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The polymer composite electrolyte with polyethylene oxide-grafted graphene oxide as fillers toward stable highcurrent density lithium metal anodes

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Abstract

Adding inorganic fillers to polymer electrolytes is one of the main means to improve ionic conductivity. However, the filler will aggregate, causing the problem of incompatibility between the electrolyte and the metal lithium negative electrode. According to reports, the flaky structure of graphene oxide (GO) has the characteristics of large specific surface area and stable performance, which has attracted widespread attention as a filler for polymer electrolytes. However, the electrolyte membrane has poor compatibility with lithium negative electrodes when GO as a filler, which is not conducive to the full performance of lithium batteries. In this paper, we grafted polyethylene oxide (PEO) on the surface of GO (the product is referred to as GO@PEO) to improving the dispersibility of the filler and the compatibility of the gel polymer electrolyte (GPE) with lithium metal. It is worth that after GO grafting PEO, the impedance of the interface between GPE and lithium metal is significantly reduced, and the compatibility of the electrolyte with the lithium negative electrode is significantly improved. Assembled into Li/Li battery, the surface current cycle is stable, up to 1 mA cm\(^{-2}\). GO@PEO further improves the ionic conductivity of the ion conductivity up to 1.6 mS cm\(^{-1}\). Further prepared into LiFePO\(_4\) (LFP)/Li battery, GPE-GO@PEO showed excellent cycle stability and the discharge specific capacity retention rate was 95.6% after 100 cycles. The rate performance of the battery is also significantly improved. At 5C, the discharge specific capacity of the LFP/Li battery remains 40 mAh g\(^{-1}\).

1. Introduction

Lithium metal has the highest theoretical specific capacity (3860 mAh \cdot g\(^{-1}\)) and the lowest electrochemical potential (\(-3.04\) V), and has been considered as an ideal battery anodes electrode [1]. Due to the uneven precipitation of Li ions on the pole pieces of traditional liquid lithium batteries, Li dendrites can pierce the separator, causing a short circuit of the battery [2]. The high reactivity of the Li anodes electrode with the electrolyte will also reduce the effective transport of Li ions, destroy the stability of the SEI membrane [2, 3], and cause the capacity of the battery to decrease. Polymer solid electrolytes have become an effective way to solve the above problems due to flame retardancy and good thermal stability [4–6]. Polvinyldene fluoride-hexafluoropropylene (PVDF-HFP), because of its strong covalent bond [7, 8] and high dielectric constant [9], is conducive to promoting the full dissociation of lithium salts and increasing the carrier concentration. At the same time, PVDF-HFP has good stability and film formation, and has gradually become a research hotspot of polymer solid electrolyte matrix.

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PVDF-HFP-based polymer electrolytes have high crystallinity, low ionic conductivity, poor stability and compatibility with lithium metal [10, 11]. These problems result in the high capacity advantage of Li anodes not being fully realized. The ionic liquid in the gel polymer electrolyte (GPE) can effectively reduce the crystallinity of the electrolyte. Therefore, this type of electrolyte has gradually become the development trend of polymer electrolytes. According to reports, adding fillers into the gel polymer electrolyte, such as SiO2 [8, 12–14], TiO2 [15–17] and Al2O3 [18–20], can not only further improve the ionic conductivity, also improve the mechanical properties and stability of the electrolyte [21]. The GO with lamellar structure has attracted much attention because of its light texture and stable performance as a filler, and its hydrophilic and hydrophobic properties can well reduce the interface energy [22, 23]. GO can not only reduce the crystallinity of the polymer, but also improve the mechanical properties and flexibility of the polymer electrolyte [24–29]. Previously, we prepared gel polymer electrolysis using GO as fillers under the PVDF-HFP system [30]. The study found that the electrochemical performance of GPE has been significantly improved, and the migration number of lithium ions can reach up to 0.79. However, the interface resistance of GPE to the lithium anode is too large, and the rate performance improvement after assembly into LFP/Li battery is not obvious.

Improving the interface between the electrolyte membrane and the lithium anode has been an important way to improve the performance of lithium batteries [31]. The interface impedance value is small and stable, indicating that the electrolyte membrane has good compatibility with the lithium anodes, which is ultimately beneficial to the cycle performance and rate performance of the lithium battery [32, 33]. According to reports, Liu [34] grafted polyacrylamide (PAM) (GO-g-PAM) on the surface of GO and applied it to commercial separators for liquid lithium batteries. Because of the presence of polymer segments, GO-g-PAM promotes interface compatibility and reduces interface resistance. Inspired by this, grafting polymer segments onto the filler GO of GPE is bound to improve the interface compatibility. At the same time, according to the principle of similar compatibility, GO has better fusion with the polymer matrix after grafting the polymer segment, which makes the dispersion of GO more uniform. Lee [35] grafted small molecular weight polyethylene oxide (PEG) onto GO (denoted as PGO) can improve the mechanical and electrochemical properties of electrolytes. The report shows that our work will not sacrifice the mechanical and electrochemical properties of GPE.

In this paper, PVDF-HFP, lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) salt and 1-methyl-3-ethylimidazole bis(trifluoromethylsulfonyl)imide salt (EMITFSI) prepared as a gel polymer electrolyte. Polyethylene oxide (PEO) is grafted onto GO (note: GO@PEO). Add GO@PEO as a filler to the above electrolyte to obtain a modified gel polymer electrolyte (GPE-GO@PEO), as shown in figure 1. GO@PEO significantly improves the dispersion of GO itself and reduces the interface impedance. After GPE-GO@PEO-2.4% is assembled as a Li/Li battery, it can cycle stably at a rate of 1 mA cm⁻², indicating that the interface compatibility has also been improved. At the same time, the electrochemical performance has been improved. The specific discharge capacity of the LFP/GPE-GO@PEO-2.4%/Li battery can reach 142 mAh g⁻¹, and it can still maintain 40 mAh g⁻¹ at the rate of 5C.
2. Experimental

2.1. GO@PEO preparation
We used edge click chemical reaction to graft PEO on the edge of GO, and obtained modified graphene oxide: GO@PEO. The reaction mechanism is shown in figure S1 (available online at stacks.iop.org/MRX/8/105305/mmedia). Comprehensive analysis by figure S2 FTIR and figure S3-1H-NMR, we successfully synthesized GO@PEO. Figure S4 indicated the modified GO changed the original moisture.

2.2. Preparation of gel-type electrolyte membrane
1 g of LiTFSI, 1 g of PVDF-HFP and 1.4 g of EMITFSI were dissolved in 15 ml of dimethylformamide (DMF) at a mass ratio of 5:5:7, and stirred at 60 °C for 1 h to form solution A. Different amounts of GO@PEO (20.4 mg, 40.8 mg, 61.2 mg, 81.6 mg, 102 mg, mgGO@PEO: mPVDF-HFP + mLiTFSI + mEMITFSI = 0 wt%, 0.6 wt%, 1.2 wt%, 1.8 wt%, 2.4 wt%, 3 wt%) were dispersed in 5 ml of DMF and through thorough sonication to form solution B. Then, the two solutions were mixed and stirred at room temperature for 30 min and then ultrasonically treated for 30 min. The two solutions A and B were mixed and stirred at room temperature for 30 min, and then sonicated for 30 min. Finally, the solution was cast in a mold and dried under a vacuum of 60 °C for 24 h to obtain electrolyte membranes with different GO@PEO contents (note: GPE-GO@PEO-X). The thickness of the resulting membranes was around 150 μm, which were expressed as GPE-GO@PEO-0%, respectively. GPE-GO@PEO-0.6%, GPE-GO@PEO-1.2%, GPE-GO@PEO-1.8%, GPE-GO@PEO-2.4%, GPE-GO@PEO-3%. The GPE-GO@PEO was cut into a 19 mm diaphragm and placed in an argon atmosphere until use. All the above processes are performed in a glove box or vacuum environment, as shown in figure 1.

2.3. Structural and morphological characterization methods
The surface and cross-section morphology of the membranes were investigated by a HITACHIS-4800 scanning electron microscope (SEM). All of the samples were sputtered with gold prior to the SEM measurement. The crystallinity of the prepared polymer electrolyte membrane could be calculated from x-ray diffraction pattern using equation (1):

\[ Xc = \frac{Ac}{Ac + KAa} \times 100\% \]  

where, \( Ac \) represents the area of the diffraction peak of the crystal region under the diffraction curve; \( Aa \) represents the area of the diffraction peak of the amorphous region under the diffraction curve; \( K \) represents the correction factor, and \( K \) can be set to 1 for the purpose of comparison. Before the test, the environment of the instrument should be dehumidified and the ambient temperature should be controlled at 25 °C. Polarizing microscope was used to observe GO distribution in electrolyte membrane. The electrolyte film to be tested was cut into the size of 60 mm long, 20 mm wide and about 150 μm thick. The tensile modulus of the electrolyte was detected by the tensile testing machine (kunshan precision electronic instrument company), and the tensile speed was set as 5 mm min⁻¹. The stress change of electrolyte can be calculated by formula equation (2):

\[ \sigma = \frac{f}{b \times d} \]  

where, \( f \) represents fracture strength of sample (N); \( b \) represents the sample width (mm); \( d \) represents the sample thickness (mm). The interaction and influence between GO-PEO and electrolyte were further analyzed by FT-IR to detect the change of chemical bond and verify the test results of lithium ion migration number.

2.4. Electrochemical application method
The ionic conductivities of the GPE-GO@PEO were measured by electrochemical impedance spectroscopy (EIS). The GPE-GO@PEO were sandwiched between two stainless steel (SS) electrodes in a blocking type cell. Impedance data was obtained by an electrochemical working station (Prinston Versa STAT MC) with a frequency range from 0.1 Hz to 10 kHz from 30 to 100 °C. The ionic conductivity (\( \sigma \)) of the GPE-GO@PEO was calculated using the following equation (3):

\[ \sigma = \frac{h}{R_b \times S} \]  

where, \( h \) is the thickness of the GPE-GO@PEO, \( R_b \) is the bulk resistance, and \( S \) is the area of the stainless steel electrode.

The \( Li^+ \) transference numbers of the GPE-GO@PEO were estimated using a combination of AC impedance and DC polarization techniques [36]. It was obtained through measuring in non-blocking type cells where the GPE were sandwiched between two lithium metal electrodes. The step potential amplitude was 10 mV. The value of \( t_{Li^+} \) was calculated from equation (4):
Where, $V$ is 10 mV, $I_0$ and $I_{ss}$ are the initial and final currents, respectively, and $R_{et}^0$ and $R_{et}^{ss}$ are the cell resistances before and after the polarization. GPE-GO@PEO was assembled into ss/GPE-GO@PEO/Li, and the electrochemical window of GPE-GO@PEO was measured by linear scanning voltammetry (LSV) using a scan rate of 0.1 mV·s$^{-1}$. The voltage range is 0–6 V. The LFP/GPE-GO@PEO/Li battery charge–discharge testing was conducted in the range of 2.7–3.85 V at 25 °C temperature, according to the LAND CT2001A.

3. Results and discussion

3.1. The dispersion of GO

3.1.1. Polarized optics photograph and comparison of tensile of electrolyte film

Figure S5 is the optical photographs of GPE-GO@PEO. They are all flexible and self-supporting. Ionic liquids are adsorbed in the polymer and no separate liquid phase exists. All samples showed uniform colors, indicating that GO@PEO was evenly distributed in the polymer matrix. As the content of GO@PEO increases, the appearance of the electrolyte membrane gradually changes from white to gray, and finally to dark brown.

Figures 2(a)–(c) are the optical photographs of GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% under a polarized light microscope, respectively. Figure 2(a) looks like there are many bubbles piled together, that is the polymer segment in the crystalline state [25, 26]. The bubbles in figures 2(b) and (c) decrease successively, corresponding to the successive decline of the electrolyte membrane’s crystallinity, which conforms to the change law of crystallinity in figure 4(b). In addition, the irregular block particles in figures 2(b) and (c) are GO. GO in figure 2(b) is too large and there is obvious aggregation. Based on the comprehensive analysis of figure S6, it can be seen that the size of block particles is smaller than that of figure 2(b), which indicates that the dispersion effect of GO in electrolyte has been improved after GO grafting PEO.

3.1.2. SEM image of GPE-GO@PEO

In order to further understand the apparent morphology of the prepared electrolyte membrane, the morphology of GPE-GO@PEO was observed by electron microscopy (SEM). Figure 3 is the SEM images of GPE-GO@PEO. This type of GPE-GO@PEO is composed of irregular blocks and gaps. The irregular mass is that the polymer matrix swells and wraps LiTFSI, EMITFSI and GO@PEO. No bare GO@PEO was observed on the surface of the GPE-GO@PEO film, indicating that GO@PEO can be well covered by the polymer matrix. In addition, SEM shows that there are obvious gaps between the polymer segments of the electrolyte, which are caused by the volatilization of the solvent during the drying process of the electrolyte. In the electrolyte preparation process, we have achieved uniformity and standardization, but electrolyte gaps with different morphologies still appear. Therefore, it can be considered as a permissible systematic error. It does not have general regularity and does not do in-depth research.

3.2. GPE-GO@PEO interface compatibility

The interface impedance and interface compatibility of GPE-GO@PEO are our focus. Here, we compare GPE-GO@PEO with unfilled GPE and GPE-0.3 wt% GO, studying the effect of GO grafting PEO on GPE in detail.

3.2.1. Ionic conductivity of GPE-GO@PEO

Ion conductivity is a way to reflect the speed of ions in GPE-GO@PEO. Figure 4(a) shows the ionic conductivity of GPE-GO@PEO with concentrations of 0.6%, 1.2%, 1.8%, 2.4%, and 3% GO@PEO at different temperatures. As the temperature increases, the ionic conductivity tends to increase. When the concentration of GO@PEO is
2.4%, the ionic conductivity can reach 1.6 mS cm\(^{-1}\) at 30 °C, which is also the time when GPE-GO@PEO has the lowest crystallinity. By comparison in table S2, the ionic conductivity of GPE-GO@PEO has obvious advantages in gel polymer electrolyte systems. The curve was fitted by the VTF equation to calculate the ion migration activation energy Ea of GPE-GO@PEO is 0.641, 0.633, 0.612, 0.603, 0.63 J mol\(^{-1}\), respectively. Figure 4(b) shows the distribution of ionic conductivity with temperature for electrolyte without filler (GPE-0%), electrolyte with 0.3% GO as filler (GPE-0.3%) and GPE-GO@PEO-2.4%. On the one hand, the smaller the crystallinity, the higher the ionic conductivity. The test of crystallinity in the figure 7(b) below supports this conclusion. The crystallinity of GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% are 41.4%, 20.2%, and 19.7%, respectively. On the other hand, it is because grafting polymer molecular chains on GO can optimize the internal interface between filler and polymer matrix. Therefore, the modified GO can further enhance the ionic conductivity of the polymer electrolyte. This initially shows that the idea of modified GO is that the electrolyte can be further optimized to effectively improve the performance of the battery.
3.2.2. Electrochemical AC impedance of GPE-GO@PEO

Figures 5 (a) and (b) are the AC impedance before and after the interface of Li/GPE-GO@PEO/Li battery is stabilized; (c) Li/GPE-0%/Li, (d) Li/GPE-0.3%/Li and (e) Li/GPE-GO@PEO-2.4%/Li battery's AC impedance changes with time; (f) GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% AC impedance after fitting the impedance change rule with time.

Figure 5. (a) and (b) are the AC impedance before and after the interface of Li/GPE-GO@PEO/Li battery is stabilized; (c) Li/GPE-0%/Li, (d) Li/GPE-0.3%/Li and (e) Li/GPE-GO@PEO-2.4%/Li battery's AC impedance changes with time; (f) GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% AC impedance after fitting the impedance change rule with time.

3.2.2. Electrochemical AC impedance of GPE-GO@PEO

Figures 5(a) and (b) are the impedance changes of GPE-GO@PEO with different GO@PEO content before and after self-activation. The interface impedance of GPE-GO@PEO with different GO@PEO content is basically the same as the law of ionic conductivity and crystallinity. GPE-GO@PEO-2.4% has the lowest interface impedance and the lowest crystallinity and highest ionic conductivity. Combining figures 4(a) and 5(a), (b), GPE-GO@PEO-2.4% has the highest ion conductivity and the smallest interface impedance. Therefore, the best ratio of GO@PEO in GPE is 2.4%.

Figures 5(c)–(e) is the change of AC impedance between GPE-0%, GPE-0.3%, GPE-GO@PEO-2.4% and lithium electrode with time. It shows that the body impedance of the electrolyte hardly changes with time, and the interface impedance to the lithium electrode first increases, then decreases, and tends to stabilize at the end. This occurrence is related to the formation of SEI film during the activation of the lithium battery itself. In the early stage of SEI film formation, the interface impedance becomes larger. After the SEI film is formed, Li⁺ performs ordered insertion/extraction at the interface, so the impedance gradually decreases and eventually stabilizes. Comparing the time required for the interface impedance of GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% to stabilize by figure 5(f), the interface impedance of GPE-0% and GPE-GO@PEO-2.4% can be stabilized quickly. It shows that GO and GO@PEO as fillers can reduce the reactivity of the interface between the electrolyte and Li anode, make the interface quickly generate SEI film, and ensure the stability of interface
contact. The interface resistance of GPE-0.3% is the largest because of the existence of GO as a ‘rigid’ part, which makes the interface contact of the SEI film poor. Due to the presence of PEO branches, the modified GO achieves the effect of ‘rigidity and flexibility’, improves interface contact, and provides more transmission paths for Li\(^+\) transmission at the interface [34]. Therefore, the interface impedance of GPE-GO@PEO-2.4% becomes smaller again. However, the interface impedances of GPE-0.3% and GPE-GO@PEO-2.4% are larger than GPE-0%.

### 3.2.3. Cycle of Li/GPE-GO@PEO/Li

By assembling Li/Li battery, the stability of GPE to lithium and the reversibility of lithium ion insertion/extraction are further compared. Figure 6 shows the voltage changes of Li/GPE-0%/Li, Li/GPE-0.3%/Li and Li/GPE-GO@PEO-2.4%/Li at different current densities with time. As the circulating current density increases, the voltage of the symmetrical battery gradually increases, which is a polarization phenomenon that occurs for lithium batteries. At low current density of 0.05 mA cm\(^{-2}\), the Li/GPE-0%/Li battery starts to show unstable polarization voltage. When the current density is 0.2 mA cm\(^{-2}\), the Li/GPE-GO@PEO-2.4%/Li battery shows smaller polarization voltage. Figure S7 shows the morphology of Li after 500 h cycling, respectively. After cycling, the lithium metal electrode is not as smooth as before cycling. The clumping of the clusters is caused by the uneven deposition of lithium ions. This is also the main cause of battery cycle performance degradation and short circuit phenomenon. In figure S7(b), the lithium ion cluster is the most serious, and the cluster height is seriously inconsistent, which is easy to squeeze out the electrolyte film and cause short circuit of the battery. This is also why the cycle of this Li-Li battery quickly becomes too polarized. Although the lithium ions in figure S7(c) also have serious clusters, the whole is still flat. Figure S7(d) shows the least amount of lithium ion clusters, and just confirms figure (c) cycle performance is best. Through the comparison in table S1, under the high current density, GPE-GO@PEO has obvious stability advantages over other electrolytes for lithium metal anodes.

### 3.3. Electrochemical and mechanical properties of GPE-GO@PEO

#### 3.3.1. Crystallinity of GPE-GO@PEO

Crystallinity is used to indicate the proportion of crystalline regions in the polymer. Reducing the crystallinity is very important to improve the ionic conductivity. We use x-ray diffraction to study the effect of GO@PEO content on GPE-GO@PEO crystallinity, as shown in figure 7(a). There are obvious peaks at 16° and 22°, which...
correspond to the crystallization peak of PVDF-HFP. With the addition of GO@PEO, the crystallization peak of PVDF-HFP shifted and the intensity decreased significantly, which shows that GO@PEO can reduce the crystallization area of PVDF-HFP. The formula 1 can be used to quantitatively estimate the crystallinity of GPE-GO@PEO, which is 41.4%, 22.3%, 21.3%, 21.2%, 19.7%, and 20.6%, respectively. It is worth noting that the crystallinity of GPE-GO@PEO after adding GO@PEO is smaller than before adding GO@PEO. The crystallinity of GPE-GO@PEO first decreases and then increases. As the amount of GO@PEO added increases, when the content of GO@PEO exceeds 2.4%, the crystallinity of the polymer can no longer be reduced. Figure 7(b) shows that GPE-GO@PEO-2.4% has lower crystallinity than GPE-0% and GPE-0.3%. The smaller crystallinity of the polymer, the more conducive to ion transport. Therefore, the change of the GPE-GO@PEO crystallinity by GO@PEO can also be confirmed from the distribution of the GPE-GO@PEO ion conductivity.

3.3.2. Lithium ion migration number of GPE-GO@PEO
The lithium ion migration number refers to the proportion of lithium ions in the process of ion transmission. The higher the lithium ion migration number, the less side reactions of the battery poles occur. The more stable the battery performance and the better the rate performance. The prepared electrolyte membrane was assembled into a Li/GPE-GO@PEO/Li battery, and the migration number of lithium ions was calculated using equation (3) by the steady-state current method. Figures S9 and 8 are GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% steady-state voltage test, respectively. Through calculation, the $t_{Li^{+}}$ of GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% are 0.28, 0.79 and 0.82, respectively. After adding GO, the $t_{Li^{+}}$ of the polymer electrolyte gradually increases because GO can increase the amorphous region of the electrolyte and provide more space for Li$^{+}$ transport. On the other hand, because of the presence of oxygen-containing groups, GO’s carbon network is deficient in electrons, which produces adsorption for anions and prevents the migration of anions [28], thereby increasing the number of Li$^{+}$ migrations. According to figure S8, With the addition of LiTFSI, the infrared absorption intensity of -CF$_2$ group is significantly reduced, indicating that strong electronegative F atom
interacts with Li⁺, which reduces the stretching vibration of -CF₂ and promotes the dissociation of lithium salt. According to reports by Guo [37] and Chen [28], there is a competitive reaction to Li⁺ between the -CF₂ group of the polymer, the ionic liquid EMITFSI and the filler GO. So Li⁺ migrates in the short-range gel state instead of the long-range jump movement along the polymer chain segment, which ultimately improves the mobility of lithium electrolyte ions. After the addition of GO@PEO, figure 8(c) shows that the mobility of lithium ions is further improved. It may be attributed to PEO effectively alleviates the incompatibility problem between GO and PVDF-HFP. At the same time, GO@PEO enhances the ability to compete with -CF₂ groups for Li⁺, which ultimately promotes the migration of Li⁺ in the gel state.

Through the characterization of GO dispersion, interface performance and electrochemical test analysis, it is found that GO grafted with PEO reduces the interface impedance and improves the cycle performance of Li/Li batteries. GO@PEO further improves the ion conductivity of GPE and the number of lithium ion migration. At the same time, the tensile modulus of GPE is quantitatively estimated by formula 2, and the linear fitting is shown in figure S10. The tensile moduli of GPE-0%, GPE-0.3% and GPE-GO@PEO-2.4% are 1.7 MPa, 1.22 MPa and 1.32 MPa, respectively. With the addition of GO and GO@PEO, the maximum tensile strength of the electrolyte gradually increases. This is because the filler can disperse and absorb external forces, so that the polymer can withstand greater tension. When GO grafts PEO, there will be mutual attraction between PEO and PVDF-HFP, which increases the tensile strength of GPE-GO@PEO-2.4%.

3.4. Characterization of LFP/GPE/Li battery performance

In order to test the application effect of GPE-GO@PEO in batteries, we assembled LiFePO₄ (LFP)/GPE-GO@PEO/Li batteries and tested at 25 ℃. Figure 9(a) shows 100 cycles of LFP/LiGPE-0%, Li/Li, LFP/GPE-0.3% /Li and LFP/GPE-GO@PEO-2.4% /Li at 0.2C rate. We found that the distribution of specific capacity of LFP/LiGPE-0%, Li, LFP/GPE-0.3% /Li and LFP/GPE-GO@PEO-2.4% /Li showed an increasing trend. After cycling, the discharge capacity retention rates of these three batteries relative to themselves were 86.9%, 94.4%, and 95.6%, respectively, which still conform to the law of sequential increase. Through the comparison in table S3, GPE-GO@PEO has good discharge specific capacity retention rate. The difference in coulombic efficiency of these three batteries is not very large, and basically remains above 95%. Figure S11 shows the CV comparison before and after 100 cycles of the LFP/Li battery. The voltage increase is the charging process, and the voltage decrease is the discharging process. Figures S11(a)–(c) respectively show a pair of redox peaks, which indicates that LFP/Li batteries rarely undergo side reactions during charging and discharging. Among them, the CV curve in figure S11(a) changes significantly before and after the cycle, which is due to the decrease in battery discharge capacity. At the same time, the redox peak is obviously asymmetry and the distance is slightly increased, which is caused by the failure of active material and weak polarization [38]. However, the symmetry and consistency of the CV curve before and after the cycle increase sequentially in figure S11, which is due to the increase in ion conductivity and lithium ion migration number and the improvement of the lithium anode interface. Figure 9(b) shows the cycle performance of these three batteries at 0.1C, 0.2C, 0.5C, 1C, 2C, and 5C rates. As the current rate increases, the battery’s reversible discharge specific capacity is gradually decreasing. Among them, LFP/GPE-0.3% /Li has better rate performance than LFP/GPE-0% /Li, mainly because GO improves the lithium ion migration number of the electrolyte and the interface stability to the lithium metal anode. As the rate increases, the capacity of LFP/GPE-0.3% /Li decreases significantly. This may be due to the large interface impedance between GPE-0.3% and the Li negative electrode, which has an effect on the insertion/extraction of lithium ions at the interface certain hindrance. In contrast, the LFP/GPE-GO@PEO-2.4% /Li battery has the best rate performance,
maintaining a specific discharge capacity of 40 mAh g⁻¹ at 5C. This is because GPE-GO@PEO-2.4% not only has good electrochemical performance, but also solves the problem of large interface impedance between electrolyte and lithium anode.

4. Conclusions

We successfully synthesized GO@PEO and used it as a filler to prepare gel polymer electrolytes. It was found that GO@PEO can further reduce the crystallinity of the electrolyte than GO, and the ionic conductivity also increased from 1.3 mS cm⁻¹ to 1.6 mS cm⁻¹. GO@PEO not only effectively reduces the interface impedance between the gel electrolyte membrane and the lithium anode, but also maintains the high lithium ion migration number of the electrolyte membrane. GPE-GO@PEO is assembled into Li/Li battery, which can circulate stably at 1 mA cm⁻² and exhibit small polarization voltage value. It shows that GPE-GO@PEO has good compatibility with lithium anodes. Further assembled into LFP/Li battery, the discharge specific capacity of GPE-GO@PEO is 40% higher than GPE-0%. The rate performance of the LFP/Li battery is also significantly improved, and the specific discharge capacity of GPE-GO@PEO remains 40 mAh g⁻¹ at 5C.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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