

Poly(Ethylene Glycol-*block*-2-Ethyl-2-Oxazoline) as Cathode Binder in Lithium-Sulfur Batteries

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Functional binders constitute a strategy to overcome several challenges that lithium-sulfur (Li-S) batteries are facing due to soluble reaction intermediates in the positive electrode. Poly(ethylene oxide) (PEO) and poly(vinylpyrrolidone) (PVP) are in this context a previously well-explored binder mixture. Their ether and amide groups possess affinity to the dissolved sulfur species, which enhances the sulfur utilization and mitigates the parasitic redox shuttle. However, the immiscibility of PEO and PVP is a concern for electrode stability. Copolymers comprising

ether and amide groups are thus promising candidates to improve the stability the system. Here, a series of poly(ethylene glycol-*block*-2-ethyl-2-oxazoline) with various block lengths is synthesized and explored as binders in S/C composite electrodes in Li-S cells. While the electrochemical analyses show that although the sulfur utilization and capacity retention of the tested electrodes are similar, the integrity of the as-cast electrodes can play a key role for power capability.

1. Introduction

Lithium-sulfur (Li-S) batteries has been considered as one of the electrochemical energy storage systems of the post-Li-ion generation. Its high theoretical gravimetric energy density (2552 Whkg⁻¹)^[1] and the low price of sulfur as a byproduct of the oil industry enable it to stand out from other candidates. However, several fundamental differences between Li-S and Li-ion batteries complicate the application of the former. Besides the low efficiency and non-uniform reaction of metallic Li on the negative electrode, the insulating end products and soluble reaction intermediates of the positive electrode make the Li-S reaction chemistry very distinct from that of the commercial graphite-metal oxide systems.^[2,3] The insulating nature of fully charged and discharged states (S and Li₂S, respectively) requires an electronically conductive host to facilitate the electrochemical reactions. The reaction intermediates (Li₂S_x, x=2–6, also referred to as polysulfides) are soluble and hence mobile in the electrolyte, and can thereby diffuse out of the positive electrode and give rise to the well-recognized “redox shuttle”, which results in poor Coulombic efficiency (CE) and self-discharge.^[4–6]

To tackle both of these issues, micro- and/or mesoporous carbon materials are commonly chosen to serve as both the battery “current collector” and polysulfide confinement.^[2,7] Nevertheless, porous carbon materials alone do not alleviate the fast decay of discharge capacity and CE of Li-S cells to an acceptable level, i.e. the expected cycle life of Li-ion cells. Thus, a broad range of additional solutions have been reported to prevent the loss of active material and/or conductive surface area on the S/C composite electrode. Strategies include, for example, modification of the separator,^[8] addition of interlayers,^[9,10] modification of the carbon host,^[11] metal oxide host materials,^[12,13] electrolytes with very little polysulfide solubility,^[14,15] and employment of functional binders.^[16,17]

Among the above-mentioned options, manipulating the functional groups on the polymer binders constitute an attractive approach, since binders are a necessary but otherwise inactive part of composite electrodes. For Li-ion systems, the binder has primarily one task-maintaining the structural integrity of the composite electrode by fixing the active materials and conductive additives together and onto the current collector throughout the lifetime of the cell.^[16] While this is also the case for typical sulfur-based electrodes, considerations unique to the Li-S chemistry have motivated efforts to introduce polar functional groups that have an affinity to polysulfides.^[16] For example, ether groups on poly(ethylene oxide) (PEO)^[18] and amide groups on poly(vinylpyrrolidone) (PVP)^[19] or poly(2-ethyl-2-oxazoline)^[20] have been demonstrated to increase the discharge capacity and capacity retention in comparison to a binder that does not interact with polysulfides. Binder that swell in the electrolyte also allows better access to the sulfur in the electrochemical reactions. In this context, the popular binder in Li-ion batteries, poly(vinylidene difluoride) (PVdF), is poorly compatible with the Li-S system since PVdF blocks the pores of the porous carbon.^[21] This makes the search for better binder systems for the S/C composite electrode necessary. Strategies for functional binders^[17] tailored to the Li-

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S system include incorporating cationic functional groups,^[22,23] constructing a three-dimensional binder network,^[24,25] etc.

In recent years, our group has developed the concept of a functional binder system based on the combination of polyether and amide functionalities. We previously reported that the combination of PEO and PVP in the binder system could enhance sulfur utilization and coulombic efficiency relative not only to a common water-based reference binder (carboxymethylcellulose sodium salt:styrene butadiene rubber, CMC:SBR), but relative also to either material used alone.^[26] The same effect was later reported also in optimized sulfur electrodes with higher loading and reduced electrolyte volume for both the PEO:PVP binder system as well as related polymers bearing the same functionality.^[27] In that study, the comparatively small differences between the electrodes with the polyether and amide-containing binders indicated that the chemical functionality of the binder had a significantly larger influence on the electrochemical performance than, for example, its mechanical properties.

Despite these performance improvements, there are some known and potential drawbacks with this type of binder concept. Firstly, the previously observed tendency of PEO and PVP mixtures to show limited miscibility and phase separation^[28–30] raises the concern that this could constitute a performance degradation mechanism over time, or lead to inefficient material usage. A further issue related to potential upscaling is our observation, not discussed in the previous studies, that the viscosity of electrode slurries containing PEO:PVP was significantly higher with more porous C/S composites; this was attributed to a preferential absorption of PVP over PEO in the composite.

One solution to address both of these issues is to pursue co-polymers combining both of these functionalities. A copolymer containing both the polyether functionality of PEO and the amide functionality of PVP would avoid both the potential issues of phase separation leading to reduced performance, and preferential absorption leading to difficulty in electrode preparation. Moreover, the development of an effective co-polymer synthesis approach can provide a platform on which to develop new materials with more tailored or targeted functionality. In this work, we thereby report the preparation of a series of poly(ethylene glycol-*block*-2-ethyl-2-oxazoline) polymers with various ratios of polyether and 2-ethyl-2-oxazoline units (referred to in this work as PEO and PEtOx blocks, respectively) as binders of the S/C composite electrodes in Li-S batteries, and explore their functionality in state-of-the-art electrochemical cells. The use of 2-oxazolines as a route to the amide functionality is attractive due to a relatively simple synthesis method as well as possibilities for further functionalization via modification of the 2-oxazoline monomer.^[17]

2. Results and Discussion

In Figure S1 in the Supporting Information, the structures of the synthesized PEtOx and intermediate were confirmed by ¹H NMR and compared with pristine PEG. The intermediate shows two

specific peaks nearby sulfonate that is a stronger electron withdrawing group than single oxygen. The peaks of the six protons (H_c) of the methyl groups appear at 3.1 ppm and proved that the reaction is completed without the residual reagent by calculating their internal areas as compared with 4.3 ppm of four protons (H_d). In the case of PEtOx, the peaks of four protons (H_e) near nitrogen appear at 3.5 ppm closed to PEG's protons (H_a, H_b) and the peaks of ethyl protons (H_g, H_f) appear at 2.3 ppm and 1.1 ppm, respectively.

The Lewis structure and differential thermal calorimetry (DSC) results of the series of copolymers are demonstrated in Figure 1 and 2, respectively. The copolymers are numbered with the molecular weight of the PEO block in the center and the molar ratio of PEtOx and PEO blocks, e.g. PO1k-212 means the PEO block with a molecular weight of 1000 is sandwiched by two PEtOx blocks with twice as many repeating units as the PEO block. It can be observed that the endothermic peak around 50 °C, corresponding to the melting point of PEO which for the macromolecular homopolymer is found around 60 °C,^[31] is only present when the molecular weight of the PEO block is higher than 4000. Below this molecular weight, the PEO domains are likely too small to crystallize properly. Comparing PO4k-212 and PO6k-212, the peak shifts towards higher temperatures as the length of the PEO block increases and the PEO domains are allowed to grow. A similar shift in melting point is also observed when the relative length of PEtOx blocks increases, as manifested by comparing PO6k-212 and PO6k-414, most likely due to an increased phase separation. On the other

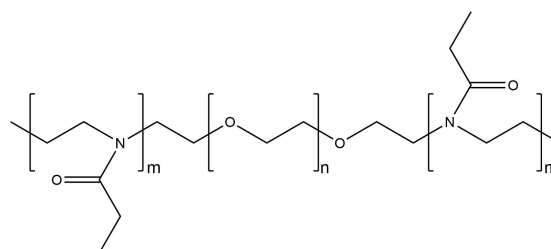


Figure 1. Lewis structure of poly(ethylene glycol-*block*-2-ethyl-2-oxazoline). *m* and *n* denotes the ratio of the number of the 2-oxazoline and ethylene oxide units, being either 2:1:2 or 4:1:4 in the synthesized series of block-copolymers.

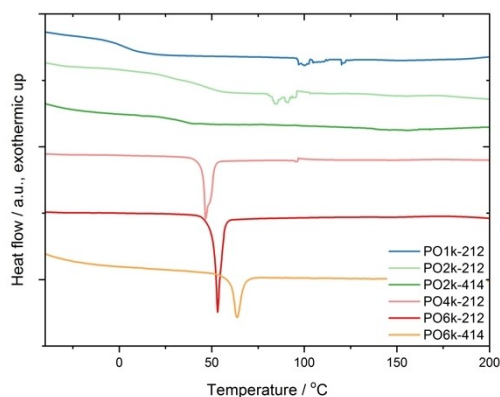


Figure 2. DSC results of the synthesized copolymers

hand, the samples with shorter PEO blocks (1000 and 2000 in molecular weight) merely display a glass transition, instead of a melting point. The glass transition temperature ranges from 0 to 30 °C, which are lower than that of poly(2-ethyl-2-oxazoline)^[32] but higher than that of PEO, which indicates good miscibility. The observed T_g increases with the length of both the PEO and the PETox blocks in these largely amorphous polymers. Nevertheless, the melting and glass transition temperatures are unlikely to significantly influence the properties of the copolymers as binders in the final electrodes, since both PEO and PETox blocks are expected to swell in the electrolyte.^[18,20]

The cycling data of the Li-S cells with different copolymers as the cathode binder is displayed in Figure 3. All cells deliver a specific capacity close to 1250 mAh g_S⁻¹ (g_S denotes the mass of sulfur in grams) in the first discharge. Together with the cell with the baseline binder (PEO:PVP, 4:1 in weight), all cells, except for PO1k-212 and PO6k-414, cycle with a stable discharge capacity of 950 mAh g_S⁻¹ up to the 75th cycle. Cells made with PO1k-212 and PO6k-414 exhibit a lower stable discharge capacity of 850 mAh g_S⁻¹, but for 120 and 100 cycles, respectively. While a tradeoff in discharge capacity and cycle life can be observed, the overall performance is rather uniform. This behavior is fully consistent with our previous finding that the functionality present in the binder system generally has a more significant effect on discharge capacity and CE compared to other factors, such as mechanical integrity, deriving from the binder structure.^[27] This behavior is attributed as a consequence of the high 'swellability' or even solubility of polymers bearing these functionalities, which means they can be considered to act as a local electrolyte additive.^[26]

Compared to the specific capacity, the CE is more scattered among cells, as displayed in the top part of Figure 3. The binders with lower molecular weights (PO1k-212 and PO2k-212)

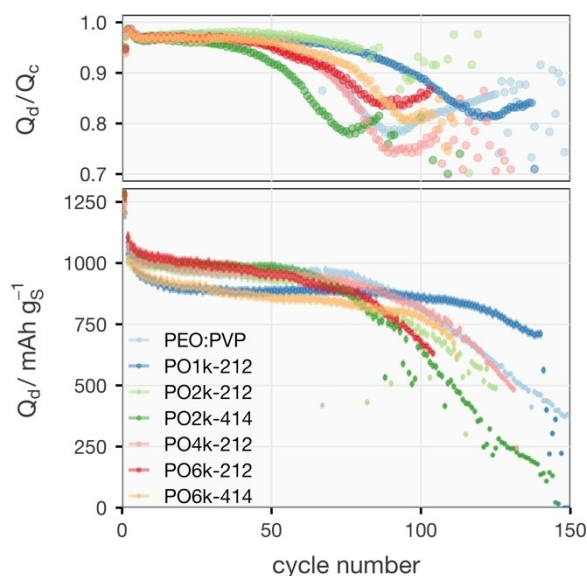


Figure 3. Specific discharge capacity (Q_d) and CE (Q_d/Q_c) of the Li-S cells consisting of S/C composite electrodes made with different binders. Data from the first 75 cycles are shown in an zoomed-in plot in Figure S8.

appears to improve the stability of CE compared to the baseline (PEO:PVP).

However, in the cycling statistics of the repeated cells with PO1k-212, PO2k-212 and PO4k-212 in Figures S1–S3 in the Supporting Information, the variation in CE among the cells with the same binder is rather large after the 50th cycle. On the other hand, the specific discharge capacity, at least for the first 50 cycles, is highly reproducible. Moreover, the resistance profiles in Figures S4–S6 in the Supporting Information, which will be analyzed in greater detail in the following paragraphs, also demonstrate good agreement between the cells with the same binder. The good consistency observed in terms of sulfur utilization and electrode resistance indicates that the relative inconsistency in CE is more influenced by other cell components, in particular the decomposition of the electrolyte by the Li negative electrode, as has been previously reported for this and similar test cell formats.^[6,33–35]

For the 10th, 30th, 50th and 90th cycles, the profiles of the potential (E), internal resistance (R) and diffusional resistance coefficient (k) are plotted against the state of discharge/charge in terms of specific capacity (Q) in Figure 4. The potential profiles of the cells resemble each other, except for those of PO2k-212. This similarity, like the one displayed by the cycling statistics in Figure 3, is less surprising due to the same functional groups on the copolymers. A suppressed lower discharge plateau of the cell with PO2k-212 binder stands out from the other. The associated overpotential can be broken down into two components, R and k, through the application of the ICI method. R corresponds to the sum of electronic, solution and charge transfer resistances,^[36] while k characterizes the transport properties in the porous positive electrode.^[37] In the resistance plots of the 10th discharge, it can be observed that R of PO2k-212 is significantly higher than that of the other cells on the lower plateau ($Q=300\text{--}1000\text{ mAh g}_S^{-1}$) while k is only slightly higher. This substantial difference was reproduced in repeated experiments with this binder, as shown in Figure S6 in the Supporting Information. A noteworthy feature of PO2k-212 is that the coating quality with this binder was observed to be noticeably poorer than the other materials; this may indicate that the poorer electrochemical performance is connected to the quality of the electrode coating, for example through an increased contact resistance, and that this may be an important factor in cycle life.

In the 50th cycle, the lower discharge plateau of the cell PO2k-212 is further deviated from the other cells. This stems from the even higher R and k values on the lower plateau, which indicates the deteriorated transport properties in the porous carbon matrix of the S/C composite electrode. This faster increase in R and k may explain the early onset of the scattering CE of the cell around the 80th cycle, as shown in Figure 4.

In Figure 3, the cell with PO1k-212 as the binder appears to be more stable than the other cells, both in terms of discharge capacity and CE. Therefore, a cell with the same electrode was repeated, whose cycling data are displayed in Figure S2 in the Supporting Information. Despite the identical discharge capacity up to the 60th cycle and the matching voltage and

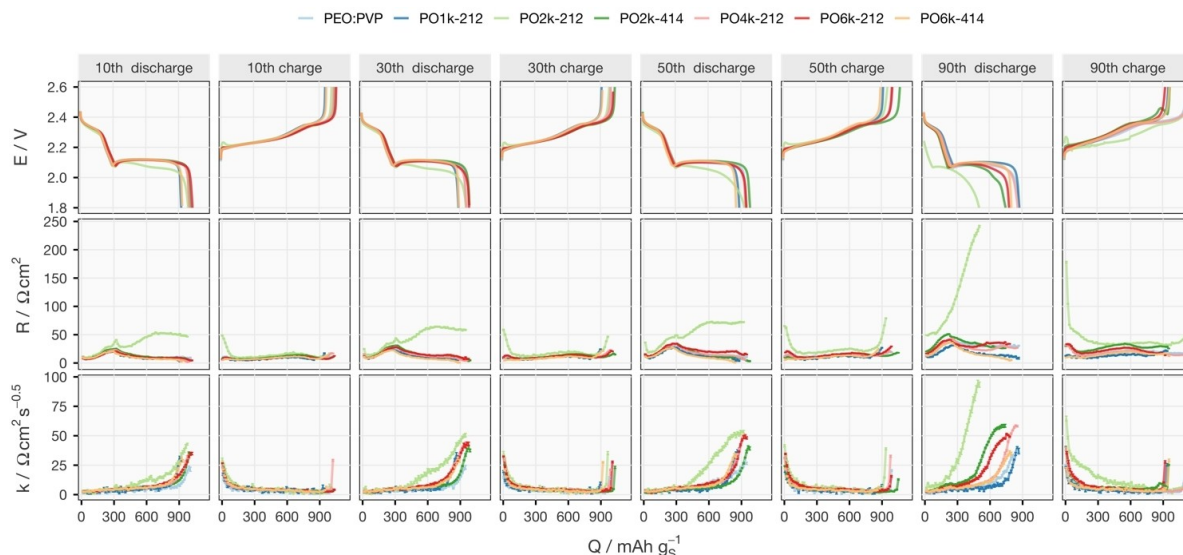


Figure 4. Potential (E), internal resistance (R) and diffusional resistance coefficient (k) of the S/C composite electrodes made with different binders in selected cycles. The experiment was carried out in a two-electrode half-cell but the resistance of metallic Li counter electrode is not negligible after extensive cycling.

resistance profiles in Figure S5 in the Supporting Information, the repeated cell shows much earlier degradation of CE and discharge capacity. The same agreements in the first 50 cycles and deviations afterwards are also observed in other repetitions with the same electrodes in Figures S3–S5 in the Supporting Information. It can thus be concluded that the electrochemical properties after the 50th cycle is dominated by factors other than the binders of the positive electrodes. Given the excessive amount of Li on the negative electrode, the cell setup presented here is by definition a half cell. This allows us to assume that the limiting factor of the performance and thus the main contributor to the resistances are the in this sense comparatively smaller positive electrode. However, the fast degradation of the metallic Li electrode can nullify this assumption.^[38] The CE is especially sensitive to the surface chemistry of metallic Li electrodes in Li-S cells due to its importance in regulating the polysulfide redox shuttle.^[6,35] It is therefore less meaningful to use the measurements after extended cycling when analyzing the influence of the binders in the positive electrodes.

To summarize, the S/C composite electrodes made with the synthesized copolymers perform generally similarly to those made with a mixture of PEO and PVP. A worse electrode integrity is observed from the electrode coating made with PO2k-212, which is manifested by the higher resistances and lower stability of the cells. The electrodes made with PO1k-212 and PO6k-414 have slightly lower specific discharge capacity. The rest of the cells demonstrate almost identical cycling statistics and resistance profiles. A summary of the performance of the cells can be found in Table S1 in the Supporting Information.

3. Conclusion

In this work, a series of poly(ethylene glycol-*block*-2-ethyl-2-oxazoline) with various lengths of both blocks are successfully synthesized, characterized, and utilized as binders in S/C composite electrodes for the Li-S system, thereby introducing positively contributing binder functionalities into the same polymer chain. The electrodes made with this series of block-copolymers show comparable electrochemical properties with each other, as with a mixture of PEO and PVP. This corroborates our previous finding that the mere presence of the functional groups on the binders generally has as a more significant effect on the electrochemical performance, than other properties deriving from their macromolecular structures.^[27] The only significant difference is the higher resistance of the cell made with the binder PO2k-212, which we attribute to the observed poor mechanical integrity of the electrode coating. The results presented here demonstrate that the PEO:PVP combination remains a robust choice as the binder system for S/C composite electrodes, and while their immiscibility does not seem to pose a noticeable impact – neither positive nor negative – on the electrochemical performance of the S/C composite electrodes in Li-S batteries, the possibility to utilize these functionalities in block-copolymers opens up possibilities for improvements in electrode fabrication and new concepts based on further binder optimization and functionalization.

Experimental Section

Materials

For the synthesis of polymer binders, poly(ethylene glycol) (PEG) with molecular weights (M_w) of 1000, 2000, 4000 and 6000 (PO1k, PO2k, PO4k and PO6k, Sigma-Aldrich), methanesulfonyl chloride (Sigma-Aldrich), 2-ethyl-2-oxazoline (Sigma-Aldrich), acetonitrile

(VWR), diethyl ether (VWR), dichloromethane (VWR) were obtained from the suppliers and used without further purification.

For the fabrication of the S/C composite electrodes, sulfur powder (S, Sigma-Aldrich), Ketjenblack (KB, EC-600JD, Akzo Nobel), Super C65 (Imerys), carbon nanofibers (CNF, 20–200 nm × 100 μm, Sigma-Aldrich), carbon-coated aluminum-foil (20 μm thick, SDX, Showa Denko) were used in addition to the synthesized binders. Poly (ethylene oxide) (PEO, $M_w \sim 4000000$, Sigma-Aldrich) and poly (vinylpyrrolidone) (PVP, $M_w \sim 360000$, Sigma-Aldrich) in mass ratio 4:1 were used as the baseline binders^[27] to evaluate the synthesized binders.

The electrochemical characterization of the positive electrodes was carried out in CR2025 coin cells (Hohsen) with the following cell components. Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI, BASF) and lithium nitrate (LiNO₃, Sigma-Aldrich) were dried at 120 °C under vacuum for 12 h. 1,2-Dimethoxyethane (DME, BASF) and 1,3-dioxolane (DOL, Sigma-Aldrich) were dried with 3 Å molecular sieves overnight. Celgard 2400 separators were dried under vacuum at 80 °C for 12 h. Lithium metal foil (Li, Cyprus Foote Mineral, 125 μm thick) was used as received but stored under Ar atmosphere.

Synthesis of Poly (Ethylene Glycol-Block-2-Ethyl-2-Oxazoline)

The copolymers were prepared based on a method reported by Ionescu-Vasii et. al.^[39] First, the initiator for polymerization was prepared by converting the hydroxyl group of PEG to methanesulfonate. In an 100-mL round-flask, PO1k (5 g) was dissolved into dichloromethane (20 mL) and the solution was retained for 10 min at 0 °C before adding to methanesulfonyl chloride (1.16 mL, 15 mmol) and trimethylamine (2.16 mL, 15.5 mmol). The mixture was stirred for 24 h at room temperature. Deionized water (10 mL) was poured into the flask to remove residual reagents. The organic phase was separated and the solvent was evaporated. Then, the white product was washed twice with diethyl ether and dried in a vacuum oven at 40 °C after filtration. The procedure was repeated for 5 g of PO2k, PO4k and PO6k, respectively.

PEG-methanesulfonate (0.5 g) and 2-ethyl-2-oxazoline (2.02 mL, 20 mmol) were dissolved in acetonitrile (10 mL). The mixture was degassed by nitrogen bubbling before it was stirred at 90 °C for 2 days. The solvent was evaporated and the polymer was dissolved in methylene chloride. The product was precipitated in diethyl ether and dried in a vacuum oven at 40 °C. The molar ratio of the 2-oxazoline units to the ethylene oxide units was determined by nuclear magnetic resonance spectroscopy (NMR) to be 2:1:2. PO2k and PO6k were also used for synthesizing copolymers with longer PEtOx chains by increasing the amount of 2-ethyl-2-oxazoline to 10.1 mL. These two products will be referred to as PO2k-414 and PO6k-414 since the number of 2-oxazoline units are doubled relative to the number of ethylene oxide units (i.e. 4:1 as compared to 2:1).

The synthesized copolymers were characterized by DSC. The samples (~10 mg) sealed in Al pans were preheated at 10 °C min⁻¹ to 100 °C, then cooled down at 20 °C min⁻¹ to -80 °C. The measurement was done from -80 to 250 °C at 10 °C min⁻¹.

Preparation of S/C Composite Electrodes

The composition of the electrodes was adopted from a previously optimized recipe.^[27] 65 % S, 21 % KB, 3.5 % C65, 3.5 % CNF, 7 % binder (by weight). S was impregnated into pores of KB by mixing in a mortar and then heated to 155 °C for 20 min. The resulting S/KB composite and other ingredients were weighed as dry powder

and mixed with a solution of 5.5 vol% ethanol in deionized water. The mixture was made into a slurry by 2 h planetary ball-milling, which was tape-cast onto a carbon-coated aluminum foil. The electrodes were cut into Ø13 mm discs and dried under vacuum at 55 °C for 12 h. All positive electrodes had a similar thickness and sulfur-loading between 2.01 and 2.44 mg_s cm⁻².

Electrochemical Characterization

The electrodes were tested in a CR2025 coin cell format with a Celgard 2400 separator (Ø17 mm) and an Li disc (Ø15 mm) as the counter electrode. In each coin cell, 6 μL mg_s⁻¹ of 1 m LiTFSI 0.25 m LiNO₃ DME:DOL (1:1, v:v) was added with an automatic micropipette as the electrolyte. The cells were rested for 6 h before discharged at C/50 (1 C = 1672 mA g_s⁻¹) to 1.9 V and charged at C/25 to 2.6 V. After the formation cycle, the cell was cycled at C/10 between 1.8 and 2.6 V with a 1 s current pause every 5 min for the resistance measurement by the Intermittent Current Interruption (ICI) method.^[36,37] Each discharge and charge step was limited to 10 h (time required to discharge/charge a cell with the theoretical capacity) to stop infinite discharging/charging (mostly charging) cause by polysulfide shuttling. The cells were tested on an Arbin BT-2043 battery testing system. The internal and diffusional resistances were derived from the electrochemical data with the ICI method^[36,37] using the R programming environment.^[40] The raw data and the R scripts used for the analysis are available online via Zenodo.^[41]

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: binder · block-copolymer · lithium-sulfur batteries · polyethylene · polyoxazoline

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