Researchers have been developing high-temperature-resistant separators for lithium-ion batteries to improve their safety and performance. A research article in the Journal of Nanomaterials describes the development of an electrospun high-thermal-resistant inorganic composite nonwoven material as a lithium-ion battery separator.

**Research Article**

**Electrospun High-Thermal-Resistant Inorganic Composite Nonwoven as Lithium-Ion Battery Separator**

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**Abstract**

Separators are key materials that ensure the safety of lithium-ion batteries and improve their performance. Current commercial lithium-ion battery separators are mainly polyolefin organic diaphragms, but their temperature instability leads to battery short circuit and fire risk. A flexible SiO₂ nano-fiber membrane combined with a poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) nano-fiber membrane is prepared by an electrospinning method. The mechanical strength of the SiO₂/PVDF-HFP composite nano-fiber membrane (SPF) is twice as high as the pure SiO₂ nano-fiber membrane and at 200°C, there are almost no dimensional changes of the SPF separators. Compared to commercial polyethylene (PE) separators, SPF shows excellent thermal stability and large-area closed cells at 180°C when used in lithium-ion battery separators. The porosity of SPF is 89.7%, which is more than twice than that of an ordinary PE separator. The liquid absorption rate of SPF is much higher than an ordinary PE separator and has reached 483%. Furthermore, the cycle and rate performance of lithium-ion batteries prepared by SPF has been improved significantly. These excellent properties, as well as the potential for large-scale production of electrospinning technology, make SPF an ideal choice for high-power battery separators.

**1. Introduction**

With the continuous advancement of information, materials, and energy technologies, lithium-ion batteries have become a hotspot in the research and application of new power supply technologies in recent years due to their long cycle life, high energy density, and fast charge and discharge rates [1, 2]. A separator is an important component of lithium-ion batteries. It is used to prevent direct contact between positive and negative electrodes, thus effectively avoiding a short-circuit phenomenon and at the same time allowing lithium ions to move quickly between positive and negative electrodes to achieve charging and discharging [3, 4]. The performance of the diaphragm directly affects the internal resistance and interface structure of the battery, which determines the cycle and safety performance of the battery. With the development of wireless electronic products and hybrid electric vehicles, lithium-ion battery separators with good safety performance, high cycle performance, and long service life have been extensively studied. At present, conventional microporous polyolefin separators are most widely used in lithium-ion batteries because of their good chemical stability, adequate thickness, and good mechanical strength. However, the poor thermal stability, poor wettability, and low porosity of the separator itself lead to low energy density, high battery impedance, and low rate performance during storage and discharge of the energy storage battery. Poor thermal stability makes the battery a serious safety hazard during charging and discharging, which greatly hinders the development of the lithium battery [5, 6]. Thus, it is of great significance to study separators with high temperature resistance and strong thermal stability for greatly improving the operating temperature range and safety of lithium-ion batteries. To improve the thermal stability of lithium battery separators, researchers around the world have made great efforts. Using polyolefin separators combined with inorganic...
nanoparticles [7–10] and developing new materials [11–15] are currently two mainstream strategies for the preparation of high-performance battery separators which are resistant to high temperatures and have an enhanced battery rate performance. Among them, lithium battery separators prepared using inorganic materials have absolute safety [16–18]. However, the problem of poor flexibility and mechanical strength of pure inorganic battery separators limit their practical application.

Electrospinning technology is a simple and effective method for preparing nanofibers [19–22]. The nonwoven membrane prepared via electrospinning has many advantages when applied to lithium battery separators, including small pore size, high porosity, large specific surface area, and good gas permeability [23–28]. In addition, electrospinning is also an effective way to solve the problem of lack of flexibility of pure inorganic microporous membranes [29, 30]. Previously, we had successfully fabricated flexible inorganic SiO₂ battery separators via electrospinning [18]. It has excellent heat resistance; however, the low mechanical strength of the pure SiO₂ membrane limits its practical application. PVDF-HFP is a polymer that has been frequently studied for preparing lithium battery separators because of its good absorption of a liquid electrolyte solution, high electrochemical stability, and ideal adhesion to the electrode. In addition, the mechanical support provided by the PVDF crystalline phase gives the PVDF-HFP considerable mechanical strength [31, 32]. Herein, we prepared a SiO₂/PVDF-HFP composite porous fiber membrane (SPF) via electrospinning and rolling bonding and applied it to the battery separator. Such a composite separator was expected to have excellent mechanical stability and a function of heat-closed pores in addition to excellent thermal stability.

2. Experimental

2.1. Preparation of SiO₂/PVDF-HFP Composite Separator

The basic procedure for preparing the composite membrane is shown in Scheme 1. The precursor solution for preparing the SiO₂ fiber membrane was mainly divided into three parts. First, PVA (Mw = 66,000, Aladdin Shanghai) was mixed in deionized water at a mass fraction of 10 wt%, and the mixture was stirred for 6 h at 60°C in a water-bath-heated environment. Next, tetraethyl orthosilicate (TEOS, Aladdin Shanghai), H₂O, and H₃PO₄ (analytical grade, Aladdin Shanghai) were mixed at a mass ratio of 1 : 1 : 0.02 and stirred at room temperature for 4 h. Finally, the above two kinds of uniformly stirred solutions were mixed at a mass ratio of 1 : 1 and stirred at room temperature for 4 h. Thus, a SiO₂ precursor solution which can be used for electrospinning is obtained. Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP, Mw = 455,000, Sigma-Aldrich) was mixed at a concentration of 18 wt% in a solution in which dimethyl formamide (DMF) and acetone were mixed at a mass ratio of 1 : 1, and stirred at a temperature of 40°C for 5 h to obtain the PVDF-HFP spinning solution.

The SiO₂ precursor solution was placed in a plastic syringe and electrospun at a voltage of 18 kV. The distance from the needle to the receiving device was 17 cm. The advance speed of the syringe was 17 μl min⁻¹. The speed of the roller was 200 rpm, and the forward speed of the reciprocating device was 2 cm s⁻¹. Thus, a hybrid fiber membrane was obtained after electrospinning for 4 h in an environment where the temperature was 28°C and the air humidity was 20%. The hybrid fiber membrane was placed in a muffle furnace and calcined to remove the components of the PVA. The heating rate of the muffle furnace was 2°C min⁻¹, and the temperature was maintained at 600°C for 2 h. After natural cooling, a pure inorganic SiO₂ fiber membrane was obtained. The PVDF-HFP solution was then injected into a plastic syringe to prepare nanofibers by electrospinning. The distance between the needle and the receiving device was 15 cm and the spinning voltage was 16 kV. The advance speed of the syringe was 26 μl min⁻¹. The speed of the roller was 200 rpm, and the forward speed of the reciprocating device was 2 cm s⁻¹. Thus, the PVDF-HFP fiber membrane was obtained by electrospinning for 2 h in an environment where the temperature was 25°C and the air humidity was 20%.

The composite process was mainly divided into the following steps: First, the battery-specific glue (Zeon, Japan) was mixed at a concentration of 2% in a mixed solution of deionized water and alcohol at a mass ratio of 2 : 3 and stirred uniformly. The main component of the battery-specific glue is styrene butadiene rubber (SBR). The glue adopts a structure that ensures a lithium-ion channel, so that a large current easily passes. Then, the solution was sprayed to wet the tilled SiO₂ fiber membrane. Finally, the PVDF-HFP fiber membrane was uniformly spread on the SiO₂ fiber membrane and subjected to a rolling treatment at normal temperature. Thus, the SPF was obtained.

2.2. Electrode Preparation and Cell Assembly

The button battery positive electrode prepared for a subsequent battery performance test was prepared from a slurry comprising 5% (mass) of conductive carbon black, 5% by mass of PVDF, and 90% by mass of LiMn₂O₄. Commercial PE separators (Nantong Tianfeng New Electronic Materials Co., Ltd.) and prepared SPF were used as the battery separators, respectively. A mixture of 1 mol L⁻¹ LiPF₆ (DMC : EC : DEC = 1 : 1 : 1) was used as an electrolyte solution, and the assembly process was carried out in an argon atmosphere glove box.

2.3. Characterization of the Separators

Scheme 2 demonstrates the structure of a separator and how the shutdown effect occurs. The surface topographies of the PE separator and SPF were obtained by a Phenom Pro scanning electron microscope (SEM). The composition of SPF was analyzed by a Nicolet iS50 Fourier transform infrared spectrometer (ATR). The mechanical properties of the samples were tested using an Instron 3300 tensile tester. The heat shrinkage test on the separators was carried out in an HD101A-3 electric blast drying oven.
The contact angle test of the separators to the electrolyte was performed by a Theta-type optical contact angle tester. The porosity of the separator was calculated by the \( \text{n-butanol adsorption method} \) and calculated as follows:

\[
P\% = \frac{M_{\text{BuOH}}}{(\rho_{\text{BuOH}} \times (M_{\text{BuOH}}/\rho_{\text{BuOH}} + M_{\text{m}}/\rho_{\text{P}}))} \times 100\%,\]

where \( \rho_{\text{BuOH}} \) and \( \rho_{\text{P}} \) represent the density of \( \text{n-butanol} \) and the polymer, respectively, and \( M_{\text{BuOH}} \) and \( M_{\text{m}} \) represent the mass of \( \text{n-butanol} \) and the separator, respectively. The electrolyte absorption rate of the separators was calculated as follows:

\[
\text{Uptake} \% = \frac{W - W_0}{W_0} \times 100\%,
\]

where \( W_0 \) is the mass before the membrane absorbs the liquid electrolyte, and \( W \) is the mass after the liquid electrolyte is absorbed.

### 3. Results and Discussion

The SEM and diameter distribution of the nanofibers of both sides of SF are shown in Figure 1. It can be seen from the figure that the average diameter of SiO\(_2\) nanofibers is smaller than PVDF-HFP; however, PVDF-HFP fibers are relatively straighter. Figure 1(b) shows the fiber diameter distribution of SiO\(_2\). As can be seen from the figure, the fiber diameters of SiO\(_2\) were mainly concentrated between 130 and 250 nm. However, the diameters of PVDF-HFP nanofibers were mainly concentrated between 300 and 450 nm (Figure 1(d)). Figures 1(e) and 1(f) are the microscopic topographies of the SPF cross-section, which are 1500x and 800x, respectively. In general, SPF composite membranes have good fiber fineness. The pores of the composite fiber membrane are formed by the random distribution of fibers, and we can see that the pore size distribution is relatively uniform.

A gradient was set every 10°C in the range of 150°C-180°C, and the composite membrane was placed in an incubator for 20 min to observe the characterization of PVDF-HFP during melting. Figure 2 shows the specific SEM image. It is apparent from Figure 2 that the SiO\(_2\) fiber did not undergo a significant change after heat treatment at 160-180°C, while the PVDF-HFP fiber did not undergo a significant change after heat treatment only at 150°C. After heat treatment at 160°C, it can be seen that the fibers began to melt at the point of bonding of the fibers to the fibers, and after the heat treatment at 170°C, the melting phenomenon had spread from the fiber bond to the entire fiber. After heat treatment at 180°C, the melting of PVDF-HFP fibers had closed most of the pore size. In the side view of Figure 2(l), we can see that a dense nonporous film was formed on the surface of SiO\(_2\) with SiO\(_2\) fibers as a skeleton. This kind of closed-cell phenomenon has great potential and application
prospects in solving the thermal safety problems caused by high charge and discharge rates of lithium-ion batteries.

The infrared spectrum characterization of both sides of SF is shown in Figure 3(a). The Si-O-Si antisymmetric stretching vibration peak near 1090 cm$^{-1}$, the Si-O bond symmetric stretching vibration peak near 800 cm$^{-1}$, and the Si-O bond symmetric stretching vibration peak near 470 cm$^{-1}$ from the infrared spectrum of the SiO$_2$ surface of the separator [33] can be seen. In the infrared spectrum of the PVDF-HFP surface of the separator, the peak near
Figure 2: Photographs in (a)–(d) sequentially correspond to the SEM image of the PVDF-HFP surface of SPF after heat treatment at 150°C, 160°C, 170°C, and 180°C for 20 min. Photographs in (e)–(h) sequentially correspond to the SEM image of the SiO₂ surface of SPF after heat treatment at 150°C, 160°C, 170°C, and 180°C for 20 min. Photographs in (e)–(l) show the cross-section SEM of the composite separator after heat treatment at 150°C, 160°C, 170°C, and 180°C for 20 min.

Figure 3: (a) SPF two-sided infrared spectrum test chart. (b) The mechanical strength test of SPF and SF.
1403 cm$^{-1}$ corresponded to the deformation vibration of the CH$_2$ group, the peak at 1177 cm$^{-1}$ was the absorption peak of CF$_3$, the peak at 1072 cm$^{-1}$ was the vibration absorption peak of the C-F bond, and the peak near 879 cm$^{-1}$ was the characteristic absorption peak of the amorphous phase [34]. This proved that the prepared separator is a composite separator made of SiO$_2$/PVDF-HFP.

In practical applications, the separator needs to have a certain mechanical strength to suit the assembly process of the battery. The mechanical strengths of the pure SiO$_2$ fiber separator (SF) and the SiO$_2$/PVDF-HFP composite separator were tested and the results are shown in Figure 3(b). It can be seen from Figure 3(b) that the mechanical strength of pure SF can barely reach 2 MPa, and the mechanical strength of SPF can reach 5 MPa. Although SPF has not reached the mechanical strength of commercial diaphragms, it is of great significance for the practical application of inorganic SiO$_2$ separators.

SPF inherits the “absolute” safety advantages of SiO$_2$ diaphragms in terms of thermal stability. Figure 4 shows the results of the PE separator and SF and SPF after being exposed to a heat treatment environment at 200°C for 20 min. As shown in Figure 4, the dimensional changes of SPF and SF were negligible, while the PE separator had melted and shrunk by more than 90%. In fact, PVDF-HFP composited on the SiO$_2$ separator had begun to melt at around 160°C, but the skeleton provided by SiO$_2$ remained unchanged, and the melting of PVDF-HFP just played the role of closed cells. The heat resistance and closed-cell phenomenon of SF have great application prospects for solving the thermal safety problem of lithium-ion batteries.

The affinity of the liquid electrolyte is also one of the important characteristics to judge the performance of the battery separator. During ion transport, ions need to pass through the battery separator through the electrolyte. In order to achieve high ionic conductivity and low resistance inside the battery, the battery separator must retain and absorb a large amount of liquid electrolyte [35–37]. In addition, the faster the liquid electrolyte is absorbed during battery assembly, the easier the electrolyte is injected. Therefore, we used an electrolyte solution to conduct a contact angle (CA) test on both sides of SPF and the PE separator (Figure 5). The CA of the PE separator was 69.31° at 1 s and dropped to 66.91° at 10 s. The SiO$_2$ side of SPF was 21.07° at 1 s and had fallen below 10° after 8 s; the PVDF-HFP side of SPF was 17.69° at 1 s and had fallen below 10° after 4 s. The larger the contact angle, the worse the wettability of the battery separator to the electrolyte. The result indicated that SPF has excellent electrolyte wettability.

Battery separators with high electrolyte absorption and porosity can retain more electrolyte to improve battery performance. The film thickness, porosity, and liquid absorption properties of the PE separator and SPF are summarized in Table 1. The porosity of the PE separator and SPF were 42.3% and 89.7%, respectively. And the porosity of SPF has reached twice that of the PE separator. The electrolyte absorption rates of the PE separator and SPF were 42.3% and 89.7%, respectively. The electrolyte absorption rate of SPF was 8 times that of the PE separator. High porosity

![Figure 4](image1.png)  
(a) Normal temperature photographs of PE, SF, and SPF. (b) Photographs of PE, SF, and SPF after heat treatment at 200°C for 20 min.

![Figure 5](image2.png)  
Figure 5: Contact angle variation of the PE separator and SPF on both sides.

|Separator| PE separator| SPF|
|---|---|---|
|Thickness (μm)| 40.3 | 45 |
|Porosity (%)| 42.3 ± 6 | 89.7 ± 0.5 |
|Average uptake (%)| 60 ± 5 | 483 ± 10 |

Table 1: The porosity and electrolyte uptake of the separators.
Figure 6: Continued.
and electrolyte absorption can enhance the property of lithium-ion batteries.

The battery was tested for cycle performance by charging and discharging the battery at a rate of 1 C from 3.0 V to 4.2 V for 50 cycles. The charge and discharge curves of batteries with different separators and their coulombic efficiency are shown in Figures 6(a), 6(c), and 6(e). Figures 6(b), 6(d), and 6(f) show the charge and discharge curves for the first five cycles and the last five cycles of SPF, PVDF-HFP, and the PE separator, respectively. The capacity of the three batteries did not differ much at the rate of 1 C, and the battery prepared using SPF showed higher capacity at high magnification. This is related to the higher electrolyte absorption rate and porosity of the SPF separator. The battery using SPF, PVDF-HFP, and the PE separator reached a high specific capacity of 120 mAh g⁻¹ in the first cycle, and after a stable cycle of 50 times, the battery using the PE separator maintained a capacity of 50.9% and the PVDF-HFP separator was 66.6%, while the battery using SPF remained at 91.8% capacity. Improvements in battery performance using SPF are associated with high electrolyte absorption rates and better wettability with the electrolyte.

The battery was tested for rate performance by charging and discharging at the rates of 1 C, 2 C, 5 C, 10 C, and 1 C and the results are shown in Figure 6(g). The discharge capacity of a battery prepared using the PE separator and SPF gradually decreased as the magnification was increased. The capacity of the two batteries did not differ much at low magnification, and the battery prepared using SPF showed a higher capacity at high magnification. This is related to the higher electrolyte absorption rate and porosity of SPF.

4. Conclusions

The SiO₂/PVDF-HFP composite separator prepared by the electrospinning method has excellent thermal stability and has the potential for closed cells at high temperatures, which is of great significance for the safe development of lithium batteries in the future. The porosity and electrolyte absorption rate of the SiO₂/PVDF-HFP composite separator were 89.7% and 483%, respectively, which are much higher than that of the traditional PE separator, and the advancement can significantly improve battery performance. The mechanical strength of the SiO₂/PVDF-HFP composite membrane has also been enhanced to 5 MPa, more than doubled compared with the pure SiO₂ separator, while maintaining its own thermal stability, which greatly promotes the practical application of inorganic membranes in lithium-ion batteries.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors’ Contributions

Yuan Xu and Jian-Wei Zhu contributed to this work equally.

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Supplementary Materials

Figure S1: (a) normal temperature photographs of PVDF-HFP; (b) photographs of PVDF-HFP after heat treatment at 200°C for 20 min. (Supplementary Materials)

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