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Research Progress of Thermal Conductive Materials for Lithium-ion Batteries

Zhirui Zheng

Fuzhou No.8 High School, Fuzhou, China
zray51243@gmail.com

Abstract. Fast charging easily leads to heat accumulation inside batteries, resulting in large temperature differences among cells, material aging and even thermal runaway risks. As the core of thermal management, thermal conductive materials directly determine the temperature uniformity and heat dissipation performance of battery modules, and are critical to guarantee the safe and long-term operation of energy storage devices. This paper systematically reviews the respective characteristics of three types of thermal conductive media for lithium-ion batteries: metal-based, carbon-based and phase change materials. Metal-based materials feature high thermal conductivity and high mechanical strength yet suffer from heavy weight and lack heat storage capacity; carbon-based materials are lightweight with excellent in-plane temperature homogenization, but their through-plane thermal conductivity is poor and high-end fillers cost a great deal; phase change materials realize constant-temperature heat storage while their intrinsic thermal conductivity is extremely low. Studies indicate that single-component thermal conductive materials all possess obvious drawbacks. Carbon-metal and phase change composite systems can complement multiple properties and have become the mainstream improvement schemes at present. In the future, the industry will carry out technical research focusing on three directions: multi-phase composite modification, low-cost large-scale preparation and integrated coupling of thermal management. The summary and review in this paper can provide important theoretical references for power battery PACK design, optimization of energy storage temperature control schemes and development of novel thermal conductive fillers.

Keywords: lithium-ion battery, thermal conductive material, thermal management, phase change material

1. Introduction

Promoted by the innovation of global energy structure and domestic Dual Carbon goals, the traditional energy system is gradually transforming to clean electric energy, and the market scale of new energy vehicles, large-scale energy storage equipment and various portable electronic devices keeps expanding. Lithium-ion batteries exhibit outstanding advantages including high energy density, long cycle life and low self-discharge loss, which make them the core carriers in the field of

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electrochemical energy storage and indispensable key materials for the implementation of low-carbon development routes [1].

With the popularization of high-voltage fast charging technology, the energy capacity of single cells is continuously improved. Lithium-ion batteries continuously generate various forms of heat during charging and discharging. If accumulated heat cannot be quickly dissipated, significant temperature differences will form among cells inside the module, accelerating the aging of electrode active materials and shortening the overall service life of batteries. Under severe working conditions such as continuous high-power charging and high ambient temperature, continuous heat accumulation will trigger internal chain exothermic reactions, further causing swelling, combustion and other potential safety hazards, hindering the large-scale commercialization of lithium-ion batteries. All active and passive heat dissipation schemes on the market rely on thermal conductive media for heat transfer. The heat transfer capacity and lightweight degree of thermal conductive materials directly determine the working performance of the whole temperature control system. Therefore, developing high-performance thermal conductive materials matching high-power cells is a core problem urgently to be solved in the power battery industry.

At this stage, thermal conductive media applied to lithium-ion battery heat dissipation in the industry are mainly divided into three categories: metal-based, carbon-based and phase change materials, which differ greatly in heat transfer mechanisms and inherent performance defects [2]. Metal thermal conductive materials deliver high heat transfer efficiency and stable mechanical structure, but their excessive weight fails to meet the lightweight design requirements of whole vehicles. Carbon materials are lightweight and perform well in in-plane heat transfer, yet they may induce short circuits due to conductivity and the modification and processing cost of high-end raw materials remains high. Phase change materials can store heat through phase transition to stabilize temperature, but their intrinsic thermal conductivity is weak, which cannot satisfy the heat dissipation demand of high-power equipment alone. Every single type of material has unavoidable performance defects. Preparing composite thermal conductive materials by blending metal, carbon fillers and phase change matrices has become a mainstream research direction in recent years. However, composites produced with different proportions and preparation processes differ remarkably in long-term cycle stability and industrial production cost, and existing studies lack complete and systematic horizontal comparative reviews [3].

To fill the gaps in current research, this paper systematically sorts out the heat transfer mechanisms, merits and demerits as well as practical application scenarios of the three kinds of thermal conductive materials, summarizes the technical limitations existing in the development and application of thermal conductive materials, and predicts the future industrial development trend from three dimensions: multi-component composite modification, low-cost mass production technology and integrated design of thermal management systems. The paper can offer theoretical references for the development of new thermal conductive fillers and optimization of lithium-ion battery heat dissipation structures.

2. Classification, advantages and disadvantages of battery thermal conductive materials

Lithium-ion batteries continuously generate heat during the whole charge-discharge cycle. Long-term heat accumulation accelerates battery aging and may even induce thermal runaway under severe conditions. Bernardi established a universally accepted battery heat generation analysis model in 1985, which classifies heat produced during battery operation into four types: Joule heat, polarization heat, reversible reaction heat and side reaction heat. This heat generation model still serves as the basic theoretical tool for analyzing battery temperature rise [4]. Joule heat is

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irreversible heat generated in line with Joule's law when current passes through internal resistance regions of batteries. Local high-temperature zones are easily formed inside cells under fast charging and high-rate discharge conditions. Polarization heat originates from potential loss caused by resistance to lithium-ion migration, and temperature rise becomes more obvious with larger discharge current. Reversible reaction heat comes from enthalpy change during lithium intercalation and deintercalation, absorbing heat in charging stages and releasing heat in discharging stages, and the heat generation amount depends on the real-time state of charge of batteries. When the internal cell temperature exceeds 80 °C, a large number of side reactions such as electrolyte decomposition, abnormal thickening of SEI film and lithium precipitation on negative electrodes will occur, releasing massive extra heat. Sustained chain exothermic reactions will ultimately lead to cell swelling, fire and even explosion [5]. Traditional air cooling has limited heat exchange capacity, while thermal conductive materials can effectively reduce contact thermal resistance between components and balance the overall temperature of modules. The three mainstream thermal conductive materials are analyzed separately as follows.

2.1. Metal-based materials

Metal-based thermal conductive materials take copper, aluminum, metal foam, gallium-indium-tin liquid metal alloy as main heat transfer carriers, transferring heat through directional movement of free electrons inside, and their heat transfer rules conform to Fourier's law of heat conduction, exhibiting superior heat transfer performance to all non-metallic thermal conductive media. Measured data from literatures show that the thermal conductivity of pure copper and pure aluminum reaches 401 W/(m·K) and 237 W/(m·K) respectively [6], which can rapidly dissipate local high temperature of cells and effectively inhibit the outward spread of thermal runaway. Metal substrates possess mature forming technology; aluminum alloy shells can simultaneously realize cell protection and heat dissipation, and waste metals enjoy high recycling rate. Aluminum water cooling plates and metal foams have sufficient heat exchange area, while gallium-indium-tin liquid metal features ultra-low contact thermal resistance and fits filling of compact battery packs with narrow spaces.

Nevertheless, copper and aluminum have high density, and mass application will increase the overall weight of vehicles, conflicting with lightweight design ideas. Metals are conductive by nature, and direct contact with cells may trigger internal short circuits, requiring additional insulating coatings and raising processing costs. Metals are prone to corrosion under humid and liquid leakage environments, resulting in continuous attenuation of heat dissipation performance after long-term cycles. Copper raw materials also carry relatively high procurement cost. Moreover, metals only possess heat conduction function without heat storage and constant-temperature capacity, making it difficult to suppress temperature peaks generated by continuous high-rate discharge. At present, aluminum water cooling plates and aluminum alloy shells are widely matched with liquid cooling systems of ternary lithium batteries for passenger vehicles. Metal foams compounded with phase change materials are applied to high-power energy storage modules, and liquid metal thermal interface materials are mostly used in high-end fast-charging pouch batteries. Pure metals are generally only adopted as underlying heat dissipation substrates and rarely used alone for heat dissipation of lightweight consumer electronics.

Metal foam acts as a common skeleton material for metal-phase change composite thermal conductive systems. Its internal three-dimensional interconnected porous structure can construct continuous heat conduction channels, and the pore structure can adsorb phase change matrices. The micro-structure of metal foam intuitively reflects the structural advantages of this material.

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2.2. Carbon-based materials

Carbon-based thermal conductive materials adopt graphite, graphene and carbon nanotubes as heat transfer functional components, transferring heat via phonon vibration inside crystal lattices with outstanding in-plane heat transfer performance. They are often processed into thermal conductive films and thermal conductive gaskets, or used as fillers to improve the heat transfer capacity of polymer and phase change matrices. Relevant test results demonstrate that the maximum in-plane thermal conductivity of graphene can reach 5300 W/(m·K), with material density ranging only from 0.1 to 1.8 g/cm³, presenting prominent lightweight advantages [7]. Graphite films have good flexibility and can closely fit the surface of pouch batteries to reduce interfacial contact thermal resistance, adapting to compact assembly space of mobile phones and energy storage pouch cells. Directionally arranged carbon materials can balance the internal temperature of modules, narrow temperature differences among cells and improve the consistency of battery charge and discharge. A small dosage of carbon nanotubes and graphene can significantly boost the thermal conductivity of silica gel and phase change materials, which makes them universal modified fillers in the industry.

However, carbon materials show obvious anisotropy in heat conduction, and their thermal conductivity along the vertical direction of lamellae is less than 10 W/（m·K）, requiring metal skeletons for auxiliary vertical heat dissipation [8]. Pure carbon is conductive, and direct application may induce micro-short circuits of cells; compounding with insulating matrices will lower the overall thermal conductivity. High-quality graphene involves complicated preparation processes with high mass production cost, and graphite films are brittle, unable to bear mechanical protection of cells independently. In practical applications, graphite thermal conductive films are mostly used for heat dissipation of consumer electronics, graphene composite gaskets fit the temperature homogenization demand of new energy pouch cells, carbon nanotubes are commonly added into various thermal conductive matrices as modified fillers, and graphene foams are applied in thermal management of high-end energy storage equipment. Carbon-based materials are rarely used as the main heat dissipation structure alone and mostly compounded with metals and phase change materials.

2.3. Phase change materials

Phase change materials are mainly divided into two categories: organic paraffin and inorganic hydrated salts, realizing constant-temperature control of cells through heat storage and release via solid-liquid phase transition, yet their intrinsic thermal conductivity is weak, usually requiring doping of carbon and metal fillers to strengthen heat transfer efficiency [9]. Existing experimental data prove that the thermal conductivity of pure paraffin only ranges from 0.2 to 0.3 W/(m·K), which cannot meet the heat dissipation demand of high-power batteries independently [10]. Such materials do not need supporting fans or liquid cooling pumps and can adjust temperature autonomously to reduce energy consumption of the whole thermal management system, suitable for low-speed electric vehicles and household energy storage scenarios. After cells are fully wrapped by phase change materials, the temperature difference of modules can be controlled within 3 ℃, effectively improving uneven temperature distribution during fast charging. Shaped phase change materials can flexibly match various cell shapes, possess better thermal stability than electrolytes and slow down the spread of thermal runaway.

However, liquid paraffin has leakage defects, and shaping modification will increase production cost. Inorganic hydrated salts suffer from supercooling and phase separation after long-term cycles, leading to continuous attenuation of heat storage capacity. Phase change materials heat storage has

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an upper limit; once completely melted, they lose temperature control effect and must cooperate with air cooling or liquid cooling for heat dissipation. In engineering scenarios, shaped composite phase change materials are applied to low-speed electric vehicles and small energy storage equipment, carbon/metal foam composite phase change materials serve as auxiliary heat dissipation media for passenger vehicles, and inorganic hydrated salts fit constant-temperature energy storage power stations. Pure phase change materials cannot be separately used in fast-charging power batteries and need to couple with water cooling plates and carbon-based temperature homogenization films [11].

Coating cells with shaped phase change materials is the most basic implementation scheme of passive thermal management at present, and the heat dissipation structure of the whole module clearly shows the layout form and working logic of phase change materials.

2.4. Comprehensive comparison and summary

Metal-based materials deliver fast heat conduction and high mechanical strength, suitable for high-power liquid cooling heat dissipation, yet they are heavy and lack heat storage and constant-temperature functions. Carbon-based materials are lightweight with excellent lateral temperature homogenization, but their vertical heat conduction is weak and high-end fillers cost much. Phase change materials realize zero-energy constant temperature, while their intrinsic heat conduction capacity is poor and they can only act as auxiliary temperature control media. All single-component materials have irreparable performance defects. Carbon-metal and phase change-carbon composite thermal conductive materials have become core research directions in recent years, which can integrate multiple performance advantages through multi-component complementation and simultaneously satisfy multiple demands including efficient heat conduction, module temperature homogenization, lightweight design and constant-temperature heat storage [12].

3. Prospect of battery thermal conductive materials

Power batteries keep iterating toward high energy density and ultra-high fast charging rate, and single-component thermal conductive materials can no longer meet the comprehensive temperature control demands of battery packs for new energy vehicles, energy storage supporting high-power charging piles and large-scale energy storage power stations. Future research and development of thermal conductive materials will focus on developing multi-phase composite thermal conductive systems, optimizing comprehensive performance by reasonably adjusting filler proportions, simplifying preparation processes to reduce material cost, and collaboratively designing thermal conductive components with battery PACK and water-cooling structures to improve the thermal safety and cycle life of lithium-ion batteries from an overall perspective. AI simulation algorithms can rapidly screen optimal proportions of composite fillers and simulate the full-domain thermal field distribution of batteries to greatly shorten the research and development cycle of new materials; digital twin technology can also carry out integrated intelligent optimization of battery heat dissipation structures. This paper systematically sorts out the performance defects and composite modification advantages of three types of thermal conductive materials and summarizes existing research gaps, which can provide clear theoretical basis for the design of battery thermal management schemes and development of novel thermal conductive fillers. Continuous improvement of relevant technologies can effectively reduce thermal safety risks of lithium-ion batteries and boost the large-scale popularization of clean energy storage equipment.

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4. Conclusion

This paper systematically reviews the heat transfer mechanism, comprehensive performance and engineering application scenarios of three kinds of lithium-ion battery thermal conductive materials: metal-based, carbon-based and phase change materials. Combined with Bernardi's classic heat generation model, it analyzes the generation rule of various heat sources of batteries, and draws core conclusions through horizontal comparison: Firstly, single-component thermal conductive materials all have obvious performance drawbacks. Metals are heavy, carbon-based materials deliver poor vertical thermal conductivity, and pure phase change materials have extremely low thermal conductivity, failing to adapt to high-demand working conditions such as high-power fast charging of charging piles and long-endurance vehicle power batteries. Secondly, carbon-metal and phase change composite thermal conductive materials can integrate the advantages of various substrates, balancing heat dissipation speed, lightweight design and constant-temperature heat storage capacity, which serve as core schemes to solve thermal runaway and narrow temperature differences among cells. Thirdly, current composite thermal conductive materials still suffer from high mass production cost, insufficient long-term cycle stability and imperfect research on adaptation to different scenarios. Existing literatures lack special comparative analysis targeting energy storage of charging piles and power batteries for commercial vehicles, forming research gaps.

From the perspective of research value, this paper completely summarizes the parameters, merits, demerits and application limitations of three kinds of substrates, which can provide clear material selection basis for new energy enterprises to develop vehicle battery PACK and temperature control components matched with high-power charging pile energy storage, as well as reference ideas for proportion and process optimization of novel composite thermal conductive fillers in laboratories. Facing the future development of the industry, follow-up researches should focus on low-cost carbon composite fillers and improved shaping technology of phase change materials, and promote the integrated design of thermal conductive materials with whole-vehicle water cooling and heat dissipation of energy storage cabinets, so as to further reduce thermal safety hazards of new energy vehicles, public charging piles and grid energy storage equipment and facilitate the large-scale popularization of clean energy.

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