



Review article

Materials insights into low-temperature performances of lithium-ion batteries



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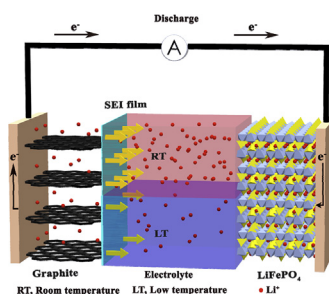
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HIGHLIGHTS

- Discusses main limitations of low-temperature performances for lithium batteries.
- Reviews methodology for improving low-temperature lithium batteries.
- Analyzes advanced materials for low-temperature lithium battery performances.

GRAPHICAL ABSTRACT



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ABSTRACT

Lithium-ion batteries (LIBs) have been employed in many fields including cell phones, laptop computers, electric vehicles (EVs) and stationary energy storage wells due to their high energy density and pronounced recharge ability. However, energy and power capabilities of LIBs decrease sharply at low operation temperatures. In particular, the charge process becomes extremely sluggish at temperatures below $-20\text{ }^{\circ}\text{C}$, which severely limits the applications of LIBs in some cold areas during winter. Extensive research has shown that the electrolyte/electrode composition and microstructure are of fundamental importance to low-temperature performances of LIBs. In this report, we review the recent findings in the role of electrolytes, anodes, and cathodes in the low temperature performances of LIBs. Our overview aims to understand comprehensively the fundamental origin of low-temperature performances of LIBs from a materials perspective and facilitates the development of high-performance lithium-ion battery materials that are operational at a large range of working temperatures.

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1. Introduction

To mitigate the energy crisis and environmental impact of the

fossil-fuel based economy, energy storage technology has been an important component of current energy strategies [1]. Lithium-ion batteries (LIBs) represent a promising energy storage technology for the integration of renewable resources and have been efficient power sources for a large range of applications owing to a number of advantages including high energy density, flexible and light-weight design, and long lifespan [2–8]. Rechargeable LIBs have been available in many commercial applications, such as cell

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phones, laptop computers, HEVs, EVs, and stationary energy storage wells. However, the poor low-temperature performances of LIBs limit the applications in cold conditions, such as certain defense and space applications [9,10], and transportation systems, including plug-in hybrid electric vehicles (PHEVs) and electric vehicles (EVs) [11–13]. There is a sharp loss in the energy and power capabilities as the operating temperature is below 0 °C [13–15]. In addition, under –20 °C the lithiation process is typically slower than the delithiation process in the electrode [16]. To avoid the cell damage at low temperatures, the current common method is to increase the working temperature by heating. Nevertheless, this method reduces the energy efficiency, and does not allow for large-scale applications.

In this review, we firstly describe the main existing limitations of LIBs at low temperatures, followed by discussion on the recent improvement in low-temperature performances by developing novel electrolyte, anode and cathode materials. Last, we conclude by providing prospectives on future developments in improving the low-temperature performances of LIBs. This report is focused on elucidating main limitations of low-temperature performances and aims to provide an insightful guidance for enhancing the overall low-temperature performances of LIBs.

2. Main limitations for low-temperature performances of lithium-ion batteries

LIBs lose most energy and capacity as the temperature is below –10 °C [13,14]. Zhang et al. [16] reported that graphite only retains 12% of room temperature capacity at –20 °C. The energy density of 18650 Li-ion batteries decreases from ~100 Wh l⁻¹ to ~5 Wh l⁻¹, and the power density decreases from ~800 W l⁻¹ to 10 W l⁻¹ as the operating temperature decreases from 25 °C to –40 °C [17]. In addition, due to the high charge-transfer resistance at the charged state LIBs can be efficiently charged up only as the temperature is higher than –10 °C [13,18]. Furthermore, the mass transport is hindered at low temperatures due to the increase of inactive lithium ions, giving rise to dendrite formation and growth. This dendritic electrode could lead to battery explosion and damage due to short circuits [19,20]. The poor low-temperature performance hinders substantially further application of LIBs. Here, we discuss the main low-temperature performance limitations of LIBs in the charge/discharge process and structure evolution.

2.1. Main limitations on charge/discharge process

LIBs directly convert chemical energy into electric energy upon discharge. In this process, lithium ions are deintercalated from the layered graphite intercalation host, pass across the solid electrolyte interface (SEI), diffuse through the electrolyte and are intercalated between the LiCoO₂ layers in the cathode [7]. Charge reverses this process and converts electric energy into chemical energy. In the first cycle, a preliminary SEI layer is formed, which has been considered as an essential process for the fabrication of LIBs. The SEI layer protects electrodes and only permits the transport of lithium-ions [21]. The efficient transport of Li⁺ and electrons ensures the efficient operation of LIBs. However, low temperatures impede all the key steps, such as the charge-transfer at electrode/electrolyte interface [22], and Li⁺ transport in the SEI, the electrolyte and the electrodes [13,22,23], leading to the loss in power capability. The commercial electrolyte, LiPF₆ typically dissolves in ethylene carbonate (EC)/ethylmethyl carbonate (EMC) and also freezes at the temperature below –30 °C. Consequently, the slow ionic conduction in such an electrolyte is the major challenge for efficient operation of low-temperature LIBs [9,23]. Wang et al. [24]

also suggested that the resistance of SEI increased by a factor of over 27 at –30 °C and largely limited the low-temperature performance. Huang et al. proposed that the poor performance was caused by the sluggish lithium-ion diffusion in the carbon anode at a low temperature [13]. This indicates that the lithium insertion reaction was kinetically limited at a low temperature. This proposition was then verified by Zhang et al. [15]. The resistance of the batteries is predominated by the charge-transfer resistance R_{ct} as the temperature is below –20 °C. R_{ct} reflects the kinetics of the cell electrochemical reaction. A small R_{ct} value corresponds to a fast kinetics of the electrochemical reaction [15]. In fact, to ensure the operation of the electrolyte at low temperatures, the charge-transfer at the electrode/electrolyte interface must be efficient enough [14,16,25]. Zhang et al. [15] studied the low-temperature performances of LIBs *via* electrochemical impedance spectroscopy (EIS). Typically, the impedance spectra of LIBs is composed of two overlapping semicircles at high and medium frequency regions, and a straight line at low frequency region. The high and medium frequency regions represent the resistances of SEI (R_{sei}) and electrode/electrolyte interface (R_{ct}), respectively, and the low frequency region represents the Warburg impedance associated with Li⁺ diffusion in the electrode [12,24]. R_b is the bulk resistance of the cell, including the resistances of the electrolyte, separator, and electrodes. Fig. 1 shows that at 3.87 V and 3.45 V, R_{ct} increases substantially at low temperatures [15]. Although R_{sei} and R_b increase with decreasing temperature, the percentage of R_{ct} vs. the total cell resistance achieves nearly 100% as the temperature is below –20 °C. The high charge-transfer resistance at low temperatures results in a high electrochemical polarization [15,17,24,26]. In addition, low-temperature mass transport limitation at the electrode/electrolyte interface increases the deposition of metallic phases on carbon anode, giving rise to concentration polarization [13,14,27,28]. As the deposited lithium is not uniform, the fragile SEI film could be broken under mechanical stress. Continuous deposition of lithium also gives rise to the formation of lithium whiskers that could be easily cut at low temperatures, leading to the accumulation of dendrite [20]. In short, the lithium-ion conductivity is decreased in electrode/electrolyte interface, electrolyte, SEI, anode, and cathode at low temperatures. In all these processes, the low Li⁺ transfer rate at electrode/electrolyte interface caused by low-kinetics electrochemical reaction is the rate-determining step in low-temperature charge and discharge.

The delithiated state has a larger R_{ct} compared with the lithiated state in a Li/graphite cell, and it has been shown that the charge process is slower than the discharge process at –20 °C and –30 °C [16]. In the anode discharge process, lithium-ions leave LiC₆, and C₆ forms [4]. An exothermic electrochemical reaction occurs in this discharge process while the charge experiences an endothermic electrochemical reaction. Lowering the temperature hinders the endothermic electrochemical reaction due to the lack of heat. Thus, lowering the temperature could result in the electrochemical equilibrium moving to lithiated state, making the charge process difficult.

2.2. Main limitation on structure evolution

The structure of materials affects significantly the performance of the batteries. The atomic resolution image in Fig. 2b shows that pristine Li-[Li_{0.2}Ni_{0.2}Mn_{0.6}]O₂ cathode exhibits well-defined layered structure, with a stacking sequence of Li/O/TM (transition metal)/O [29]. Li-[Li_{0.2}Ni_{0.2}Mn_{0.6}]O₂ transforms from layered phase to spinel phase due to the extensive removal of lithium ions during charging (Fig. 2b,c,f). At the same time, part of the lithium ion diffusion channels are blocked by the migration of TM ions to Li layers (Fig. 2f), which leads to capacity degradation. After 300

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