A non-Newtonian fluidic cellulose-modified glass microfiber separator for flexible lithium-ion batteries

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Abstract
The separator is of great importance for regulating ion transmission, maintaining electrode stability, and device safety in lithium-ion batteries. Despite their many advantages, glass microfiber separators have shortcomings that often give rise to poor cycle performance and fatal short-circuit risk when used in flexible batteries. Here, we propose the use of a scalable non-Newtonian fluidic cellulose permeation-diffusion strategy to develop a cellulose/glass microfiber composite film with an hourglass-gradient structure. Due to the unique gradient morphology resulting from the presence of cellulose macromolecules, the as-prepared composite film showed an improved surface mass density, adjustable pore structure, controllable ion transport, ideal mechanical flexibility, and robustness. When used as the separator in an assembled lithium-ion battery, the composite film exhibited rapid ion diffusion and Li dendrite inhibition, which endowed the battery with excellent cycle stability (specific capacity of 108.5 mAh g⁻¹ after 1000 cycles) and good rate performance. This composite film shows great potential application in flexible lithium-ion batteries including but not limited to pouch cells.

KEYWORDS
cellulose, flexible batteries, glass microfiber, molecular self-assembly, separator

1 INTRODUCTION

Due to the increasing demand for portable communication devices, flexible electronic equipment, and electric vehicles, there is an urgent need to develop flexible rechargeable energy storage and conversion devices such as supercapacitors, lithium-ion batteries (LIBs), metal-air batteries, and so on. The separator is a part of these electrochemical energy devices that ensures ion transmission and maintains electrode stability and device safety.

Glass microfiber film (G-film) is environmentally friendly and has an affinity for the electrolyte, as well as high-temperature resistance and shows potential commercial applications in LIBs; however, the poor mechanical properties and brittleness of G-film often fail to resist Li dendrite penetration, which gives the resulting LIBs poor cyclic stability. The large pores of G-film...
prevent the uniform diffusion and transmission of ions; thus, more electrolyte is required to fill these spaces to ensure stable battery operation, which increases the risk of flammable liquid leakage. Although several strategies have been developed to improve the properties of G-film, such as dip-coating, casting, immersion, or layer-by-layer assembly, problems still exist, including a lack of true flexibility and non-adjustable pore sizes.

Cellulose is a well-known natural biopolymer that is renewable, processable, and designable. Due to its appealing mechanical performance and voltage resistance, it is believed that cellulose can be used as a sustainable and green material to improve the flexibility and ion transmission of separators. Cellulose can be easily dissolved in ionic liquids by disrupting the hydrogen bonds inside its molecular structure, which results in a fluid system composed of cellulose macromolecules. Compared with the micro- or nano-level ones, molecular cellulose has the unique advantage of the operable spatial-network design, which permits the spatial penetration and assembly for constructing gradient structured materials. In contrast to homogeneous composite structures, gradient structures can be used to produce materials with continuous, gradient changes that give rise to unique properties such as strong mechanical flexibility or robustness; therefore, the use of a cellulose fluid system to controllably optimize the surface mass density and internal structure of G-film is expected to significantly enhance the mechanical performance and optimize their interior structures with massive adjustable mesopores.

In this work, a spatial penetration and assembly strategy using a cellulose fluid system to modify G-film was used to develop a composite film as the separator, termed as Cel/G-gradient film. Based on the non-Newtonian fluidic characteristics of the dissolved cellulose, the molecular cellulose fluid slowly and orderly diffused into the three-dimensional framework of the G-film. Due to hydrogen-bonding of cellulose macromolecules and the

FIGURE 1
Construction and properties of the Cel/G-gradient film. (A), Schematic of the formation of the Cel/G-gradient film. (B), Comparison of the tensile stress of G-film and Cel/G-gradient film. (C), G-film and Cel/G-gradient film pore-size distribution obtained with Barrett–Joyner–Halenda (BJH) nitrogen desorption.
steric hindrance of glass microfibers, a gradient and hierarchical porous structure was constructed in the G-film after converting liquid cellulose into the solid-phase by antisolvent self-assembly (Figure 1(A)); therefore, the loosely-coupled architecture of the G-film and its macropores were transformed into a sturdy structure with lamellar mesoporous separators. Using this strategy, the resultant Cel/G-gradient film presented outstanding flexibility and robust mechanical properties, that is, the tensile strength was 16 times higher than the G-film (Figure 1(B)). The pore structure was optimized to have much smaller and homo-dispersed mesopores (Figure 1(C)), which was more conducive to the rapid and uniform shuttling of ions and the inhibition of Li dendrite growth.

2 | RESULTS AND DISCUSSION

2.1 | Non-Newtonian fluidic performance of dissolved cellulose

The molecular cellulose fluid was prepared by dissolving cellulose in 1-butyl-3-methyl imidazolium chloride ([Bmim]Cl), which physically breaks the hydrogen bonds inside cellulose (Figure S1). By controlling the cellulose content from 0 to 2.0 wt%, the viscosity of the molecular cellulose fluid can be adjusted (Figure 2(A)). The molecular cellulose fluid shows typical non-Newtonian fluid characteristics, that is, the shear stress changes slowly upon increasing the shear rate and $n$ (non-Newtonian index) is less than 1.0 (Figure S2A,B). The rheology results indicate that the viscoelastic behavior of the molecular cellulose fluid is dynamically adjustable (Figure 2(B) and S2C,D). When the cellulose content was less than 1.0 wt%, the cellulose fluid was viscous; inversely, if it was higher than 1.0 wt%, it tended to be elastic. As shown in Figure 2(C), a low-viscosity cellulose fluid leads to low cellulose retention in the G-film (Figure S3A), which is insufficient to construct the secondary structure to improve the properties of the G-film. In contrast, a high-viscosity cellulose fluid with a cellulose content higher than 1.0 wt% allowed large amounts of cellulose to accumulate on the surface and partially penetrate into the inner layers of the G-film. A very high-viscosity cellulose fluid (e.g., cellulose content 2.0 wt%) could not penetrate into the inner layers (Figure S3B,C).

By investigating the pore size distribution and porosity of the Cel/G-gradient film using different viscous cellulose fluids (Figure S4), 1.0 wt% was determined to be the optimal content for preparing the cellulose fluid to construct...
the Cel/G-gradient film (Figure 2(D),(E)). Additionally, by controlling the penetration time, the porosity and microstructure of the Cel/G-gradient film could also be regulated (Figure 2(F) and S5). When the treatment time was 20 min, the porosity of the gradient film reached 77.6%, and changed little with the increase of penetration time. Therefore, the penetration time of 20 min was adopted.

2.2 Morphology and microstructure of Cel/G-gradient film

As a separator in batteries, G-film has good insulation, high-temperature resistance, and chemical stability; however, they often break when twisted or bent due to the physical build-up of microfibers, which makes them poor candidates for use in flexible devices. Luckily, Cel/G-gradient film introduces flexibility into the cellulose into G-film and forms a sturdier structure than the pristine G-film. The composite film can undergo twisting, bending, and even folding into complicated shapes without breaking (Figure 3(A),(B)). To better understand the structural changes due to the presence of cellulose, the microstructure (the surface and cross-sectional images) of the Cel/G-gradient film was observed by SEM. As shown in Figure 3(C), the G-film was composed of loosely interlaced microfibers with an irregular pore size, which is not conducive to the uniform deposition of Li-ions and cannot inhibit dendrite growth. For the Cel/G-gradient film (Figure 3(D),(E)), layered porous cellulose membranes existed both on the surface and in the interior of the G-film and filled most of the space. This modified structure could undoubtedly improve the mechanical robustness and flexibility of the G-film as a separator. As observed from the cross-sectional SEM images, the Cel/G-gradient film presented an hourglass-shaped density gradient distribution and a rhombic-shaped pore size gradient distribution (Figure 3(F),(G) and S6A). The element percentages and map scanning analyses further

FIGURE 3  Morphology and microstructures of Cel/G-gradient film. (A), Digital photos of G-film. (B), Digital photos of Cel/G-gradient film (cellulose content 1 wt%, penetration time 20 min). (C), SEM images of G-film. (D)-(E), SEM images of Cel/G-gradient film. (F)-(G), Cross-sectional SEM images of Cel/G-gradient film.
verified these results (Figure S6B,C). This unique gradient structure of Cel/G-gradient film makes it suitable for use as a separator and the improved surface density can control ion shuttling and inhibit dendrite growth; moreover, the gradient cellulose network structure further strengthened the interior connections of the glass microfibers (Figure S7), which improved the resistance to electrolyte swelling disintegration and large bending deformation.

2.3 Mechanical properties of Cel/G-gradient film

A separator with a high mechanical strength can withstand puncture by lithium dendrites and prevent direct contact between positive and negative electrodes to ensure battery safety. Benefiting from its hourglass-shaped gradient structure, Cel/G-gradient film exhibits a maximum tensile strength of 6.25 MPa, which is about 16 times higher than that of G-film (0.38 MPa, Figure 4(A)). Even when Cel/G-gradient film was wetted by LB-002 electrolyte (1.0 M LiPF₆ in EC: DMC: EMC = 1: 1: 1 vol%), its mechanical strength was 4.75 MPa, which far exceeded that of the G-film (0.25 MPa). The wide gap of 19 times between the two films clearly demonstrates the improved tensile strength due to cellulose modification.

The elastic modulus is also an important parameter that reflects the elastic deformation ability and stiffness of a separator. In both the dry state and when wetted by the LB-002 electrolyte, Cel/G-gradient film exhibited much higher elastic moduli (193.7 and 150 MPa, respectively) than the G-film (33.5 and 16.5 MPa, Figure 4(B)). These results clearly indicate that our strategy involving the penetration and assembly of cellulose fluid is effective for improving the mechanical properties of the G-film. As we can see, the Cel/G-gradient film not only owns an outstanding load-bearing capacity (Figure S8), but also a robust tear resistance when used to lift a 100 g weight, while the G-film could only lift a 50 g before displaying severe damage (Figure 4(C)). In addition to improved mechanical properties, Cel/G-gradient film also exhibited excellent anti-liquid disintegration ability. As shown in Figure 4(D), the Cel/G-gradient film maintained its structural integrity and flexibility even after being immersed in water for 24 h with stirring, whereas the G-film underwent severe breakage and disintegration after only 1 h of immersion.

![Figure 4](image-url)

**Figure 4** Mechanical properties of Cel/G-gradient film. (A), Tensile stress-stain curves of Cel/G-gradient film and G-film. (B), Comparison of the elastic moduli of G-film and Cel/G-gradient film in both the dry and wet states. (C), Photos showing the good tear resistance of Cel/G-gradient film. (D), Comparison of the structural stability of G-film and Cel/G-gradient film after immersion in water.
2.4 Electrolyte absorption and ion transfer of Cel/G-gradient film

The affinity between a separator and electrolyte can directly affect the ionic conductivity and the rate performance of a LIB. To investigate the affinity of G-film and Cel/G-gradient film, contact angle tests were conducted. As shown in Figure 5(A), the Cel/G-gradient film showed a contact angle of 19.1° with the LB-002 electrolyte, which is similar to that of the G-film (17.3°). The electrolyte immersion-height tests confirmed that the Cel/G-gradient film was well-wetted after 60 min (bottom of Figure 5(A)), and the uptakes of KOH solution and LB-002 electrolyte were as high as 602 wt% and 553 wt%, respectively (Figure S9). Even after 30 min of desorption at room temperature, the LB-002 electrolyte retention in Cel/G-gradient film still reached 73.9%, which is higher than that of the G-film (Figure 5(B)). Moreover, Cel/G-gradient film displayed good dimensional stability at high temperatures, showing perfect flexibility even after being baked in a muffle furnace at 200°C for 1 h (Figure 5(C)).

Due to its rich mesoporous structure, ideal electrolyte retention, and structural stability, Cel/G-gradient film is expected to show great potential as a separator in LIBs. As can be seen from the linear sweep voltammetry (LSV) curves in Figure 5(D), a stable electrochemical window between 2.5 and 5.0 V was obtained using the Cel/G-gradient film. To further evaluate the self-discharge performance of the LIBs using Cel/G-gradient film as a separator, LiFePO4/Li half cells were assembled, and G-film was used as the reference. The open-circuit voltage (OCV) curves were measured, and the results are shown in Figure 5(E). After a self-discharge process for up to 2 h, Cel/G-gradient film showed a higher open-circuit voltage plateau of over 3.5 V, which is due to the unique gradient structure and the lithium-philicity of cellulose hydroxyl groups. In contrast, only 3.4 V was obtained using the G-film. In addition, the ion conductivity and Li$^{+}$ transference number of Cel/G-gradient film were measured (Figure 5(F)), which were calculated by electrochemical impedance spectroscopy (Figure S10) and chronoamperometry (Figure S11). The results show that the ion conductivity and Li$^{+}$ transference number of Cel/G-gradient film were 4.34 ms cm$^{-1}$ and 0.57, respectively, which were better than those of the G-film (3.37 ms cm$^{-1}$ and 0.53), indicating its excellent ion diffusion and transport performance.

2.5 Electrochemical properties of the LIB with Cel/G-gradient film

The Cel/G-gradient film, with its pore structure, excellent mechanical strength, and high affinity for Li-ions, is
potentially suitable for use in LIBs. As shown in Figure 6 (A), a LiFePO₄/Li cell was assembled using Cel/G-gradient film as a separator, and the initial discharge specific capacity reached 160.6 mAh g⁻¹ (0.2 C), which is 94.47% of the theoretical specific capacity and higher than that of the G-film (156.5 mAh g⁻¹) (Figure 6(B)). Since the gradient structure promotes Li⁺ shunting and current distribution, the Cel/G-gradient film showed excellent electrochemical properties including a lower charge transfer impedance of 102.2 Ω (Figure S12, Table S1), higher discharge specific capacity at all rates (i.e., 0.2 to 2 C), and better rate capability of 145.9 mAh g⁻¹ at 2 C (Figure 6(C)).

The cyclic performance of the LiFePO₄/Li cell with the G-film or Cel/G-gradient film as the separator was also evaluated at a high current rate of 2 C. After charging and discharging for 1000 cycles (Figure 6(D) and S13), the cell with Cel/G-gradient film exhibited a more stable Coulombic efficiency of over 99% and high retention capacity of up to 108.5 mAh g⁻¹, which is significantly better than that of the G-film-based cell (48.9 mAh g⁻¹). To probe the surface changes of the Li foil after long-term cycling tests, SEM was used to observe the morphology of Li detached from the cells. Figure 6(E), (F) show that after charging/discharging, the Li foil that detached from the Cel/G-gradient film-based cell showed a smoother surface than those detached from the G-film-based cell, indicating that Cel/G-gradient film inhibited the growth of Li dendrites. Due to superior electrolyte retention and ion orientation, the Cel/G-gradient film-based cell still displayed a small charge transfer impedance of 226.2 Ω after 1000 charge/discharge cycles, which was half that of the G-film-based cell (527.6 Ω) (Figure 6(G)).
To validate the possibility of using the Cel/G-gradient film in practical flexible LIB applications, a 6 × 7 cm pouch cell was assembled using LiFePO₄ as the cathode, Li metal as the anode, and the Cel/G-gradient film as the separator (Figure 7(A)). As shown in Figure 7(B), the pouch cell retained a high capacity of 151.1 mAh g⁻¹ even after 40 cycles at 0.5 C and achieved an outstanding cycle retention of nearly 100%. The flexible pouch lit up the “NEFU” pattern in various bending states such as 60° or 180°, and the brightness was barely affected (Figure 7(C)). Throughout these large-scale bending and destructive folding experiments, the strong potential of the as-prepared Cel/G-gradient film separator for Li metal cells or flexible devices has been clearly demonstrated.

3 | CONCLUSION

In this work, we designed a functional gradient separator using molecular cellulose fluid as a non-Newtonian system to permeate the G-film to reconstruct its inner structure. The prepared Cel/G-gradient films possessed excellent flexibility, strong mechanical properties, high surface mass density, abundant mesoporous structure, high electrolyte retention, and anti-disintegration. These properties promote the rapid transmission of ions and inhibit the growth of Li dendrites. When used as the separator in LiFePO₄/Li cells, the cycle performance of the cell based on the Cel/G-gradient film showed excellent stability for over 1000 cycles as well as a high retention capacity of over 100 mAh g⁻¹, which was superior to that of the G-film. In addition, the Cel/G-gradient film has also been demonstrated to be a suitable flexible separator for use in pouch cells. Overall, our study presents a new strategy for developing functional G-film separators using natural biopolymer to construct flexible energy-storage devices such as LIBs.

4 | EXPERIMENTAL SECTION

4.1 | Preparation of Cel/G-gradient film

Cellulose (0.2 g) and 9.8 g [Bmim]Cl were added into a three-neck flask followed by stirring at 80°C for 10 min until the cellulose was completely dissolved (preparation method is consistent with our previous study)¹⁰ Then, 10 g dimethyl sulfoxide (DMSO, AR) was added to dilute

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**FIGURE 7** Electrochemical performance of a flexible Li-ion pouch cell with the Cel/G-gradient film as the separator. (A), Schematic of the inner structure of the pouch cell. (B), Cyclic performance of the pouch cell at 0.5 C. (C), Photos of the pouch cell lighting up the "NEFU" string in different bending states.
the solution (1.0 wt%), which was stirred at room temperature. The G-film was impregnated into the above solution at room temperature and vacuumed at −0.1 MPa for 20 min. Finally, the sample was immersed in distilled water for 8 h to remove [Bmim]Cl and then dried in the oven at 80°C for 24 h.

4.2 Characterization

The SEM micromorphology of each sample was characterized using a JSM-7500F microscope (Hitachi, Tokyo, Japan) at an operating voltage of 10 kV. Nitrogen sorption analysis was performed using an ASAP 2460 Brunauer–Emmett–Teller surface area analyzer (Micromeritics Instrument Co., Ltd., Shanghai, China). The rheological properties were characterized using an ARES-G2 rheometer (TA Instruments, New Castle, DE). Oscillatory frequency sweep experiments of cellulose solutions with different weight percentages were performed from 1 to 100 rad/s with a strain of 0.1 wt% and 1.0 mm gap at 25°C. Steady-state flow experiments of cellulose solutions were performed using shear rates from 0.1 to 1000 s⁻¹ and a 1.0 mm gap at 25°C. The viscosity of homogeneous solutions with different cellulose concentrations was measured at 25°C using a digital rotational viscometer (NDJ-SS, Nimboverde Instrument Co., Ltd.). An Instron 5569 universal mechanical testing machine (Instron Corp., Canton, MA) was used at room temperature. Dumbbell samples were used at a tensile speed of 5 mm min⁻¹ and LB-002 (1.0 M LiPF₆ in EC: DMC: EMC = 1: 1: 1 vol%) electrolyte infiltration.

4.3 Cell assembly

Half cells using LiFePO₄ as the anode and a Li cathode were assembled using a 2032-type coin cell or a 6 × 7 cm pouch cell. Before cell assembly, the LiFePO₄ electrodes were dried at 80°C in a vacuum oven for 12 h. All cells were prepared in a glove box filled with argon (H₂O < 0.1 ppm, O₂ < 0.1 ppm).

4.4 Electrochemical characterization

The ionic conductivity was measured by assembling a symmetrical battery with two stainless steel (SS) electrodes. The Li-ion transfer number was tested by chronoamperometry (potentiostatic polarization method). LSV tests of the assembled Li//separator/SS asymmetrical cell were performed between 2.5 and 5.0 V with a scanning rate of 1 mV s⁻¹. The above tests were carried out on an electrochemical workstation station (CHI 660D, Chenhua Instrument Co., Shanghai, China). The rate capability and cycling performance were tested by assembling LiFePO₄/Li half cells using a Land-CT2001 battery Tester (LAND Electronic Co., Ltd., Wuhan, China) in the potential range from 2.5 to 4.2 V at different currents (1 C = 170 mA g⁻¹).

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest.

AUTHOR CONTRIBUTION

H.Y. supervised the project. D.Z. and Y.Z. designed the experiments. Y.Z. carried out most of the experiments. K.C., W.C. and S.Z. participated in the experiments and analyses. W.C. contributed to the results discussion. Y.Z., D.Z., H.Y. and S.D. collectively cowrote the paper. All authors commented on the final manuscript.

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SUPPORTING INFORMATION

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