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# Enhanced ionic conductivity and electrochemical capacity of lithium ion battery based on PVDF-HFP/HDPE membrane

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ABSTRACT

Poly(vinylidene fluoride-co-hexafluoropropylene)/high density polyethylene (PVDF-HFP/HDPE) membrane with enhanced ionic conductivity and electrochemical capacity for lithium ion battery application has been successfully prepared by non-solvent induced phase separation (NIPS) technique. HDPE particles with size smaller than 250 nm is achieved via optimized inverse emulsion polymerization method. Results indicate that the novel PVDF-HFP/HDPE membrane has ion conductivity of 2.97 mS $cm^{-1}$ at 25 °C, higher than that 1.92 mS $cm^{-1}$ of pristine PVDF-HFP membrane. Additionally, the $LiCoO_2$/Li cells assembled with PVDF-HFP/HDPE membrane show good C-rate and cycling performance which is parallel to the commercial PP membrane.

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## 1. Introduction

Since the advent of lithium ion battery, the electrolyte also experiences the development of liquid electrolyte, gel electrolyte and solid electrolyte [1–7]. Although the traditional liquid lithium-ion batteries have good electrochemical performance such as rate and cycle characteristic, its electrolyte leakage could easily lead to security problems such as short circuit, spontaneous combustion and explosion [3]. Currently, in all the three electrolytes, solid electrolyte, which has the characteristics of high safety, flexible design in cell geometry, thermal resistance and dimension stability, is the most promising electrolyte, but the poor battery electrochemical performance due to low ionic conductivity is still a huge engineering challenge [4,5,7].

Hence, the study is focused on the gel electrolyte that has better safety than the liquid electrolyte and higher ionic conductivity than solid electrolyte. Zhu reported a simple process to prepare composite glass fiber polymer electrolyte membranes with poly(vinylidenefluoride) (PVDF) and composite of a non-woven fabric with PVDF which achieve a high ionic conductivity and good electrochemical and safe performances [8,9]. Zhang prepared a nanocomposite polymer membrane derived from nano $TiO_2$-PMMA and glass fiber nonwoven [10]. In this study, to achieve gel electrolyte with high ionic conductivity, PVDF-HFP which has lower crystallinity than PVDF is selected as based membrane [11–15]. At the same time, based on our previous study [15], the addition of HDPE particles has positive effect on PVDF based membrane, hence, the HDPE particles with uniform size smaller than 250 nm are prepared via optimized inverse emulsion polymerization method and added to PVDF-HFP to form PVDF-HFP/HDPE membrane via non-solvent induced phase separation (NIPS) technique.

## 2. Experimental

10 g HDPE (Dow chemical company, HDPE3360) was added into flask with 50 ml Dibutyl phthalate (DBP, Xilong Chemical Co., Ltd., AG), and then the flask was sealed and heated at 180 °C for 1 h until most of HDPE was dissolved into homogeneous solution. Next, the solution was quickly poured into a flask containing certain amount of ethanol and was vigorously stirred to generate milky solution. After that, the milky solution was treated with an intermittent ultrasonic sonicator (KQ-100B, 100 W) at room temperature for 30 min to achieve uniform HDPE dispersion solution and avoid the aggregation of HDPE long chain. Then, the solution was subsequently filtrated and the filtrating cake was dried at 50 °C for 5 h, the dried filtrating cake was scraped to agate mortar with ethanol which could remove the residue DBP and grinded. Finally, the grinded filtrating cake was filtrated and dried once again. The achieved cake was grinded and sieved with 300 meshes

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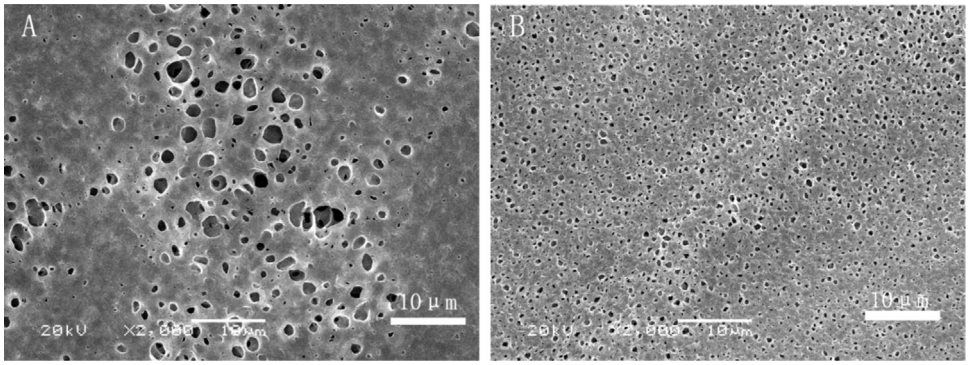


**Fig. 1.** SEM images of pristine PVDF-HFP (A) and PVDF-HFP/HDPE (B) membranes.

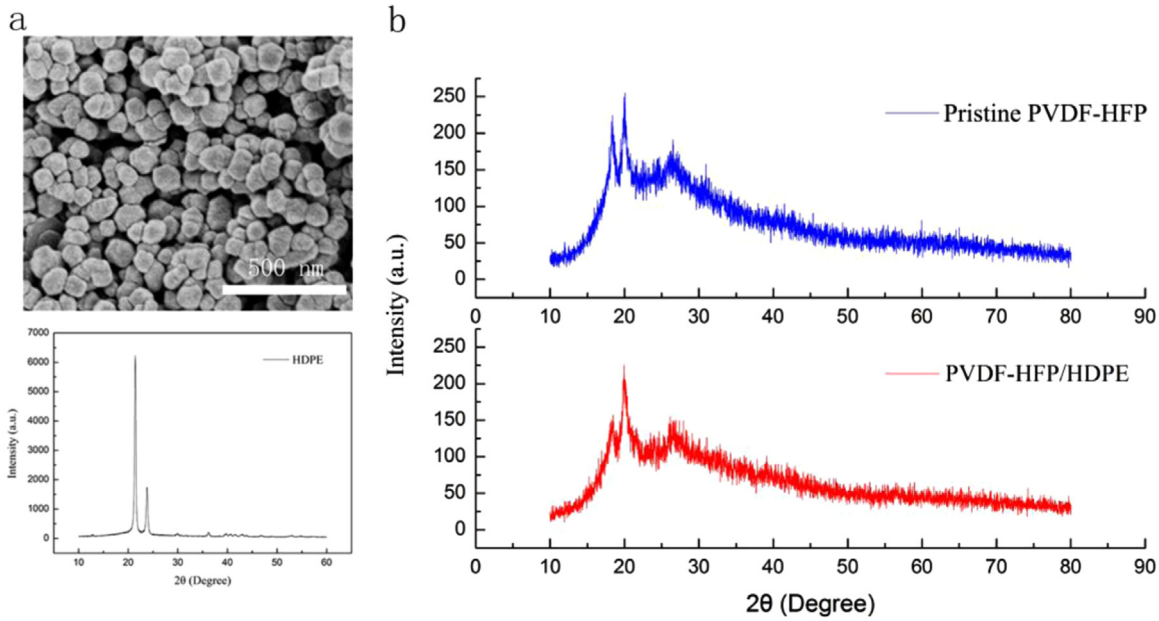


**Fig. 2.** SEM image and XRD pattern of HDPE fillers (a), and XRD pattern of membranes (b).

**Table 1**
Property and uptake of different membranes.

| Properties | PP | PVDF-HFP/ HDPE | Pristine PVDF-HFP |
|---|---|---|---|
| Thickness (μm) | 25 | 80 | 78 |
| Porosity (%) | 41 | 71 | 72 |
| Liquid uptake (%) | 80 | 300 | 210 |
| Mechanical stress (MPa) | 16 (TD), 142 (MD) | 21 | 18 |

to acquire HDPE particles with average size smaller than 250 nm shown in Fig. 1.

22 g pristine PVDF-HFP (Arkemar, Kynar Flex 2801, $M_w$ = 47,000) and 2 g polyvinyl pyrrolidone (PVP, K30, Chemically pure) (mainly as pore forming agent) was mixed in N,N-Dimethylformamide (DMF) in a beaker flask with continuously stirring. Then, 1 g HDPE particles was added to 75 g DMF and the mixture was processed with an intermittent ultrasonic sonicator (KQ-100B, 100 W) at room temperature for 30 min to get uniform HDPE/DMF solution which was finally introduced into the beaker flask with 22 g pristine PVDF-HFP and 2 g PVP (the beaker flask was impermeability). The mixture was continuously stirred at 45 °C for 4 h and then processed in the ultrasonic treatment for 1 h at room temperature. The homogeneous casting solution with 22 wt% PVDF-HFP and 1.0 wt% HDPE was also de-foamed for 5 h.

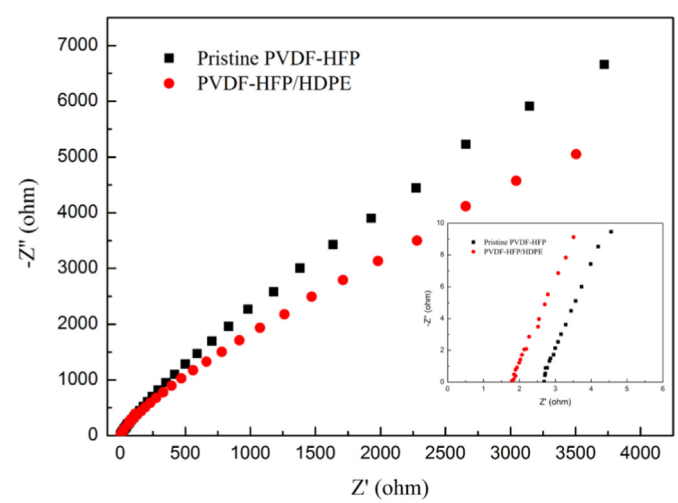


**Fig. 3.** Electrochemical impedance spectrum of pristine PVDF-HFP and PVDF-HFP/HDPE membranes.

The wet membrane was left in air for 20 s at room temperature and then was immersed into deionized water for 12 h. The resulting membrane was dried in vacuum drier at 80 °C for 10 h to remove residue solvent. At the same time, Celgard 2400 PP membrane was used as the comparison.

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