



FULL PAPER

Thermally conductive separator with hierarchical nano/microstructures for improving thermal management of batteries



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Abstract

Thermal management is critical to improving battery performance and suppressing thermal runaway. Besides developing external cooling technologies, it is important to understand and control thermal transport inside batteries. In this paper, heat transfer inside batteries is first analyzed and the thermal conductivity of each component is measured. The results show that low thermal conductivity of the separator is one major barrier for heat transfer in Li-ion batteries. To improve thermal conductivity of the separator, a hierarchical nano/micro- Al_2O_3 /polymer separator is prepared with thermal conductivity of $\sim 1 \text{ W m}^{-1} \text{ K}^{-1}$, representing an enhancement of $5 \times$ compared to commercial polyethylene-based separators. Modeling has been performed to understand mechanism behind the enhancement of thermal conductivity, which suggests that addition of nanoparticles significantly reduces thickness of polymer coating on micron-sized Al_2O_3 particles and thus increase the thermal conductivity of the composite separator. This Al_2O_3 -based separator also has similar ionic conductivity with commercial polymer separators. Such composite separator may have potential applications in developing batteries with better performance and safety.

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Introduction

High-performance batteries are important for various applications ranging from portable electronics, electric vehicles and grid-level energy storage [1,2]. Thermal management of state-of-the-art Li-ion batteries (LIBs) and future higher energy batteries is critical to their performance and safety, especially at large scale [3-6]. The high temperature significantly deteriorates cycle life and it is one important reason to trigger thermal runaway, especially for batteries with high energy and power density [7-10]. Past efforts mainly focus on modeling of external cooling technologies, such as forced air and liquid cooling, to lower battery temperature [5,11-19], where the lumped heat transfer model is widely used without considering thermal conductivity (k) of a battery itself [15-18]. Only a few references took battery thermal conductivity into account with assumed values [5,11,19]. Improving thermal transport inside batteries can also facilitate heat dissipation, reduce temperature inhomogeneity and thermal stress in batteries. In this paper, we first measured thermal conductivity of different components in batteries and identified that the battery separator is a major limiting factor for heat dissipation in batteries. Then a thermally conductive Al_2O_3 /polymer composite separator was developed to improve heat dissipation in batteries. The Al_2O_3 /polymer hybrid separator contains both micron-sized and nano-sized Al_2O_3 particles as the thermally conductive phase, and Poly(vinylidene fluoride-hexafluoropropylene) (PVdF-HFP) as the binder. The large surface area of nanoparticles reduces thickness of polymer coating on Al_2O_3 and enhances thermal conductivity of the separator. At the optimized materials loading ratio, a thermal conductivity of $1.05 \pm 0.16 \text{ W m}^{-1} \text{ K}^{-1}$ is reached, which is more than five times that of commercial polyethylene/polypropylene (PE/PP)-based commercial separator. Such separators could improve heat conduction and reduce the temperature rise of batteries in operations. Meanwhile, the composite separator shows similar ionic conductivity as commercial polyethylene-based separators, avoiding excessive joule heating due to slower ionic transport across the separator.

Materials and methods

Materials

Micro- Al_2O_3 particles ($\sim 10 \mu\text{m}$) were purchased from Sigma Aldrich. Kynar 2801 PVdF-HFP was received from Arkema. Commercial single-side coated LiCoO_2 and graphite electrodes, separators and nano- Al_2O_3 particles ($\sim 100 \text{ nm}$) were purchased from MTI Corporation. The Al_2O_3 /PVdF-HFP composite separator was prepared by dispersing micro- Al_2O_3 particles, nano- Al_2O_3 particles and PVdF-HFP binders in THF for 12 h and drop casting onto a flat glass substrate. A mold made of Aluminum is used to confine the dispersion and shape the separator to the desired dimension. The typical sample size is 3 cm by 3 cm. The weight ratio of PVdF-HFP to THF is fixed as 1:11. The ratio of PVdF-HFP to Al_2O_3 varies from 20: 80 to 10: 90. After drying, the Al_2O_3 /PVdF-HFP film is peeled off from the glass substrate and pressed under 0.1 ton for 5 min at room temperature.

Cross-plane thermal conductivity measurement

The thermal conductivity is measured by stacking multiple electrode films together to ensure that thermal resistance of sample is one order of magnitude higher than the thermal contact resistance between copper and sample. After the environmental temperature is stabilized, the thermoelectric plate cools the bottom side of the sample while the heater is used to keep temperature of the top surface the same as the environment. The heater power (Q) is recorded after temperature is stabilized, indicated as the average power over 30 s after stabilization. Temperature data is recorded by K-type thermocouple. More details can be found in Section 2 of the supporting information.

Ionic conductivity measurement

Ionic conductivity is measured by sandwiching the separator between two pieces of stainless steel with the same size and applying an AC voltage with amplitude of 10 mV at 50 kHz. The electrolyte is 1 M LiClO_4 in Ethylene carbonate/Diethyl carbonate (EC:DEC) with weight ratio of 1:1.

COMSOL simulation

In simulation of temperature rise, external heat transfer coefficient is assumed to be $1000 \text{ W m}^{-1} \text{ K}^{-1}$ and $20 \text{ W m}^{-1} \text{ K}^{-1}$ for forced liquid cooling and force air cooling, respectively. The voltage loss due to internal resistance is supposed to be 0.6 V. The simulation time for 3 C rate is 1200 s. The capacity of 18650 cell is set to 3.1 Ah. Liquid cooling is applied to all surfaces. For prismatic cells, the volumetric energy density is set to 600 W h L^{-1} for the all cells. More details can be found in Section 4 in the supporting information.

Results and discussions

A typical Li-ion battery is made up of a triple-layer structure with a porous separator sandwiched between two composite electrodes (Figure 1a). The porous separator is typically made of polypropylene and polyethylene [20], while two electrodes are mixtures of active materials (e.g. LiCoO_2 or graphite, ~ 80 -95 wt%), carbon black (2-10 wt%) and polymeric binder (1-10 wt%) (See Figure S1 for SEM images of electrodes and separators). The triple-layer structure is assembled together to form either a roll in a cylindrical cell or a cuboid in a prismatic cell (Figure 1b). Voids in electrodes and separators are filled with carbonate-based organic electrolyte. In battery operation, heat is generated throughout the cell due to joule heat and entropy change in electrochemical processes [21]. The heat produced is first conducted inside the cell through both in-plane and cross-plane directions of the triple-layer structure, followed by dissipation process outside the cell, such as air/liquid convection (Figure 1b). The in-plane direction of the triple-layer structure corresponds to the axial direction in cylindrical cells and width/length directions in prismatic cells with a thermal conductivity of $k_{//}$, while the cross-plane direction corresponds to the radial direction in cylindrical cells and thickness direction in prismatic cells

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