



# Sandwich-Structured Separator of Poly(vinylidene fluoride) and Nonwoven Fabrics for Li-ion Batteries

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## Abstract:

Sandwich-structured composite separators were prepared via thermally induced phase separation (TIPS) method for high energy density Li-ion batteries. The composite separator comprises poly(vinylidene fluoride) (PVDF) porous substrate and polyethylene terephthalate (PET) nonwoven fabrics in between. In contrast to the pure PVDF separator prepared via TIPS, the nonwoven fabrics impart the porous PVDF separator with enhanced mechanical strength and thermal stability. Besides, nonwoven fabrics provide PVDF separators with superior thermal stability and thermal-shutdown property. The combination of nonwoven fabrics can be an effective method to solve the remaining problem in this field. In addition, the nonwoven composite separator (NWCS) possesses a dual-asymmetric pore structure caused by asymmetric cooling process, which can ensure a large liquid electrolyte uptake and a low leakage. A mathematical model for asymmetric cooling process is also developed for understanding the formation of dual-asymmetric pore structure. The performances of PVDF, NWCS and Celgard 2400 were systematically compared in this work and Li-ion batteries using NWCSs display higher discharging capacity and retention under different current rates than those of commercial Celgard 2400 separators. Thus the sandwich-structured composite separator is expected to be a promising candidate in next-generation Li-ion batteries.

## Introduction

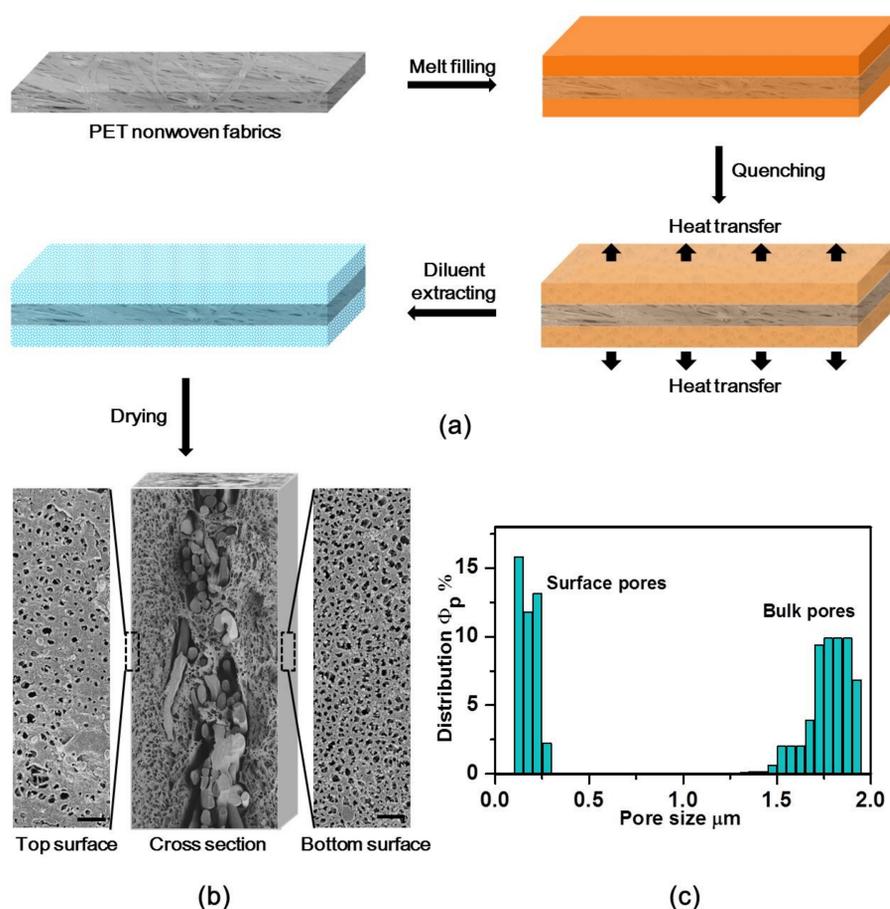


Fig. 1 a) Schematic description of the NWCS preparation process. b) FESEM images of NWCSs including cross section and surfaces (the scale bar in surface images is 1 μm). c) Pore size and its distribution of NWCSs which is detected by gas-liquid displacement method.

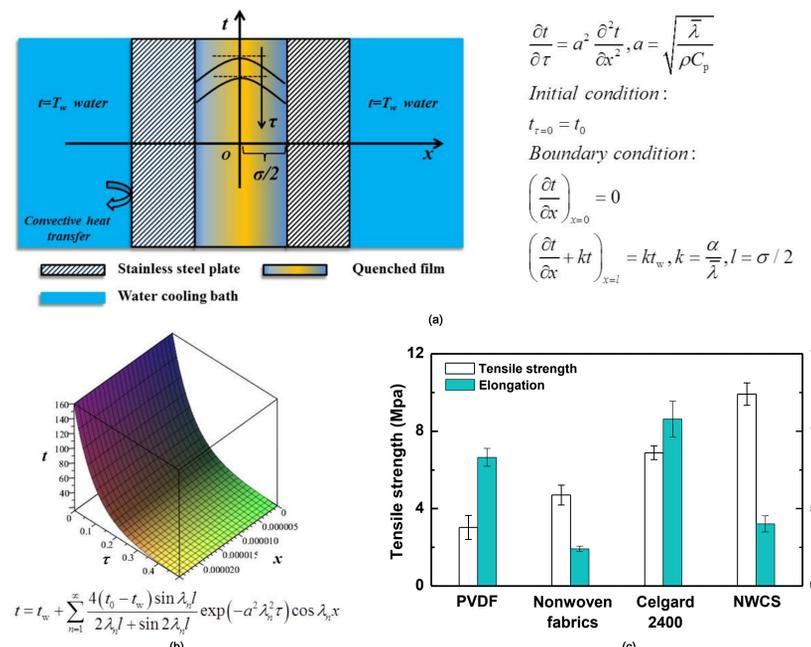


Fig. 3. (a) the mathematical model for asymmetric cooling process via TIPS; (b) the graphical representation of the analytical solution results (the calculation result of  $t$  related with time  $\tau$  and position  $x$  in polymer solution); (c) the mechanical strength of separators including Celgard 2400 (transverse direction), PVDF, nonwoven fabrics and NWCS.

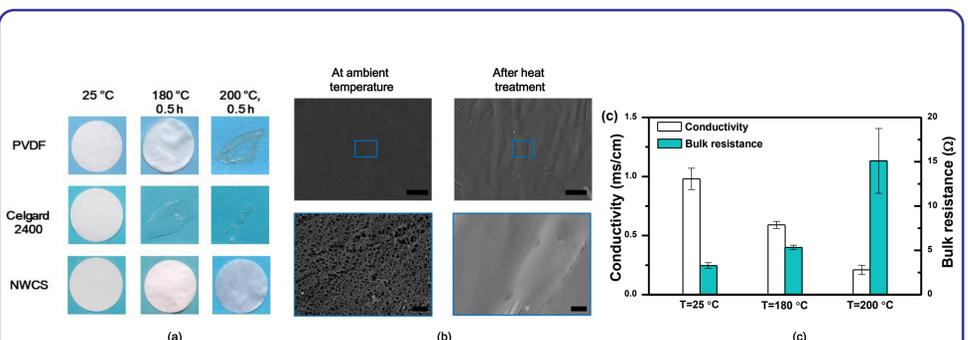


Fig. 4. (a) Thermal stability of PVDF, Celgard 2400 and NWCS before and after heat treatment; (b) Surface pores of NWCS before and after heat treatment (scale bar in SEM images is 10 μm and 2 μm, respectively); (c) The change in ionic conductivity and bulk resistance of NWCS during heat treatment.

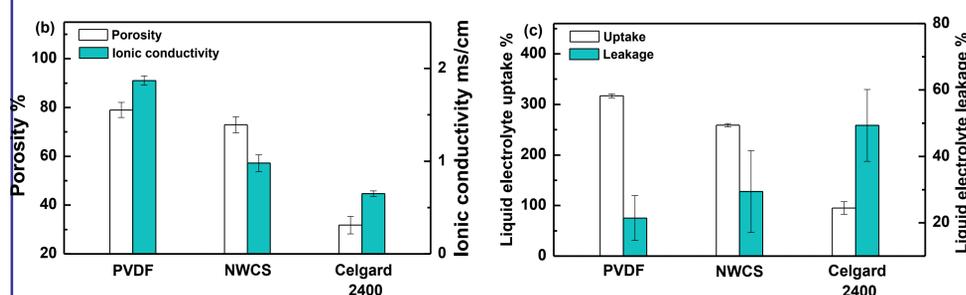


Fig. 2. Performances including porosity and ionic conductivity (a) and uptake and leakage (b) of different separators such as NWCS, Celgard 2400 and PVDF.

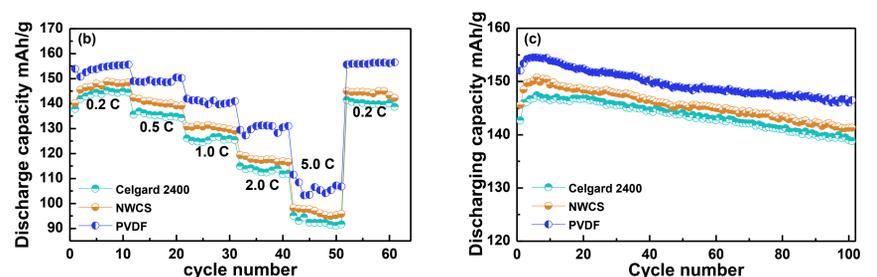


Fig. 5. Cell performance of NWCS, Celgard 2400 and PVDF: (a) C-rate discharging capacity and (b) discharge capacities as a function of cycle number.

## Conclusions

We have demonstrated a TIPS method to prepare a sandwich-structured nonwoven fabrics composite separator. In contrast to the pure PVDF separator prepared via TIPS, the nonwoven fabrics impart the porous PVDF separator with enhanced mechanical strength and thermal stability pure PVDF separator. In the meanwhile, PVDF layer fabricated via TIPS owns a dual-asymmetric structure with porous bulk and dense surface, and it can be understand from the heat transfer mathematical model developed above, because a different cooling rates and temperature gradients is generated in the cooling process between surface and internal part of polymer solution. Moreover, the PVDF matrix and unique structure endows the separator with high electrolyte uptake, large ionic conductivity and low electrolyte leakage. Besides Li-ion batteries using NWCSs display higher discharging capacity and retention under different current rate than those of commercial Celgard 2400 separator. Thus the sandwich-structured composite separator is expected to be a promising alternative in next-generation Li-ion batteries.

## Acknowledgement

The research is financially supported by the National Natural Science Foundation of China (Grant no. 21174124). We also acknowledge Dr. Qing-Yun Wu (Ningbo University) for their kind suggestions to this work.

## References

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