operation of the battery has garnered less attention, despite comprising a significant portion of the total weight of the cell (5-10 wt%).\(^9\) This is in light of the fact that the binder is responsible for "gluing" the components during large volume changes. Even small improvements to the overall performance of the battery can, on an industrial scale, lead to large energy savings. Polyvinylidene fluoride (PVdF) is currently the standard for lithium battery technology and is routinely used in lithium-ion and lithium-sulfur batteries for its chemical inertness and availability. Despite a number of its clear issues, such as delamination from the active components, it has thus far been sufficient for the development of new battery systems. One issue that PVdF does not address is the 80% volume expansion that occurs upon formation of Li\(_2\)S. This volume change disrupts electrical contacts within the cathode which in turn reduces cyclability.\(^13\) An improved binder that can mitigate these issues could help to accelerate the development of Li-S batteries as a practical alternative to current technologies. One class of polymers that have recently garnered attention are polymeric ionic liquids (PILs), whose high electrochemical stability, conductivity, and good processability is well suited for electrochemical applications.\(^14\) They have been incorporated in supercapacitors,\(^15\) electrochemical sensors,\(^16,17\) and a variety of energy devices\(^18,19\) with good success. Recently it was found that their high stability was advantageous for use as a binder in lithium-ion batteries.\(^20,21\) Linear PILs were capable of wrapping the powder components while still allowing for Li\(^+\) to flow, thus improving cyclability.\(^20\) Nanoparticle PILs were found to operate according to a different mechanism by allowing electrolyte to permeate between the free spaces.\(^21\) During cycling, these nanoparticles would maintain contacts between the conductive components while permitting the flow of electrolyte. In comparison, only a few studies have been conducted for the use of PILs in lithium-sulfur batteries.\(^22\) While molecular interactions between ionic-liquids and polysulfide species have been examined in great detail,\(^25\) the role of a
cationic polymer binder in similar environments is not well understood. There is strong
evidence by Helms et al. for such polymers to facilitate lithium-ion transport and restrict
sulfide diffusion simultaneously, which in turn leads to improved cyclability. However, no
study has been conducted to compare the effect of different cationic polymers or PILs. In
this context, we examined five PIL binders, either as linear or crosslinked nanoparticles for
their use in lithium-sulfur batteries. We found that in all cases, the PILs were generally better
than PVdF with respect to cyclability, with some PILs displaying higher discharge capacities in
extended cycling experiments. We found that this improvement is due to the compatibility
between the PIL and the produced sulfides, which inhibits swelling-induced degradation and
retention of the sulfides within the cathode. We postulate on the nature of the PIL in these
batteries and demonstrate a potential route for further PIL binder exploration.

Experimental

Materials

Azobisisobutyronitrile (AIBN) was purchased from Wake Chemicals and used as received.
Polyvinylidene fluoride (PVdF, average M<sub>w</sub> 534000, CAS 24937-79-9, Sigma-Aldrich) and
Printex XE2 (Degussa) were used for the electrode preparation. 1 M LiTFSI in TEGME:DOL
(purity 99.9%, E057, Solvionic) was used as electrolyte. Lithium foil (FMC, 500 µm) was used
as anode.

Synthesis

Linear binders PIL1-3 and crosslinked nanoparticle binder PIL5 were synthesized according to
literature procedures. Crosslinked nanoparticle PIL4 was synthesized from the TFSI based
monomer in methanol by precipitation polymerization. Briefly, monomer (7.32 g, 14.90 mmol) was dissolved in methanol (50 mL) containing 1.5 wt% AIBN (0.108 g, 0.657 mmol) and stirred for 18 h at 60 °C. The precipitate was then rinsed with methanol (4 x 100 mL) to remove excess monomer and dried in-vacuo (Scheme S1).

Electrochemistry

Carbon (Ensaco 350G, Imerys) was ball milled for 30 min at 300 rpm with sulfur in a mass
ratio of 1:2. The mixture was heated to 155 °C with a heating ramp of 0.2 C min<sup>−1</sup>, where it
was kept for 5 h. The cooling down period to room temperature was performed at a rate of
0.5 C min\(^{-1}\). The electrodes were prepared by mixing the sulfur/carbon composite (66 wt.% of sulfur), the binder (PVdF or PIL1-5), and conductive carbon black additive (Printex XE2, Degussa) in a mass ratio of 80:10:10. The slurry was prepared in N-methyl-2-pyrrolidine (NMP) and casted on a carbon coated aluminum foil. The typical sulfur loading on carbon coated aluminum foil was approximately 1.5 mgS cm\(^{-2}\). A pouch type two electrode cell was prepared inside an argon filled glovebox. The sulfur cathode (2 cm\(^2\) electrode) was separated from the metallic lithium anode with Celgard 2400 separator. The electrolyte 1 M LiTFSI in TEGDME:DOL (vol. 1:1) quantity was normalized to 15 µL mg\(^{-1}\) of sulfur. The batteries were cycled in the potential range between 1.5 V to 3.0 V by using Bio-logic VMP3 galvanostat/potentiostat at a current density of C/5 (334.4 mA g\(^{-1}\)).

**X-ray photoelectron spectroscopy (XPS)**

XPS measurements were performed using a Thermo Scientific K-Alpha+ X-ray Photoelectron Spectrometer. All samples were analyzed using a microfocused, monochromated Al K\(\alpha\) X-ray source (1486.68 eV; 400 μm spot size). The K-Alpha+ charge compensation system was employed during analysis to prevent any localized charge buildup. The samples were mounted on conductive carbon tape the resulting spectra analyzed using the Avantage software from Thermo Scientific.

**Polysulfide uptake**

Inside an argon filled glove box, 100 mg of each binder (PVdF and PIL4) were weight into two separate vial flask. 500 µL of 0.05 M Li\(_2\)S\(_2\) in 1 M LiTFSI in TEGDME:DOL solution was added to each binder containing vial flask. The solution was left inside the glove box for 24 h.

**Results and discussion**

In order to determine the effect of different PIL binders on the performance of Li-S batteries, three different linear polymers (PIL1-PIL3) and two crosslinked polymers (PIL4 and PIL5) were prepared (Figure 1). PIL1 and PIL2 possess a vinyl or styrenic backbone respectively, while PIL3 is a styrenic PIL with a methyl protected substituent at its C2 position. We also examined two crosslinked nanoparticle systems, PIL4 and PIL5, which possess both different molecular structures and a different number of polymerizable groups. These differences manifest in their size and morphology upon dispersion polymerization, with PIL5 having an
average diameter of 10-30 nm, and PIL4 a diameter of ~20 nm and comprised of smaller particles (~5 nm in diameter) (Figure S1).

Figure 1: PIL binders examined for use in Li-S systems.

The electrochemical performance of the PIL1-5 and PVdF as binders was tested in a Li-S battery at C/5 (334.4 mA g⁻¹) current density (Figure 2). PVdF is a well-known inert binder and possesses no electronic or ionic conductivity. The cell with PVdF achieved an initial discharge of 847 mAh g⁻¹, which faded to 521 mAh g⁻¹ and to 355 mAh g⁻¹ after 200 and 500 cycles respectively, with a Coulombic efficiency of approximately 83%. Figure 2 shows that PIL binders influence discharge capacities and the rate of capacity fading of the cell. PIL1 exhibits a lower discharge capacity in comparison to PVdF, however after prolonged discharge-charge cycling, severe capacity degradation was observed. This is not surprising as similar instabilities for PIL1 was observed in Li-ion battery and is a result of side reactions on the ethyl substituent. Cells with PIL2 had a lower initial capacity of 482 mAh g⁻¹ and 494 mAh g⁻¹ in the 1st and 50th cycle, respectively. However after 200 cycles, an increase of 16% of the initial capacity was observed (574 mAh g⁻¹), while after 500 cycles, a discharge capacity of 473 mAh g⁻¹ was observed. The cells with binders PIL3-5 showed an increase in capacities in the initial cycles, with cells containing binder PIL3 reaching the highest discharge capacity after 11 cycles followed by cycling behaviour similar to PVdF. The similar performances of PIL2 and PIL3 indicates that methyl protection at the C2 position is not particularly important, unlike in Li-ion batteries which benefits from C2 protection. This is likely due to the lower operating voltages in Li-S vs. Li-ion batteries and indicates that
greater tolerances for PIL structures is possible, allowing for a greater number of PILs to be examined for these applications. The Li-S cell with PIL4 attained the highest discharge capacity of 1015 mAh g\(^{-1}\) after 3 cycles and was the highest among the binders tested. The discharge capacity of the cell with PIL4 slowly faded to 657 mAh g\(^{-1}\) and to 446 mAh g\(^{-1}\) after 200 and 500 cycles, respectively. The cell with PIL5 had the lowest initial discharge capacity, however an increase of 10% was observed after 200 cycles with stable cycling.

Figure 2: a) Discharge capacity and b) coulombic efficiency for PVdF and PIL1-5 in Li-S battery at C/5 current density.

Figure 3 represents the discharge and charge voltage profiles of the Li-S batteries with PVdF and PIL1-5. In all tested samples, two Li-S characteristic plateaus were observed; the high and the low-voltage plateau.
Figure 3. Galvanostatic discharge/charge voltage profiles for the Li-S cells with a) PVdF binder, b) PIL1 binder, c) PIL2 binder, d) PIL3 binder, e) PIL4 binder and f) PIL5 binder at C/5 (334.4 mA g\textsuperscript{–1}) current density.

To facilitate the interpretation of the involved complex reactions, the voltage profile was divided into three sections, full discharge (denoted as $Q_{\text{disch}}$), high-voltage ($Q_{\text{high}}$) and low-voltage ($Q_{\text{low}}$) plateau. The high-voltage plateau (denoted as $Q_{\text{high}}$), from 2.5 V to 1.9 V correlates to a sulfur reduction into long- and mid-chain polysulfides. The low-voltage plateau (denoted as $Q_{\text{low}}$), from 1.9 V to 1.5 V correlates to the equilibrium between polysulfides and precipitated Li$_2$S.\textsuperscript{29–31} Similar to what Peled et. al. have reported,\textsuperscript{29} the binder influences both $Q_{\text{high}}$, $Q_{\text{low}}$, and the cell over-potential (Table 1). In the case of PVdF the $Q_{\text{high}}/Q_{\text{disch}}$ ratio is approximately 30% and does not change from the 50\textsuperscript{th} to the 200\textsuperscript{th} cycle. The constant $Q_{\text{high}}/Q_{\text{disch}}$ ratio shows that the capacity degradation is the same for the high-voltage and low-voltage plateau, which correlates with loss of sulfur. For PIL4 the $Q_{\text{high}}/Q_{\text{disch}}$ ratio increases from the 50\textsuperscript{th} to 200\textsuperscript{th} cycle. This may indicate that sulfur reduction to Li$_2$S\textsubscript{4} is more efficient with prolonged discharge-charge cycling. Interestingly, with PIL2 and PIL5, the $Q_{\text{high}}/Q_{\text{disch}}$ ratio is higher in comparison to the other binders, and is around half of $Q_{\text{disch}}$. However, from the 50\textsuperscript{th} to the 200\textsuperscript{th} cycle the $Q_{\text{high}}/Q_{\text{disch}}$ ratio decreases to 40%. Similar behavior is observed in the obtained discharge capacities for PIL2 and PIL5 (Figure 2), where the capacity increases during prolonged cycling. The small initial $Q_{\text{low}}/Q_{\text{disch}}$ ratio and its gradual increase with cycling is due to the less effective initial reduction from Li$_2$S\textsubscript{4} to Li$_2$S, which is an indication of an initial kinetics limitation. Furthermore, PIL2 and PIL5 have the highest over-potential. The similar capacity fading after 200 cycles between the PVdF and PILs binders indicates that similar degradation mechanisms occur. However, the fading with PILs is slower due to its favourable interaction with the polysulfide species.

Table 1. Effect of the cycling and binders on $Q_{\text{disch}}$, $Q_{\text{high}}/Q_{\text{disch}}$, $Q_{\text{low}}/Q_{\text{disch}}$ ratio and cell over-potential.

<table>
<thead>
<tr>
<th>Binder</th>
<th>$Q_{\text{disch}}$ [mAh g\textsuperscript{–1}]</th>
<th>$Q_{\text{high}}/Q_{\text{disch}}$ [%]</th>
<th>$Q_{\text{low}}/Q_{\text{disch}}$ [%]</th>
<th>Pol. [mV]</th>
<th>$Q_{\text{disch}}$ [mAh g\textsuperscript{–1}]</th>
<th>$Q_{\text{high}}/Q_{\text{disch}}$ [%]</th>
<th>$Q_{\text{low}}/Q_{\text{disch}}$ [%]</th>
<th>$\eta_{\text{tot}}$ [mV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVdF</td>
<td>644</td>
<td>34</td>
<td>66</td>
<td>146</td>
<td>521</td>
<td>35</td>
<td>65</td>
<td>152</td>
</tr>
</tbody>
</table>
To study the effect of the binders on the cathode morphology, *post mortem* SEM analysis was performed on disassembled cathodes (Figure S2). SEM images of freshly discharged (1.5 V) and charged (3 V) cathodes containing PVdF, PIL2, and PIL4 are shown in Figure 4. In the case of the fresh cathode with PVdF binder, a porous cathode structure was observed with interconnected sulfur-impregnated carbon particles. The discharged cathode with PVdF shows that the pores are blocked due to the crystallization of Li$_2$S on the cathode surface. When charged, the open porosity is re-established due to Li$_2$S consumption and the reformation of sulfur. The fresh electrode with binder PIL2 is similar to the cathode with PVdF binder, while the fresh cathode containing binder PIL4 possessed a polymeric film of interconnected sulfur-impregnated carbon particles on the surface. SEM images of the discharged cathodes with PIL2 and PIL4 were similar to each other with a “swelled particle” structure containing open pores. This is in stark contrast to the discharged cathode containing PVdF binder, which appears to have filled pores and no “swelling”. This indicates that the growth of Li$_2$S is likely different in the presence of PIL vs PVdF and that the improved cycling is derived from this interaction. According to Welton *et al.*, dissolution of ionic solutes in ILs takes place via metathesis reactions, thus resulting in an intimate mixing of the two ionic components. The observed differences in the SEM between the discharged PVdF and PIL samples can be explained by the uniform mixing and retention of sulfide species with PIL binder (in the form of ion-exchange reactions), which then influences Li$_2$S growth and battery cycling. The fluorophilic character of PVdF does not allow any of these processes to occur and thus leads to poorer cycling stability.
Figure 4: SEM micrographs higher magnification top-view for fresh, discharged at 1.5 V and charged at 3.0 V sulfur cathodes with a) to c) PVdF binder, d) to f) PIL2 and g) to j) PIL4.

Analysis of the cathodes by X-ray photoelectron spectroscopy (XPS) S 2p spectra (Figure 5) revealed that the morphology differences observed in the SEM are not met by differences in their surface chemical composition.
Figure 5: XPS S 2p spectra of the composite cathode with a) PVdF in the 1\textsuperscript{st} discharge, b) PVdF in the 1\textsuperscript{st} charge, c) PIL2 in the 1\textsuperscript{st} discharge, d) PIL2 in the 1\textsuperscript{st} charge, e) PIL4 in the 1\textsuperscript{st} discharge and f) PIL4 in the 1\textsuperscript{st} charge.

In all cases, four different components were observed in the discharged samples. They include Li\textsubscript{2}S (around 160.5 eV), terminal (around 162 eV) and bridging (around 163.6 eV) sulfur, and the decomposition products S(IV) from TFSI salts.\textsuperscript{6} As well, the TFSI anion was only observed in the charged cathodes. Thus, the stark differences between PVdF vs PIL in the charged cathodes is a result of how the respective binders affect the growth of the produced sulfides. The perfluorinated structure of PVdF prefers not to mix with the sulfide salts, resulting in sulfide precipitation on top and in between the binder/carbon interstitial spaces (Figure 6). This is supported by the dramatically low solubility of lithium sulfides in fluorinated solvents, which applies to the interaction between PVdF and the lithium sulfides.\textsuperscript{24,33,34} PILs however have shown the ability to uptake a variety of anions, direct the growth of crystalline compounds like bismuth sulfide, and generally promote the stabilization of interfaces for nanoparticle dispersions.\textsuperscript{35–38} The more favourable interactions between the produced lithium sulfides and PIL results in a mixing of these two components, a more even distribution and volume expansion within the cathode, and thus more stable cyclability (Figure 6, bottom right). This is supported by the XPS S2p spectra (Figure 5) of the cathodes in charge state. On the surface of the cathode with PVdF binder, mostly LiTFSI salts and a mixture of short- and mid-chain polysulfides are present, due to dissolution and
shuttling of long-chain polysulfides. On the surface of the cathode with PILs binders, mostly long-chain polysulfides are found. These results support the interaction and uptake of polysulfides by PILs. Furthermore, we assume it is for this reason that the carbon particles appear more swelled in the SEM images for **PIL2** and **PIL4** in comparison to PVdF. It was also noticed that lithium sulfide sublimation occurred in the SEM vacuum chamber for the discharged cathode containing PVdF and not for PIL, indicating that the lithium sulfides are strongly bound within/around the PIL. This effect does not appear to be different for **PIL2** and **PIL4** despite the former being a soluble homopolymer and the latter a nanoparticle binder. In contrast to this, high over-potentials were observed for **PIL2** and **PIL5** and not for **PIL4**. In the case for **PIL2**, easier transport of electrolyte between the interstitial spaces for the particle system which is not possible for the linear homopolymers (**Figure 6**, left). For **PIL5** the kinetics of Li$_2$S growth was hampered (**Figure 3**) and may stem from the more crosslinked and unswellable structure, preventing mixing with produced sulfides.

**Figure 6**: Schematic representation of the sulfide precipitation around and in between the binder/carbon for PVdF and PIL binders.

To test the swelling and the polysulfide uptake of the polymers, TEGDME:DOL solutions containing 0.05 M Li$_2$S$_2$ and 1 M LiTFSI TEGDME:DOL were mixed with PVdF and **PIL4** (**Figure S3**). When the green solution was mixed with the PVdF dispersion, the colour of the solution remained unchanged after 24 hr (**Figure S3a**). When the polysulfide solution was mixed with a dispersion of **PIL4**, the PIL noticeably swelled and the colour of the solution
changed from green to orange (Figure S3b). The change of colour from green to orange suggests a chemical reaction between the PIL4 and the Li$_2$S$_2$, meaning PIL4 acts as a catalyst for the disproportionation reaction of Li$_2$S$_2$\textsuperscript{40,41} as presented on the followed reaction:

$$3S_{2}^{2-} \leftrightarrow S_{4}^{2-} + 2S_{2}^{2-}$$

After 24 hr PIL4 was swelled and absorbed the polysulfide solution, forming a white gel. The ability for the particles to reversibly swell and deswell allows for their use as “sulfide reservoirs” during cycling. This feature combined with their effective electrolyte transport demonstrates an improved approach for PIL utilization in lithium-sulfur battery systems.

In Li-S batteries, the role of the PIL binder is not only to “glue” the active material together, but to also play an active role during the cycling. They efficiently improve the reduction of sulfur into lithium polysulfides and improve the Li$_2$S redistribution over the carbon surface. Furthermore, PIL-based binders can reduce volume change-induced stress by uptaking/releasing lithium polysulfides during swelling and deswelling cycles.

**Conclusions**

In conclusion, five different PILs were used as binders in lithium-sulfur batteries and compared to conventional PVdF. The PIL binders possessed different chemical structures, polymer backbones, and different polymeric architectures, including linear homopolymers and nanoparticles. We found that in all cases, PILs were equal to or superior than PVdF with respect to cyclability, with some PILs displaying higher discharge capacities across the tested lifetime. The PIL binders improved the reduction of sulfur into Li$_2$S$_4$ more efficiently while simultaneously improved Li$_2$S redistribution in the cathode, which boosted battery cyclability. We can conclude that the key to this increased performance is a result of the favourable interactions between the PIL and the produced lithium sulfide species, resulting in mixing of the two and a more even volume expansion during discharge. This is in contrast to PVdF, which does not mix with the lithium sulfide species, resulting in uneven volume expansion between the carbon particles. Such added stress within the cathode results in shorter lifetime with respect to their PIL counterparts. Finally, we show that PIL nanoparticles lower the resistance of the cell in comparison to PIL homopolymers by promoting electrolyte flow between the carbon particles during cycling. This is a result of their small size, high surface area, swellability for the produced lithium-sulfide species, and improved electrolyte transport through the interstitial spaces. Furthermore, the PIL
nanoparticles promoted the disproportionation reaction of the low soluble short-chain polysulfides into long-chain polysulfides, which has a beneficial influence on the achieved capacities. These findings illustrate the positive effects of using PILs in lithium-sulfur batteries and will aid the search for optimized binder designs. Future work will also examine how Li$_2$S crystal growth is influenced by the presence of PIL.

ASSOCIATED CONTENT

Supporting information

The Supporting Information is available free of charge on the ACS Publications website at DOI: XXXX

Contains a synthetic scheme, SEM images, and comparison photos of the swelled binders.

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