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Functionalized polypropylene separator coated with polyether/polyester blend for high-performance lithium metal batteries

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Abstract

Commercial polyolefin separators used in lithium metal batteries (LMBs) have the disadvantages of insufficient thermal stability and poor wettability with electrolytes, which causes bad safety and battery performance. Poly(ϵ -caprolactone) (PCL)-based electrolytes have drawn widespread attention in the field of polymer electrolytes owing to their electrochemical stability and high lithium-ion transference number. This work proposes a strategy of functionalizing commercial polypropylene (PP) separator coated by blending PCL ($M_w \sim 50,000$) and poly(ethylene oxide) (PEO, $M_v \sim 600,000$). Compared to commercial PP separators, PP-blended PEO_{60w}/PCL_{5w} separators possess better wettability with electrolytes and electrochemical performances. The initial discharge specific capacity of LiFePO₄-based LMBs assembled with PP-blended PEO_{60w}/PCL_{5w} separators reaches 144 mAh g⁻¹ (1C) and 103 mAh g⁻¹ (5C) at room temperature, respectively. Notably, Li/PP-blended PEO_{60w}/PCL_{5w}/LiFePO₄ shows an improved capacity retention rate of 77% after 800 cycles, confirming that the functionalized separator with coated PEO/PCL blend has great potential for application in the field of LMBs.

Keywords: Lithium metal batteries, polyolefin separators, polymer blend, coating, battery performance



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INTRODUCTION

Lithium metal as an anode has a high specific energy density of 3,860 mAh g⁻¹ and a low anode potential of -3.04 V (*vs.* standard hydrogen electrode), causing lithium metal batteries (LMBs) to have great potential to meet the high energy density demand of energy storage field^[1-4]. However, the high reactivity and conversion chemistry of lithium anode have resulted in the cycling instabilities and safety concerns of LMBs^[5-7]. Thus, it is a great challenge to design high-performance LMBs for the long cycle life with high safety. Among the important components of batteries, the separator plays an important role in separating the two electrodes, preventing internal short circuits and providing ions transport channels^[8-11]. Besides, advanced functional separators can further promote the overall performance of batteries due to the improvement of low impedance, excellent thermal stability, good interface performance, *etc.*^[12-14].

Polyolefin-based microporous separators manufactured by polyethylene (PE) and polypropylene (PP) dominate commercial battery separators due to their low cost for large-scale production, excellent mechanical properties, and good stability. However, the disadvantages of sluggish lithium-ion diffusion and poor wettability with electrolytes lead to safety issues and make satisfactory battery performance hard to achieve^[15-21]. Therefore, researchers have conducted numerous studies to modify polyolefin separators, with surface coating being one of the most effective and simplest methods^[22]. The commonly coated polymers include poly(vinylidene fluoride) (PVDF)^[23], poly(ethylene oxide) (PEO)^[24], poly(methyl methacrylate) (PMMA)^[25], polyimide (PI)^[26], and so on. Wang *et al.* coated PVDF organic particles on traditional PE separators to achieve good electrolyte wettability. However, the battery capacity based on the coated separator was not high enough (122 mAh g⁻¹ under 1C) and could not involve the long cycle performance of the battery^[27]. Tian *et al.* fabricated a composite separator by coating organic palygorskite and methyl methacrylate (MMA) onto both sides of a polyolefin-based separator. A high lithium-ion transference number (t_{Li^+}) of 0.89 and a good discharge capacity of 164 mAh g⁻¹ (0.1C) with only a 77% capacity retention rate (200 cycles) were obtained^[28]. Although coating polymers on separators could effectively improve the battery performance, new polymers should be designed to meet the increasing demands of the capacity and long cycling stability of advanced LMBs.

Poly(ϵ -caprolactone) (PCL) has excellent electrochemical stability (~ 5 V), high dielectric constant, and weak interaction between carbonyl groups and lithium ions, which has attracted great interest in the field of electrolytes^[29-31]. Li *et al.* reported a self-catalyzed strategy for brush-typed PCL-based polymer electrolyte with a high ionic conductivity [5.53×10^{-2} mS cm⁻¹ at room temperature (RT)] and t_{Li^+} (0.82)^[32]. However, due to the poor compatibility of PCL and polyolefin-based microporous separators, there is little research on improving battery performance using the PCL as a constituent of the separator. Although Di Carli *et al.* made PAN and PCL into separators using an electrospinning procedure, the strategies and electrochemical performance, especially in capacity and cycling stability, need further improvement^[33]. As a common polymer electrolyte and separator coating material, PEO not only avoids the risk of delamination between the modified layer and the separator but also adsorbs liquid electrolyte and reduces the reaction between electrolytes and lithium metal^[24]. Furthermore, introducing PEO and PCL components into the electrolyte can effectively change the coordination structure of Li⁺ and improve the ionic conductivity and lithium-ion transference number^[29]. Therefore, the polymer coating of blend PEO and PCL on the PP separator can further improve the performance of LMBs.

In this work, we propose a simple but effective strategy to blend PEO and PCL (with a mass ratio of 1:2) as the coating of commercial PP separators to provide a smooth atmosphere for ion migration and enhance the cycle stability of LMBs. The component of PEO can not only assist the transport of lithium ions^[3] but also work as a binder to make it more uniform for PCL coating on the PP separator surface. The strong

polar ester carbonyl group in PCL plays a key role in promoting the dissociation of lithium salts and improving the interfacial compatibility between electrodes and electrolytes^[34-36]. Compared to batteries containing separators with PEO-coated and pure PP separators, the capacity and cycling stability of batteries assembled with PEO/PCL blend-coated PP separator (PP-blended PEO_{60W}/PCL_{5W}) are significantly improved. The Li/LiFePO₄ (LFP) cell with the functionalized separator shows a high specific discharge capacity of 146 mAh g⁻¹ under 1C and 103 mAh g⁻¹ under 5C at RT, showing the potential for large-scale production of separators via the simple and effective method.

RESULTS AND DISCUSSION

Structure characterization of separators

A novel functionalized separator coating by blending PEO_{60W} and PCL_{5W} for PP-based separators was prepared using the blade coating method [Figure 1]. Besides, the coating with PEO_{60W} or PCL_{5W} for PP separators was prepared similarly to explore the influencing factors. Fourier-transform infrared spectra (FT-IR) are used to analyze the coating of PEO_{60W}, PCL_{5W}, and blended PEO_{60W}/PCL_{5W} on the commercial PP separator surface. As depicted in Figure 2A, an obvious absorption peak around 1,727 cm⁻¹ corresponds to the stretching vibration of C=O belonging to PCL, and the peak around 1,108 cm⁻¹ corresponds to the stretching vibration of -C-O-C- belonging to PEO, confirming the coatings of PEO and PCL were coated on the PP separator successfully^[37]. Meanwhile, both occur on the corresponding curve of PP-blended PEO_{60W}/PCL_{5W}, representing the successful coating of PEO_{60W}/PCL_{5W}. Besides, the lithium salt [lithium bis(trifluoromethanesulfonyl)imide (LiTFSI)] was only added to the PP-PEO_{60W} separator, and the molar ratio of the ethylene oxide (EO) chain segment and LiTFSI is 8:1. No absorption peak around 1,648 cm⁻¹ belongs to LiTFSI, which appeared in other separators such as PP-PCL_{5W} and PP-blended PEO_{60W}/PCL_{5W} separators. The ¹H-nuclear magnetic resonance (¹H NMR) spectra of PCL_{5W}, PEO_{60W} and PP-Blended PEO_{60W}/PCL_{5W} separators are shown in Supplementary Figure 1. The similar peaks belonging to PEO (3.5 ppm) and PCL (between 2.4 and 1.2 ppm) exist in the PP-Blended PEO_{60W}/PCL_{5W} separator, indicating that the PP-Blended PEO_{60W}/PCL_{5W} separator is successfully prepared. As shown in Supplementary Figure 2, because of the good adhesive quality and high flexibility of PEO, a well-coated PP separator can be obtained. However, for a large fabrication process, owing to the poor compatibility between PCL and PP separators, the coatings of pure PCL cannot form uniform coatings without adding PEO as a binder. Luckily, the blended PEO and PCL can overcome this dispersion issue through the intermolecular interaction between the two polymers and the enhanced compatibility of the PP separator.

Scanning electron microscopy (SEM) was used to observe more visible characterizations of separators. As shown in Figure 2B, the average thickness of blended polymer coating on the top of the PP separator is about 1.3 μm (as calculated from the SEM cross-sectional imaging). The element mapping [Figure 2B] further demonstrates the coating distributed on the surface without clogging the pores of the PP separator. The top-view SEM images of the PP separator and coated separators are shown in Figure 2C-E, indicating that a uniform polymer coating was prepared on the top of the PP separator, and its micrometer porous morphology is retained on the uncoated side.

The result of surface area and pore size distributions in the separators was verified by Brunauer-Emmett-Teller (BET) analyses [Supplementary Figure 3]. Due to the stretching process during the PP separator production, it does not contain homogeneously distributed slit pores, and the PP and PP-Blended PEO_{60W}/PCL_{5W} separators show typical type III adsorption isotherm curves^[38]. Owing to the coating on the separator, its porosity is decreased. The pore size distributions of the PP separator do not change after coating, indicating that the polymer coating only distributes on its surface, and its pore structure is preserved.

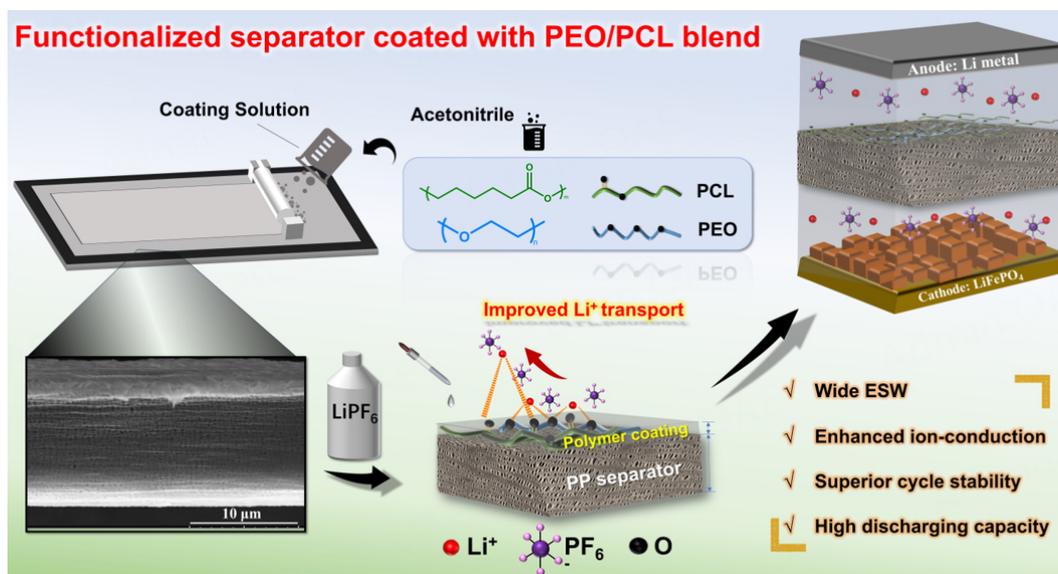


Figure 1. Schematic illustration of functionalized separator coated with polymer blend for the application in LMBs.

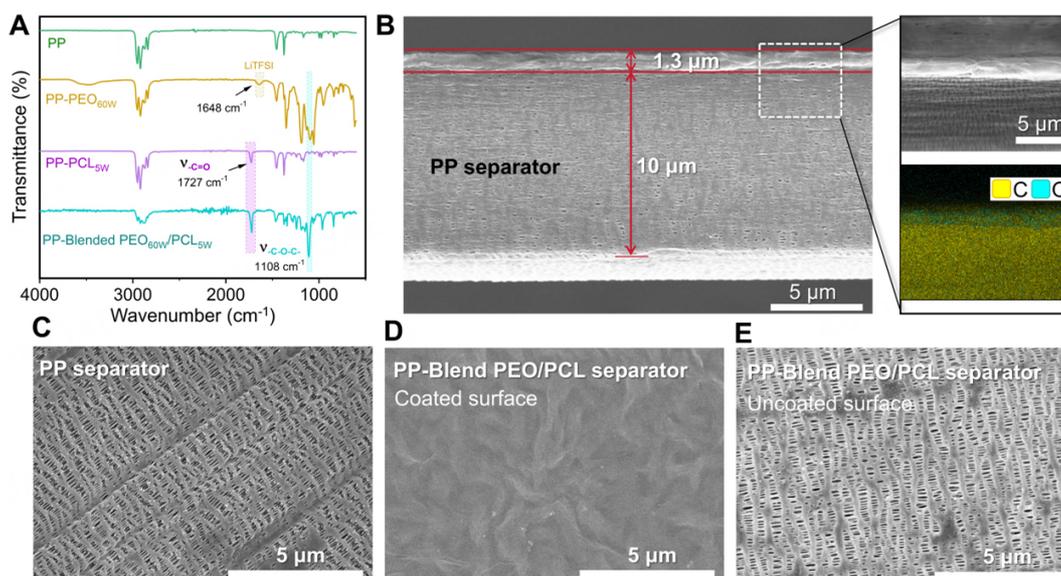


Figure 2. Characterization of the separators: (A) FTIR spectra of PP, PP-PEO_{60w}, PP-PCL_{5w}, and PP-blended PEO_{60w}/PCL_{5w} separators; (B) Cross-sectional SEM images and the corresponding EDS element mapping of C and O of the PP-blended PEO_{60w}/PCL_{5w}; Top-view SEM images of PP separator (C) and PP-blended PEO_{60w}/PCL_{5w} separator with the coated surface (D) and uncoated surface (E).

Physical property characterization of separators

In advanced LMBs, the thermal stability of the separator is one of the evaluation criteria for safety. As shown in [Figure 3A](#), the decomposition temperature of PP separators is around 400 °C. Although the decomposition temperature of coated separators is slightly decreased to 377 °C because of the lower decomposition temperature of polymer coatings^[39,40], they are still enough for battery application. As shown in [Figure 3B](#), the PP-PEO_{60w} (EO:Li⁺ = 8:1) and PP-blended PEO_{60w}/PCL_{5w} separators show improved thermal stability compared to the commercial PP separator. The PP separator shows a slight irreversible thermal shrinkage of about 5% at 120 °C, while no significant size change is found on the coated separators.

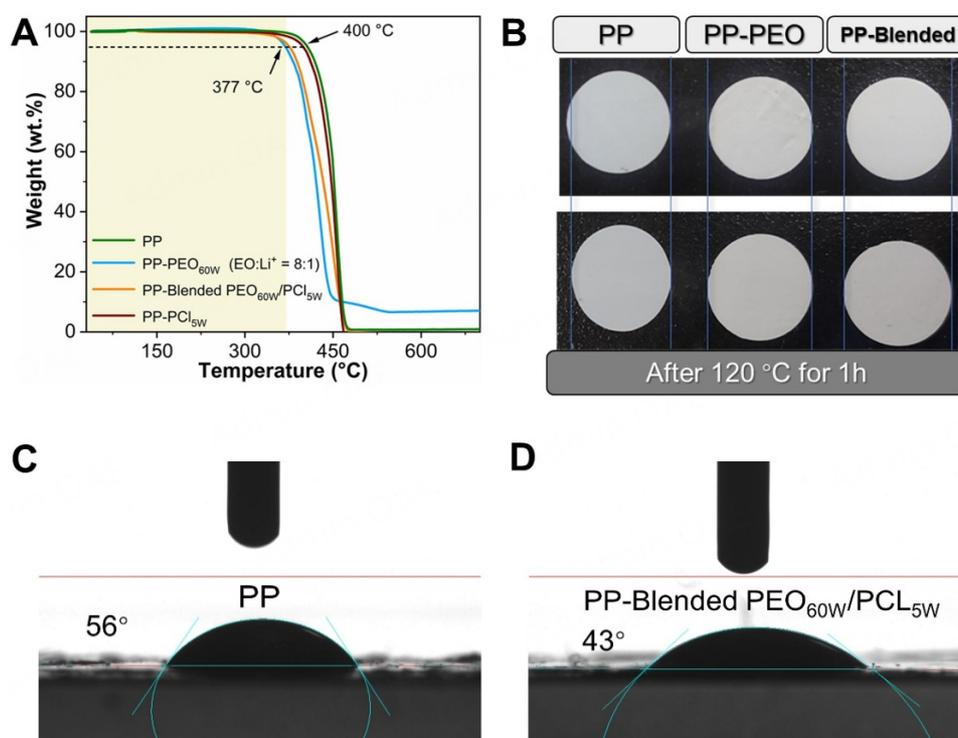


Figure 3. Physical and chemical properties of separators: (A) Thermal gravimetric analysis (TGA) curves of PP, PP-PEO_{60W} (EO:Li⁺ = 8:1), PP-PCL_{5W} and PP-blended PEO_{60W}/PCL_{5W} separators; (B) Thermal shrinkage of PP, PP-PEO_{60W} (EO:Li⁺ = 8:1) and PP-blended PEO_{60W}/PCL_{5W} separators exposed at 120 °C for 1 h; Liquid electrolyte contact angles of PP separator (C) and PP-blended PEO_{60W}/PCL_{5W} separator (D).

Besides, the PP-blended PEO_{60W}/PCL_{5W} separator shows lower thermal shrinkage than a PP separator at higher temperatures [Supplementary Figure 4], indicating the coating can slightly improve the dimensional stability of the separator. The mechanical properties of different separators on drawing directions were also tested. As shown in Supplementary Figure 5, the PP-based separator has a maximum tensile strength of about 137.7 MPa, while the flexible polymer coatings on the PP separators do not significantly reduce their mechanical properties.

Electrolyte wettability of battery separators is crucial for efficient ion conduction in polyolefin separators, and good wettability helps decrease the internal ionic resistance and shorten electrolyte penetration time during the cell assembly^[41]. The electrolyte wettability was evaluated using contact angle measurements of a liquid electrolyte (LiPF₆ in a carbonate-based electrolyte). As shown in Figure 3C, owing to the hydrophobic nature and low surface energies of the PP separator [Supplementary Figure 6A and B], the PP separator shows a large contact angle of 56° with liquid electrolyte, indicating the poor wettability between the PP separator and electrolyte. Thanks to the enhanced interaction with electrolyte by the polar ester carbonyl in PCL and EO chain in PEO, the contact angle of the PP-blended PEO_{60W}/PCL_{5W} separator reduces to 43° [Figure 3D]. In addition, the PEO and PCL coatings also have promoted contact angles (around 45°) between the coated separators and the electrolyte [Supplementary Figure 6C-E]. Because separators have varying wettability for different electrolytes^[42], we further compare the wettability of the separators with the LP-07 electrolyte, and the results are shown in Supplementary Figure 7. The PP-Blend PEO_{60W}/PCL_{5W} separator is well infiltrated by LP-07 electrolyte; meanwhile, the PP separator remains un-infiltrated after 30 s. Owing to the low wettability between carbonate-based electrolytes and PP separators, the coatings provide a more obvious improvement in wettability, which widens the selection range of electrolytes.

Electrochemical performance

The separators were packaged into cells to investigate the electrochemical properties. The ionic conductivity of cells with PP separators and various coated separators was measured. As shown in [Figure 4A](#), the PP-PEO_{60w} (EO:Li⁺) and PP-blended PEO_{60w}/PCL_{5w} separators have higher ionic conductivity than PP separators as 0.66 and 0.63 mS cm⁻¹ at 30 °C, respectively, indicating that the PEO and PCL in separators play a role in promoting the dissociation of lithium salts and assisting in the ionic migration. Due to the inferior conductivity of PCL, the ionic conductivity of PP-PCL_{5w} separators slightly decreases to 0.52 mS cm⁻¹ (30 °C). As shown in [Supplementary Figure 8](#), adding LiTFSI improves ionic conductivity by coordinating with ethereal oxygen of PEO. However, this means increased costs. Besides, it is necessary for coating to avoid being too thin to face uneven issues which may reduce ionic conductivity. Owing to the component of PCL, not only the coordination interaction between ethereal oxygen and lithium ions is weakened by weak ion coordination of polyester, but also part of the crystallization zones of PEO is destroyed, avoiding the addition of additional lithium salts. Therefore, the PP-blended PEO_{60w}/PCL_{5w} separator not only has high ionic conductivity but also balances performance and cost.

As shown in [Figure 4B](#) and [Supplementary Figure 9](#), the t_{Li^+} of PP-blended PEO_{60w}/PCL_{5w} is significantly enhanced relative to pure PP and PP-PEO_{60w} separators thanks to the PCL component. The t_{Li^+} of PP, PP-PEO_{60w} (EO:Li⁺ = 8:1), PP-PCL_{5w}, and PP-blended PEO_{60w}/PCL_{5w} separators is 0.29, 0.31, 0.63, and 0.46, respectively. The coordination interaction between the strong polar ester carbonyl group and lithium ions in PCL is less than the ethereal oxygen and lithium ions in PEO. The weaker coordination interaction promotes lithium ion transport^[35]. Compared with the reported coatings on polyolefin-based separators, the coatings of PCL and blended PEO/PCL have a good effect on improving lithium-ion conduction [[Supplementary Table 1](#)].

[Figure 4C](#) shows the electrochemical impedance spectra (EIS) of the lithium symmetric cell of PP-blended PEO_{60w}/PCL_{5w} and PP separators. It is clear that the interfacial resistance of the PP-blended PEO_{60w}/PCL_{5w} separator (about 220 Ω) is less than the PP separator (about 430 Ω), indicating a better interface between electrolyte and electrode provided by the blended PEO_{60w}/PCL_{5w} coating. Therefore, coating PEO_{60w}/PCL_{5w} on PP separators is beneficial for reducing interfacial resistance. Lower interfacial resistance positively influences electrochemical properties, especially resulting in a higher t_{Li^+} ^[43]. Also, the functionalized separator possesses enough electrochemical stability (4.4 V) for LMBs [[Figure 4D](#)].

To sum up, as shown in [Figure 5](#), because the carbonyl and ether oxygen of coatings have generated interaction with solvent molecules^[9] and coordinated with lithium ions, promoting the destruction of the solvation sheath of Li⁺ and increasing the mobility of Li⁺, the wettability and Li⁺ conductivity of coated separators have been optimized. The PP-blended PEO_{60w}/PCL_{5w} separator has high ionic conductivity, high t_{Li^+} , low interfacial resistance, and good electrochemical stability, indicating that it is extremely suitable for LMBs.

Battery performance

To investigate the interfacial performance between electrolyte and electrodes, the Li//Li symmetric cells with PP-blended PEO_{60w}/PCL_{5w} and PP separators were assembled and measured through the lithium plating-stripping measurements. [Figure 6A](#) shows the long-term performance of lithium symmetric cells with a current density of 0.5 mA cm⁻² and a cycling capacity of 0.5 mAh cm⁻² for the two kinds of separators. The overpotential of Li//Li symmetric cells with PP separators is stabilized at about 70 mV and short-circuited after 170 h due to the growth of lithium dendrites. On the contrary, the lithium symmetric cell with a PP-blended PEO_{60w}/PCL_{5w} separator has a lower initial overpotential of 60 mV and maintains stable cycling for 200 h without a short circuit. Besides, the top-view images of lithium anode from Li-Li

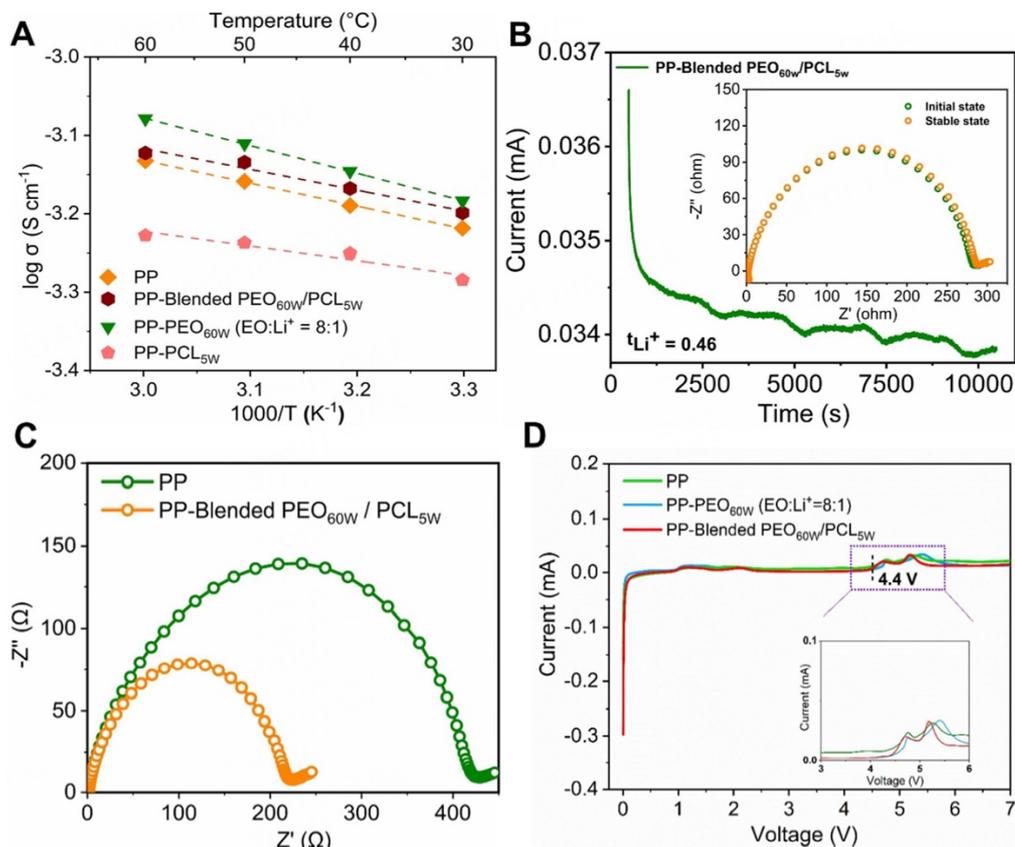


Figure 4. Electrochemical properties of separators: (A) Temperature dependence of ionic conductivity of PP, PP-PEO_{60w} (EO:Li⁺ = 8:1), PP-PCL_{5w} and PP-blended PEO_{60w}/PCL_{5w} separators; (B) Chronoamperometry profile of Li/PP-blended PEO_{60w}/PCL_{5w} separator-LiPF₆/Li symmetric battery under a polarization potential of 10 mV and the EIS before and after the polarization (insert); (C) EIS curves of PP and PP-blended PEO_{60w}/PCL_{5w} separators; (D) Electrochemical stability windows of PP, PP-PEO_{60w} (EO:Li⁺ = 8:1) and PEO_{60w}/PCL_{5w} separators.

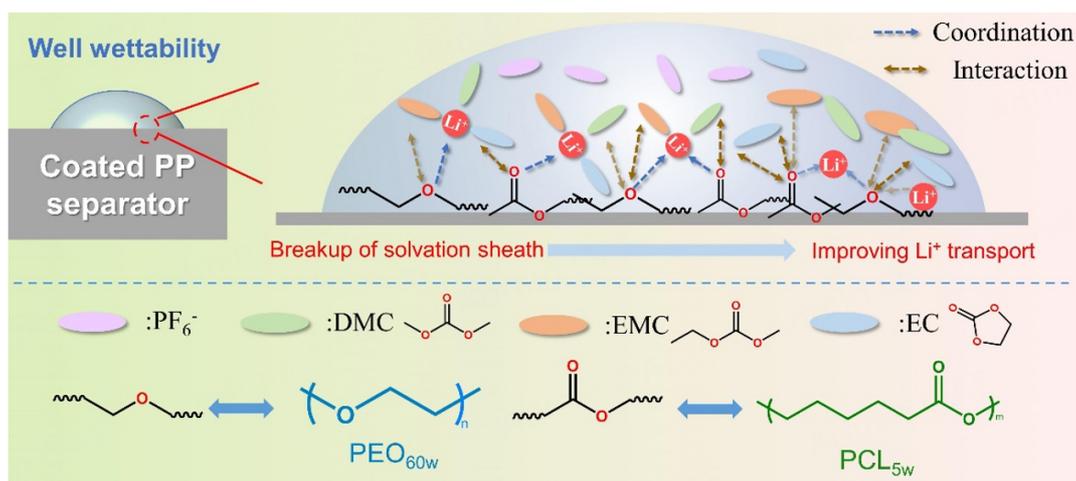


Figure 5. Mechanism diagram for improving wettability by blended PEO/PCL coating.

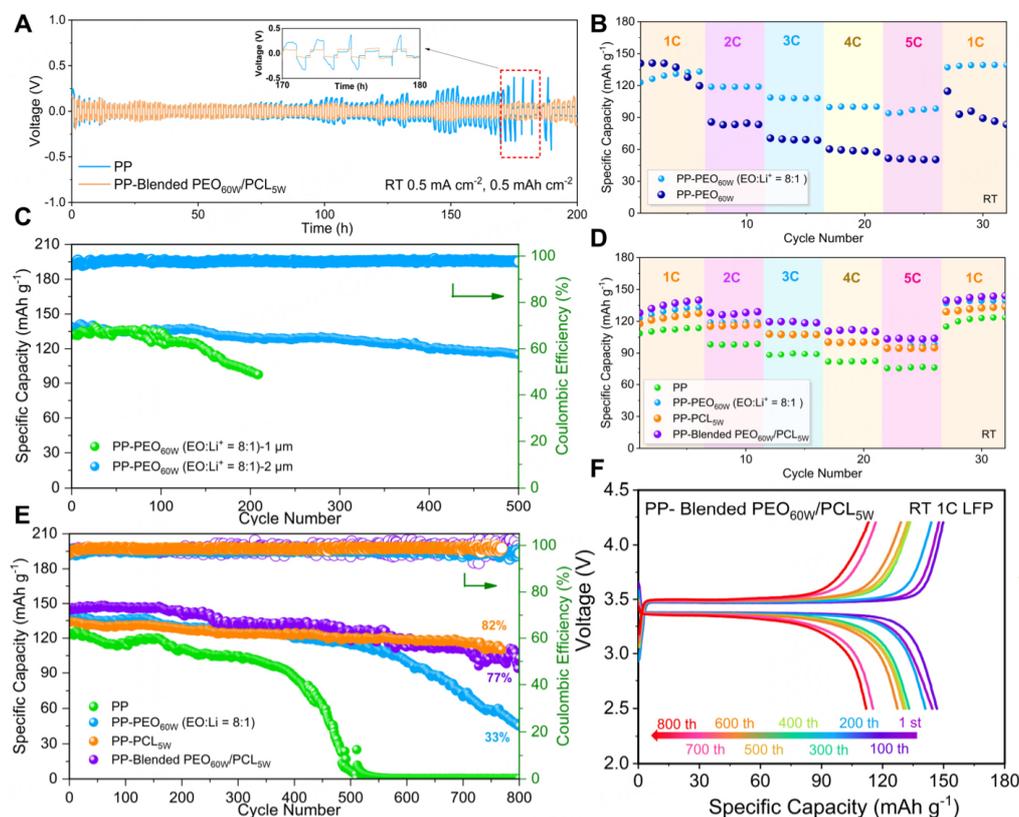


Figure 6. Performance of battery with different separators: (A) Polarization potential voltage-time curves of the Li//Li symmetric cells with PP-blended PEO_{60W}/PCL_{5W} separator and PP separator; (B) C-rate capability of the cells with PP-PEO_{60W} and PP-PEO_{60W} (EO:Li⁺ = 8:1) separators (1C-5C); (C) Cycle stability of the cells with PP-PEO_{60W} (EO:Li⁺ = 8:1) separator (thickness is 1 μm and 2 μm); (D) C-rate capability of the cells with PP, PP-PEO_{60W}, PP-PCL_{5W} and PP-blended PEO_{60W}/PCL_{5W} separators (1C-5C); (E) Cycle stability of the cells with PP, PP-PEO_{60W} (EO:Li⁺ = 8:1), PP-PCL_{5W} and PP-blended PEO_{60W}/PCL_{5W} separators; (F) Charge-discharge curves of Li/PP-blended PEO_{60W}/PCL_{5W} separator/LiFePO₄ at different cycles.

symmetrical battery assembled with PP and PP-Blended PEO_{60W}/PCL_{5W} separators were observed after lithium plating/stripping for 100 h at 0.5 mA cm⁻² by SEM [Supplementary Figure 10]. It is obvious that the lithium anode assembled with a PP separator shows a large number of mossy lithium dendrites; meanwhile, the lithium anode assembled with a PP-Blended PEO_{60W}/PCL_{5W} separator exhibits a better lithium metal surface. It indicates that the PP-blended PEO_{60W}/PCL_{5W} separator has better compatibility and interfacial stability with lithium anode thanks to the high t_{Li^+} , which help form a stable interface and suppress the growth of Li dendrites in LMBs^[44].

To explore the specific capacity and cycling stability of separators, Li/LFP cells were assembled mentioned above and tested. Firstly, the impact of lithium salts on battery performance in PP-PEO_{60W} separators is shown in Figure 6B. Compared to PP-PEO_{60W} separators without lithium salt, the Li//LFP cell with PP-PEO_{60W} (EO:Li⁺ = 8:1) separators has a better rate performance (1C-5C) and cycling stability thanks to the effect of LiTFSI, which tunes the coordination structure of EO chains and the LiF produced by bis(trifluoromethanesulfonyl)imide anion (TFSI) decomposition forms a stable solid electrolyte interphase (SEI). However, compared with LiTFSI content, polymer degradation is preferred for PCL-LiTFSI to form SEI^[45], and the lithium salts have a trifling impact on Li//LFP cells with PP-PCL separators [Supplementary Figure 11]. Regardless of the addition of LiTFSI, both PP-PCL separators demonstrate excellent cycle performance for working about 800 cycles with a high-capacity retention (80%). This

phenomenon further proves that the weak coordination capability of PCL with LiTFSI enhances the smooth ion transport and the dissociation of lithium salts, thus improving the battery performance. Besides, the coating thickness of the PP-PEO_{60w} (EO:Li⁺ = 8:1) separators also obviously influences the performance of LMBs. As shown in [Figure 6C](#), it is obvious that the discharge specific capacity of a Li//LFP cell with 1 μm of coating thickness has a sharp decline after 150 cycles. On the contrary, the PP-PEO_{60w} (EO:Li⁺ = 8:1) with 2 μm of coating thickness can work for over 500 cycles under 1C at RT, indicating that the uniform and flexible PEO coating extends the battery lifespan^[46]. As shown in [Supplementary Figure 12](#), for the PP-blended PEO_{60w}/PCL_{5w} separator, the increased coating thickness leads to a lower ionic conductivity and reduced capacity of Li/LFP cell, indicating that the effect of higher resistance of the coating on electrochemical performance needs to be considered. Moreover, the prepared coatings with the different mass ratios (PEO:PCL = 2:1 and PEO:PCL = 1:1, respectively) on PP separators have a higher t_{Li^+} with the increased PCL content, and the Li/LFP cells with different separators have a similar rate performance [[Supplementary Figure 13](#)], indicating that the battery performance mainly depends on the electrolyte, and the coatings on separator play a role in performance optimization.

[Figure 6D](#) shows the rate capability of Li//LFP cells with distinct separators, in which the Li//LFP cell with a PP-blended PEO_{60w}/PCL_{5w} separator delivers excellent performance, and the detailed specific capacities are 140.0, 128.8, 119.8, 112.1, and 103.6 mAh g⁻¹, at 1C, 2C, 3C, 4C, and 5C, respectively, which have a great improvement compared with the commercial PP separator. The long cycling properties of Li//LFP cells were explored at RT, as shown in [Figure 6E](#). A PP separator has a poor ability to tolerate Li dendrite growth^[47], and the lack of pore connection may cause an uneven Li⁺ concentration gradient across the separator, accelerating local lithium dendrite growth. The Li//LFP cell with a PP separator owns the lowest initial discharge specific capacity of 124 mAh g⁻¹ and rapidly degrades after 450 times cycling at 1C. With the coating of PEO_{60w} (EO:Li⁺ = 8:1), the initial discharge specific capacity of Li//LFP cells increases to 139 mAh g⁻¹. However, its capacity retention rate rapidly declines to 33% after 800 cycles. The PCL coating enhances the lifespan of Li//LFP cells (82%, 770 cycles), but its low ionic conductivity decreases capacity (133.9 mAh g⁻¹). When the components of PEO and PCL are involved, both the battery capacity and the lifespan of the Li//LFP cell with a PP-blended PEO_{60w}/PCL_{5w} separator are favorable. The initial discharge specific capacity of the Li//LFP cell is promoted to 144.5 mAh g⁻¹ and the battery can steadily work for 800 cycles with a high-capacity retention of 77%. By comparison, it can be found that the main role of PEO_{60w} and PCL_{5w} in separators is to improve discharge specific capacity and enhance cycling stability, respectively. The combination of the PCL and PEO has obtained a cell of high capacity and excellent cycling stability. At the same time, it also indicates that high t_{Li^+} and low interface resistance significantly affect the charging and discharging process and long-term cycling stability of Li//LFP cells^[25,48-51]. Notably, the charge-discharge curves of Li//PP-blended PEO_{60w}/PCL_{5w} separator/LFP cells nearly without voltage increase in the polarization platform [[Figure 6F](#)].

In order to further enhance the energy density, high-voltage cathodes such as Ni-rich LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂ (NCM811)^[52] were also tested at a current density of 0.5C from 2.5 to 4.2 V at RT. As shown in [Supplementary Figure 14](#), a high capacity (142 mAh g⁻¹) but rapid decay within 60 cycles in Li/NCM811 cells suggests that the commercial PP separator is not stable enough to form a fine cathode electrolyte interphase (CEI) layer between the electrolyte and the cathodes. By comparison, Li/NCM811 cells with PP-blended PEO_{60w}/PCL_{5w} separators show an increased specific capacity of 142 mAh g⁻¹ and a capacity retention rate of 76% after 95 cycles, confirming that the separator not only has superior interfacial stability and compatibility with lithium anode and LFP cathode but also positively influences NCM811 cathode. Compared with different polymer coating materials, the PP-blend PEO/PCL and PP-PCL separators enhance the t_{Li^+} and lithium deposition; the assembled Li/LFP cell has good capacity and lifespan

[Supplementary Table 2]. The effort to improve the electrochemical performances of Li//NCM ($\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$) remains ongoing, and investigations are now in progress to explore the new polymer coating methods for application in advanced LMBs.

CONCLUSIONS

In summary, we designed a novel separator coating by blending $\text{PEO}_{60\text{w}}$ and $\text{PCL}_{5\text{w}}$ (with a mass ratio of 1:2) for the commercial PP separator. Compared to the PP separator, functionalized separators obtained in this study have excellent electrochemical properties such as great ionic conductivity (0.63 mS cm^{-1} , $30 \text{ }^\circ\text{C}$), high t_{Li^+} of 0.46, and lower interfacial resistance. More importantly, the Li//LFP cell with PP-blended $\text{PEO}_{60\text{w}}/\text{PCL}_{5\text{w}}$ separators combines the high capacity of PEO and the great cycle stability of PCL, achieving superior initial discharge specific capacity of 144 mAh g^{-1} under 1C at RT, which increases by about 17% compared to the PP separator (123 mAh g^{-1}). Besides, it has excellent cycle stability with a high-capacity retention rate of 77% after 800 cycles under 1C. The PP-blended $\text{PEO}_{60\text{w}}/\text{PCL}_{5\text{w}}$ separator not only possesses excellent electrochemical and battery performance but also has good thermal dimensional stability and mechanical performance, confirming that the coating method has great potential for application in the field of battery separators.

DECLARATIONS

Authors' contributions

Methodology, investigation, and writing manuscript: Ye W

Methodology, investigation, and writing manuscript: Fan Z

Project administration and funding acquisition: Zhou X

Conceptualization and supervision: Xue Z

Availability of data and materials

The data supporting our work can be found in the [Supplementary Materials](#).

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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