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Research Progress in Polymer Separators for Lithium-Ion Batteries

Tongyu Bao

College of Material Science and Engineering, Hainan University, Haikou, China
3555728208@qq.com

Abstract. As the world pushes toward cleaner energy, lithium-ion batteries have quietly become the go-to energy storage choice for everything from phones to electric vehicles. Their appeal comes from decent energy density, a long cycle life, and steadily improving safety. Sitting right in the middle of every battery cell, the separator is easy to overlook but actually plays a huge role in how well — and how safely — the whole system performs. This paper takes a look at where polymer separators stand right now. We start with a quick recap of how lithium-ion batteries are put together and what we actually ask of a separator: electrical insulation, the right thickness and pore structure, electrochemical stability, good wetting by the electrolyte, enough mechanical strength, and the ability to hold up at high temperatures. After that, we walk through the main modification routes people are pursuing — ceramic particle coatings, organic polymer blends, entirely new separator materials, and better binder choices — and talk about why these approaches matter for the two big weak spots of commercial polyolefin films, namely poor thermal stability and lousy electrolyte wetting. We also touch on functional separators, such as nitrogen-containing designs and trilayer films with built-in safety responses, and how they might help with problems like transition-metal leaching from ternary cathodes and lithium dendrite growth. Looking ahead, the field seems to be moving toward more multifunctional, better-performing, and more deliberately tailored separator designs. If these efforts pay off, they could remove some of the remaining bottlenecks holding lithium-ion batteries back, which would be good news for both EVs and grid-scale storage.

Keywords: lithium-ion battery separators, aluminum compounds, poly(vinylidene fluoride), polyimide

1. Introduction

Fossil fuels won't last forever, and the gap between how much energy we use and what we can supply keeps widening. Add in the environmental mess burning fossil fuels leaves behind, and it's clear why clean and renewable energy has moved from a nice-to-have to a must-have. The trouble is, older rechargeable battery types just don't pack enough energy — they're too heavy and bulky for applications that demand high power over long runtimes. Fig. 1 lays out how lithium-ion batteries stack up against other rechargeable chemistries on both volumetric and gravimetric energy density, and they come out looking pretty good [1].

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Beyond energy density, lithium-ion batteries last through a lot of charge–discharge cycles and hold up well over time. Safety has also gotten a lot better as engineers have worked through the design issues. These improvements have made lithium-ion batteries small enough and light enough for portable devices, and powerful enough for grid storage. They're pretty much everywhere now.

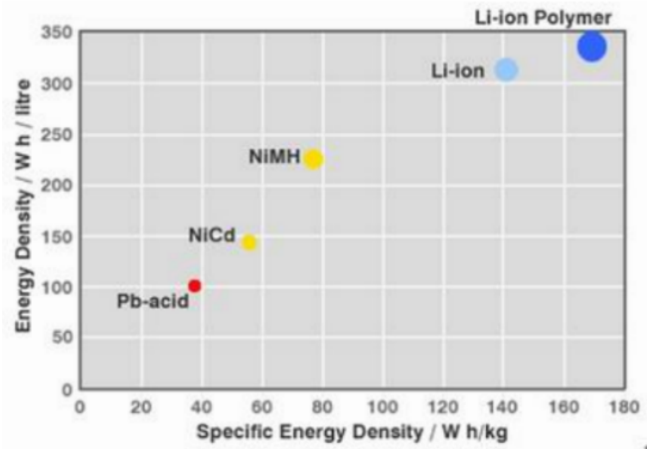


Figure 1. Comparison of the volumetric and gravimetric energy densities of several rechargeable battery technologies [1]

2. Overview of lithium-ion batteries

2.1. Structure and components of lithium-ion batteries

A lithium-ion cell has four main pieces: a cathode, an anode, a separator, and an electrolyte. None of them works alone — they all have to cooperate if the battery is going to charge and discharge reliably. A typical cell configuration is schematically illustrated in Fig. 2.

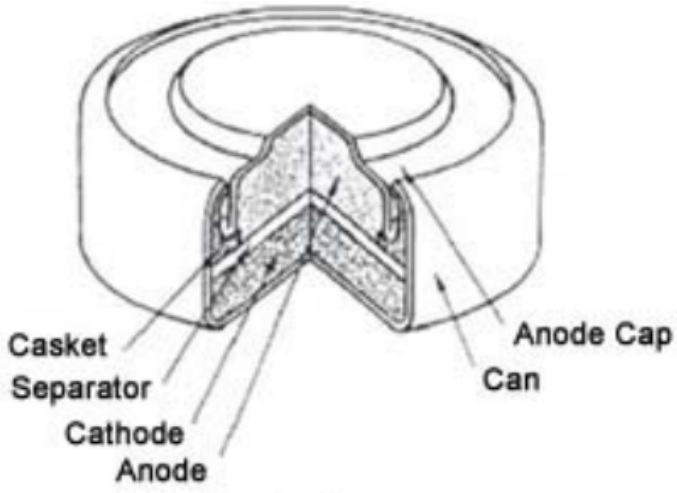


Figure 2. Typical battery configurations [2]

2.1.1. Cathode

The cathode supplies the lithium and largely sets the operating voltage of the cell. To get a high and stable voltage, cathode materials need a suitably high redox potential. They also have to survive the

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electrochemical environment without falling apart, and they should be cheap enough for mass production.

Lithium manganese oxide (LiMn2O4) is one such material. It has a spinel structure with the space group Fd3m. In that structure, Li+ ions sit in the tetrahedral 8a sites, Mn ions take the octahedral 16d sites, and O2− ions occupy the 32e sites, as shown in Fig. 3 [3].

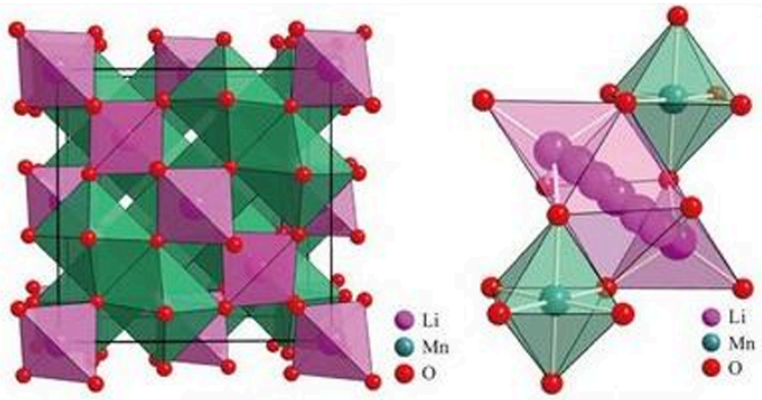


Figure 3. Crystal structure of LiMn2O4 [3]

2.1.2. Anode

The anode is where lithium gets stored and released during cycling. Early on, people tried using pure lithium metal as the anode, but it was too reactive and grew dendrites that caused short circuits — a real safety headache. So that idea was largely abandoned.

These days, carbon-based anodes, graphite in particular, dominate commercial cells. Silicon–carbon composites, which offer much higher theoretical capacity, are looking like a strong candidate for the next generation of anodes and could push energy density up even further.

2.1.3. Separator

The separator does several jobs at once. First and most obvious, it keeps the cathode and anode from touching — if they did, you'd have an internal short. At the same time, it has to let lithium ions pass through freely. On top of that, a good separator soaks up and holds onto electrolyte well, so the ion-conduction pathways stay continuous. It also needs to be chemically inert — no reactions with the electrolyte or electrodes. Get the pore structure right, and you can even slow down dendrite penetration.

A number of commercial separators also have a thermal shutdown trick built in. If the cell overheats, the micropores close up, ion transport drops to almost nothing, and the current stops — buying some time before things get dangerous.

Most commercial separators today are made of polyolefins — polyethylene (PE), polypropylene (PP), or the PP/PE/PP three-layer sandwich. They're cheap and work reasonably well at room temperature, but heat is their weak point: at elevated temperatures they shrink and distort, and they don't wet very well with the electrolyte. That's why modifying these materials and rethinking their structure is still a hot research topic [4].

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2.1.4. Electrolyte

The electrolyte is the highway lithium ions travel on between cathode and anode. It needs to be chemically and thermally stable, and it has to conduct ions well. Depending on how it's made, electrolytes fall into roughly three camps: organic liquid, gel polymer, and solid-state.

2.2. Operating principles of lithium-ion batteries

The whole idea is simple enough: lithium ions shuttle back and forth between the two electrodes. During charging, they leave the cathode, swim through the electrolyte-soaked separator, and lodge in the anode — electrons take the long way around through the external circuit to balance the charge. Discharging flips the process: lithium comes out of the anode, electrons flow through the circuit, and the ions head back to the cathode, as illustrated in Fig. 4 [5].

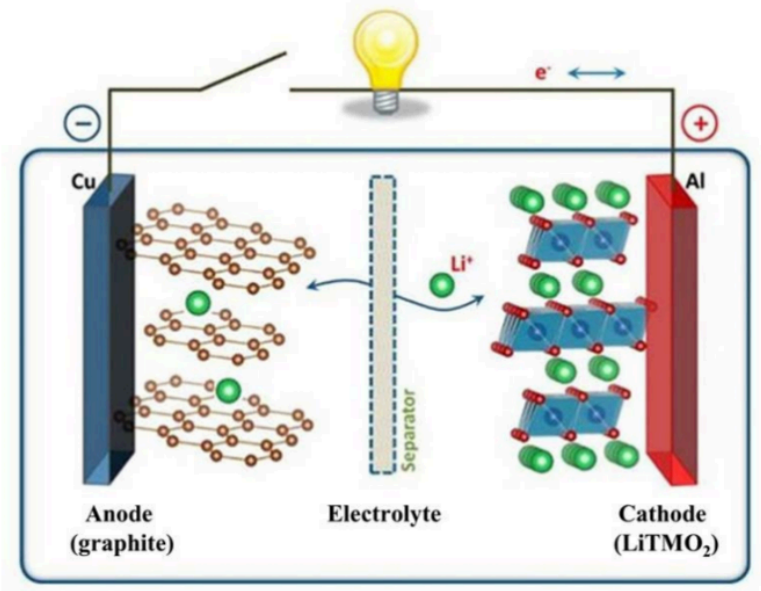


Figure 4. Schematic illustration of the operating principles of a lithium-ion battery [5]

3. Performance requirements for lithium-ion battery separators

The separator sits at the intersection of safety and performance, so it can't be optimized for just one thing. A bunch of requirements have to be met at the same time.

3.1. Thermal and electrical insulation

The separator has to block electrons. If the cathode and anode ever make contact — even through a tiny pinhole — you've got a short. So electrical insulation is non-negotiable. It also helps to have some thermal barrier capability: if a hot spot develops, a decent separator can slow the heat spread and keep things from cascading.

3.2. Appropriate porosity

Porosity is a balancing act. Too much and the film gets flimsy — it shrinks and distorts when heated, and the electrodes might touch. Too little and there isn't enough room for electrolyte, the ion

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pathways get choked off, and the battery slows down. The sweet spot gives you good electrolyte uptake, efficient ion transport, and enough structural integrity to last for thousands of cycles.

3.3. Electrochemical stability

Inside a cycling battery, the environment is harsh. The separator has to sit in it without falling apart — no side reactions with the electrolyte, no decomposition, no excessive swelling, no nasty by-products. It also needs to hold up across the full voltage window the cell sees. If the separator starts breaking down, internal resistance climbs and capacity fades, and that's the last thing you want after hundreds of cycles.

3.4. Good electrolyte affinity

If the electrolyte doesn't wet the separator quickly and evenly, you get dry patches where ions can't travel. That shows up as higher interfacial resistance, sluggish rate performance, and bad behavior in the cold. The separator also shouldn't swell too much or lose electrolyte over time — it needs to hold onto that liquid and stay compatible with whatever chemistry the battery is using.

3.5. Appropriate melting and shutdown temperatures

Ideally, a separator should shut down before it melts. Once the temperature crosses a threshold, the pores close, resistance spikes, and the electrochemical reaction grinds to a halt — that's the safety function. But you also want the film to keep its shape long enough after shutdown that the electrodes don't touch.

Differential scanning calorimetry (DSC) is the usual way to look at this. The melting peak in Fig. 5 shows how a PE separator behaves as it heats up [6].

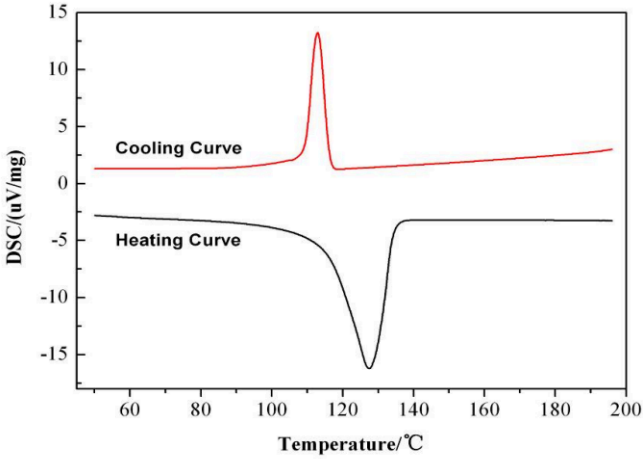


Figure 5. DSC curve of a PE separator [6]

Laman et al. [7] suggested a more direct approach: just watch how resistance changes with temperature. As you heat toward the melting range, the pores start closing and resistance shoots up. Keep heating, though, and the film collapses — resistance drops right back down because the electrodes finally touch. Fig. 6 shows exactly that.

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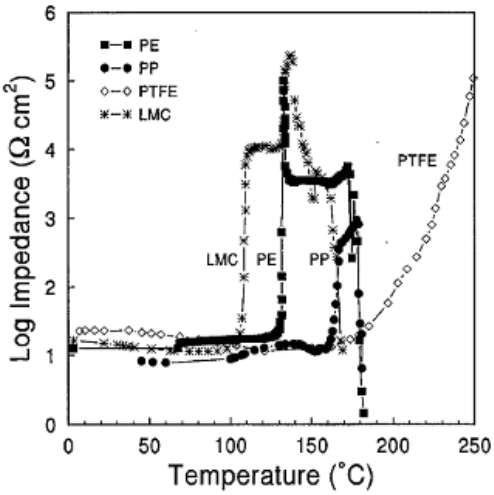


Figure 6. Temperature dependence of separator resistance [7]

4. Current research on lithium-ion battery separators

Batteries keep getting pushed harder — more energy density, faster charging, safer operation — and the old polyolefin separators are starting to look inadequate. Their main shortcomings, poor heat resistance and weak electrolyte wetting, are hard to ignore when cells run hotter and charge faster. So people have been busy modifying them. The main avenues are: mixing in inorganic ceramics, blending organic polymers, coming up with entirely new substrate materials, and rethinking the binders that hold everything together.

4.1. Inorganic composite modification

Dropping ceramic particles onto or into a polyolefin film is one of the oldest tricks in the book, and it still works. The particles boost heat resistance, improve wetting, and add mechanical strength. Al_2O_3 and SiO_2 are the usual choices, though some carbon fillers have been tried too. The key is picking the right material and loading it properly — too little does nothing, too much gums up the works.

Shi et al. found that coating thickness mattered a lot for the Al2O3 case. A thin layer helped heat resistance, but pile it on too thick and ion transport suffered, dragging down electrochemical performance [8]. Zhao et al. took a different tack: they coated PE separators directly with Al_2O_3 ceramic powder. The 6 μm coating they came up with showed less thermal shrinkage and higher ionic conductivity than bare PE, and cycling got better too [9].

4.2. Organic composite modification

The organic route is more about polymer chemistry — blending, grafting, and coating with other polymers to change how the separator behaves. It's probably the most actively studied area right now.

Polyolefins are notoriously hard to wet. Throw in something like polyacrylonitrile (PAN) or poly(methyl methacrylate) (PMMA), and the surface gets more polar, the electrolyte spreads faster, and ions move more freely. Tweaking the chain flexibility also helps the film absorb the mechanical stresses that build up during cycling.

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Li et al. made a porous poly(vinylidene fluoride) (PVDF) film about 31 μm thick using phase inversion [10]. Compared with a commercial Celgard 2500 PP separator, it soaked up a lot more electrolyte — porosity around 56% and uptake near 200% — and held up well at 5 V.

4.3. Development of novel separator materials

Sometimes tweaking polyolefins isn't enough, and you need a new material altogether. The idea is to design polymers that simultaneously give you better wetting, better heat resistance, and better ion flow.

PAN/PVDF blends are a case in point. PAN wets well but isn't the most stable electrochemically; PVDF is stable but doesn't wet as nicely. Mix them in the right ratio, and you get the best of both. These blended films can hold a lot of electrolyte while keeping their pores and mechanical properties intact.

Gopalan et al. [11] spun a PAN/PVDF solution into a nonwoven fabric by electrospinning, then soaked it in liquid electrolyte to get a working polymer electrolyte system.

4.4. Development of binders for lithium-ion battery separators

Binders get overlooked, but they're what keeps a coating or composite layer stuck to the separator. PVDF is the go-to binder, but it gets soft and loses its grip when things heat up — not ideal for batteries that need to be safe at high temperatures. So there's a push to find binders that can handle the heat while staying tough and adhesive.

Cho et al. [12] took poly(amic acid) — made by reacting pyromellitic dianhydride with oxydianiline — and used it as a coating. Heating it up drove off the water and turned it into polyimide (PI). PI is remarkable for its heat resistance, mechanical strength, and chemical stability, which makes it a natural fit for separator and interface materials. China came to PI research a bit late, but the field has grown quickly and now covers films, plastics, adhesives, and more.

4.5. Separator manufacturing processes

Commercial microporous polyolefin separators are made mainly two ways: the wet route, which leans on thermally induced phase separation (TIPS), and the dry route, which stretches a melt. Nonwoven fabrics, on the other hand, usually come out of electrospinning.

TIPS was originally proposed by Castro [13]. The idea is to melt a polyolefin with a high-boiling-point diluent at a temperature above the polymer's melting point, let it cool so the two phases separate, and then wash out the diluent to leave behind a network of interconnected micropores. Fig. 7 walks through the steps [14].

Lloyd et al. [15] dug into the phase-separation physics, thermodynamics, and crystallization kinetics of TIPS, and looked at how the choice of diluent, cooling rate, and additives all shape the final membrane. One advantage of TIPS is that it works well for highly crystalline polymers that won't dissolve at room temperature, and you have a lot of knob twiddling room. By changing the molecular weight, diluent, cooling rate, nucleating agent, or extraction solvent [16], you can dial in different pore sizes, porosities, and structures.

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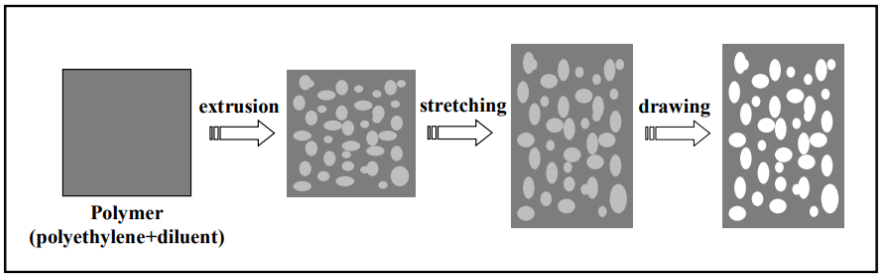


Figure 7. Schematic illustration of polyethylene microporous membrane formation via thermally induced phase separation [14]

5. Future directions for lithium-ion battery separators

Traditional separators basically do two jobs: keep the electrodes apart and let ions through. That used to be enough, but today's high-energy, high-safety batteries expect more [17]. Functional separators are the direction things are headed.

A functional separator still separates the electrodes and conducts ions, but it also takes on extra jobs — cleaning up interfacial side reactions, trapping transition metals, blocking dendrites, or surviving higher temperatures. Smart structural design and surface chemistry are opening up ways to push both performance and safety at once.

Nickel–cobalt–manganese ternary cathodes are everywhere in high-energy cells because they pack a lot of specific capacity. Adding Mn helps cut costs and stabilize the structure, so it's a key ingredient in those cathode recipes [18].

The catch is that, especially at high temperatures or high voltages, transition metals leach out of these cathodes. The dissolved ions drift through the separator, reach the anode, and mess up the solid electrolyte interphase (SEI). Every time the SEI breaks and reforms, internal resistance goes up and capacity fades — and the safety picture gets worse. So separators that can grab, block, or pin down those metal ions are getting a lot of attention, because they could make ternary-cathode batteries last longer and run hotter without falling apart.

Dendrites are the other safety headache. Kai Liu's group at Stanford tackled this with a three-layer separator that sandwiches a sheet of SiO_2 nanoparticles in the middle. When a lithium dendrite pushes toward the separator, it hits that functional layer and reacts with the SiO_2, which slows it down and keeps it from punching all the way through. That kind of active-safety design — where the separator itself fights back instead of just passively sitting there — looks like a promising way to deal with dendrite penetration and improve both safety and cycle life.

6. Conclusions

This review has walked through where polymer separators for lithium-ion batteries stand right now. As the energy transition rolls on and storage demand keeps climbing, lithium-ion batteries are only going to matter more for EVs and grid storage. The separator, though easy to ignore, is the component that ties safety and performance together — it has to insulate electrically, hold up mechanically, tolerate heat, stay stable electrochemically, and wet well with the electrolyte, all at the same time.

Today's polyolefin separators still fall short, mainly on heat resistance and wetting. People have responded with inorganic coatings, organic polymer blends, new substrate materials, and better binders — these are the four main levers right now.

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Looking further out, functional separators that can adsorb ions, regulate interfaces, and actively respond to safety threatens look like the most promising path forward, especially for problems like transition-metal dissolution and dendrite growth. The next generation will probably be more multifunctional, better overall, and more precisely tailored to specific battery chemistries. If materials design, interface engineering, and structural optimization can work together, separators should get safer, last longer, and help batteries use their energy more efficiently — which is exactly what EVs and large-scale storage need.

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