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Review: Performance Of Li-Ion Battery At Low Temperature: Challenges And Mitigation For Military Applications

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Abstract

1. Lithium ion batteries are used for enormous application across the globe. Its being widely used in armies also. The use of lithium batteries at low temperature has poor performance, whether charging or discharging, and may have an impact on life. Under low temperature conditions, the performance of lithium battery will decline, such as prolonged charging time, reduced charge and discharge, smaller battery capacity and faster power loss, which will affect the driving mileage of new energy vehicles. Low temperature conditions significantly reduces the battery power hence it is crucial to understand the various factors responsible for degradation of LIBs at various temperatures. For example a fully charged laptop in one place , if brought to a high altitude place immediately shows 10% charge just reaching there. In this paper the generic effects of both high and low temperatures on LIBs are covered and more focus will be on improving the performance of batteries in extreme cold climates. Low temperature LIBs are designed to operate in harsh environments where the temperature is below 0°C.

I. Introduction

2. In military operations, soldiers often operate in remote locations with various climatic conditions with temperatures ranging from -40^{o}C to 50^{o}C and where access to a reliable power grid is not possible. In such scenarios, batteries are the only practical solution for powering electronic devices such as radios, GPS units, night vision devices and other essential equipment. The use of batteries also provides operational security, as soldiers can operate silently without the need for noisy generators or other power sources. Furthermore, the reliability of batteries is crucial in military applications, as equipment failures due to power issues can have serious consequences. Due to these reasons, the military invests heavily in battery research and development.

3. As compared to other batteries, Lithium Ion Batteries (LIBs) have various advantages like high energy density, stable performance, long cycle life, low discharge rate etc which make it most reliable and portable power source for a wide range of military equipments. LIBs can be dominant power source for applications ranging from portable electronic devices to electric vehicles. However it is observed that the performance of LIBs degrades at negative temperature due to low ionic conductivity of bulk electrolyte because of increased solid electrolyte interface. Under low temperature environment, the electrolyte viscosity of LIBs increases thus reducing the migration speed of lithium ion which further relatively decreases its discharge capacity.

4. Low temperature conditions significantly reduces the battery power hence it is crucial to understand the various factors responsible for degradation of LIBs at various temperatures.
In this paper the generic effects of both high and low temperatures on LIBs are covered and more focus will be on improving the performance of batteries in extreme cold climates. Low temperature LIBs are designed to operate in harsh environments where the temperature is below 0°C. These batteries are essential for a wide range of applications as mentioned above. The challenges associated with operating LIBs in low temperatures include reduced performance, lower energy density, shorter lifespan, and reduced capacity. The low temperature can cause the electrolyte in the LIB to freeze, which reduces the ability of the battery to store and release energy. It can also lead to increased impedance, reduced rate capability, and increased self-discharge.

Generic effects of both Low Temperatures on LIBs

5. The experimental results showed that the energy density of Li-ion battery decreased from 100Wh/kg at 25^{o}C to 5kWh/kg at -40^{o}C and the power density reduces from 800Wh/kg at 25^{o}C to 10Wh/kg at -40^{o}C [1-4]. It

was observed that the commercially available LIBs having ethylene carbonate (EC) as electrolyte and graphite as anode, experience poor performance at temperature below zero degrees Celsius.

6. The main reasons for decrease in capacity of LIBs in low temperature are as follows:-

(i) The ionic conductivity of bulk electrolyte reduces substantially.
(ii) Internal resistance of electrolyte increases, Solid Electrolyte Interface (SEI).
(iii) Poor charge transfer kinematics.
(iv) The diffusion of Li-ions throughout bulk electrodes becomes slow.

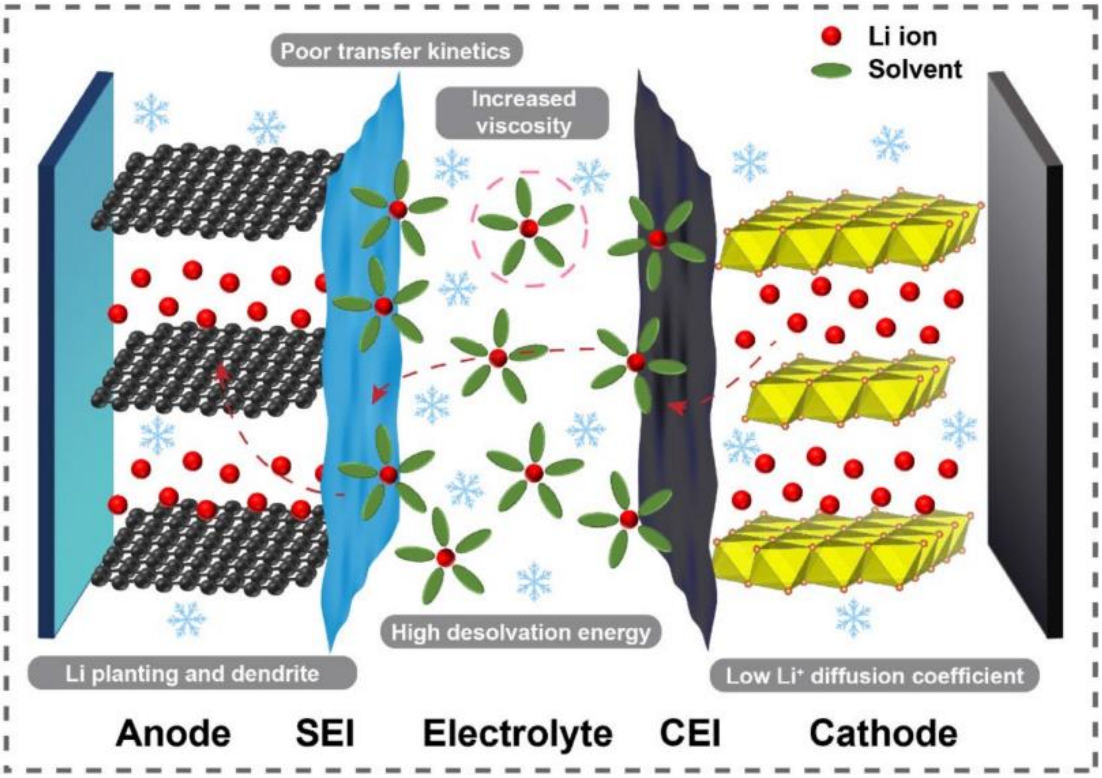

Fig 1: Problems encountered by LIBs at low temperature

7. At cold temperature, the intercalation of Li-ions on the graphite electrode and the Li plating reaction proceeds simultaneously. Under cold conditions the diffusion of Li ions in graphite is inhibited and the conductivity of electrolyte is reduced, as a result, the intercalation rate also decreases. This results in Li plating occurring more easily on graphite surface and this accumulation leads to an increase in the thickness of Solid Electrolyte Interface (SEI). Assuming that the voltage remains constant and discharge current decreases, the power output of the battery will also reduce thus lowering the output. The formation of SEI layer is inevitable on the anode material because their lithiation potential lies outside the thermodynamic stability window of typical organic electrolytes. The primary mechanism of SEI formation occurs during the oxidation and reduction of the electrolyte at the electrode surface during the first few charge discharge cycle. At low temperature, large amount of Li-ions are consumed in the first cycle forming a relatively dense SEI due to lower solubility and worse kinetic characteristics of Li-ion leading to polarisation and low efficiency.

8. Low temperature increases viscosity and tension of the electrolyte thus worsening the diffusion and movement of Li-ions. This increased battery resistance impacts battery from operating properly. This could result in upto 80% capacity loss based on the temperature and discharge rating.

Mitigation methods to overcome the challenges of low temperature

9. Various mitigation techniques can be used to improve the performance of lithium ion batteries under low temperature conditions. A few techniques are being reviewed in this paper.

(a) **Use of unconventional electrolytes**. It was observed that most of the developed unconventional electrolytes improved drastically the performance of battery. The most common approach used by various researchers is to reformulate the electrolyte for low temperature applications. One of the approaches suggested in [5-8] is to replace LiPF6 electrolyte with LiBF4. Zhang et al [43] reported a new approach to improve the low-temperature performance of LIBs by replacing LiPF6 with LiBF4. They found that at -30℃, a battery with 1 m (mol kg/1 solvent) LiBF4 dissolved in 1 : 1 : 3 (wt) propylene carbonate (PC)/ethylene carbonate (EC)/ethylmethyl carbonate (EMC) mixed solvent delivered as high as 86% capacity compared to LiPF6, which delivered 72% (in comparison to that obtained at 20℃). Furthermore, the battery with LiBF4-based electrolyte showed lower polarization. The composition of electrolyte determines the temperature range of LIB, as electrolyte affects the transport of Li-ion through bulk electrolyte thus determining the quality of formed SEI. It was observed that all fluorinated electrolytes had high ionic conductivity and were electrochemically stable. Hence researchers in [29-34] developed all fluorinated electrolytes for low temperature LIBs, however due to high affinity between fluorinated solvents and Li ions, these electrolyte based Lithium cells failed to operate below -30ºC. Hence superelectrolyte was developed by dissolving all fluorinated electrolytes into highly fluorinated nonpolar solvents. Since the affinity between the solvents and Lithium ion is reduced, this electrolyte can work from temperature -90ºC to 70ºC by maintaining the electrochemical properties of all fluorinated electrolytes. However, these super electrolytes goes for the cells at very low area capacity of 1.2mAh/cm^2, which is very much lower that that (5mAh/cm^2) of commercialized LIBs at room temperature. Hence it will take some more time for this electrolyte to be used commercially in LIBs. A novel electrolyte was developed in [35] by dissolving lithium bis (trifluorome-thanesulfonyl) imide (LiTFSI) into ethyl trifluoroacetate (ETFA) solvent, which has weal ability to combine Li-ions and the electron, thus reducing the withdrawal of F atoms. This decreased binding energy of Li-ions-ETFA enables the rapid desolvation process in the SEI at low temperatures. Pure ETFA solvent has low freezing point of -78ºC and it can tolerate a faster rate of 1C at low temperature maintaining retention of almost 50%. Liquefied gas based electrolytes developed in [36-38] has columbic efficiency (CE) of 99.5% at -20ºC, had stable polarization and high capacity retention even at extremely low temperature. This liquefied electrolyte was further modified by adding a fully coordinating cosolvent, Tetra Hydro Furan (THF). This developed hybrid electrolyte showed excellent electrochemical performance at higher rate over wider temperature range and can produce uniform dense SEI lithium metal deposition and a stable lithium metal cycle without dendrites. Liquefied gas electrolytes cannot be used in large scale practical application as it suffers from potential safety hazards. Succinonitrile (SN), a type of plastic crystal electrolytes [39] is getting considerable attention in recent years to prepare low temperature electrolytes for LIBs due to their high ionic conductivity and large electrochemical window. Since SN based electrolytes suffer from poor mechanical strength, which limits its application in solid state LIBs. Therefore, subzero SN based polymer plastic crystal electrolytes were further developed which had excellent low temperature ionic conductivity and satisfactory mechanical strength. However it was observed that the serious reaction between SN and lithium metal deteriorates electrochemical performance of lithium metal batteries. Water based LIBs are being used as energy storage system even at below the freezing point of water due to their high safety, low cost, environmental benign and ultrafast kinetics process. Highly concentrated water in salt (WIS) [40], has wide electrochemical stability thus enhancing energy density of LIBs. It was also proved that aqueous electrolytes offered low charge transfer resistances, due to which LIBs can be charged and discharged at much higher rates at low temperatures. However aqueous electrolytes still suffer from common problem of high freezing point.

(b) **Use of different solvents/cosolvents**. It was observed that at different temperatures there was improvement in discharge capacity of LIB by increasing the content of certain chemicals like ethylene carbonate (EC) in solvent mixtures.The service temperature range of LIBs can be extended by adding solvents of low freezing points and high dielectric constants to formative electrolyte. Hence it is necessary to improve the basic solvent, which has low freezing point and optimize the formation of SEI so that LIBs can work satisfactorily at low temperatures.The performance of LIBs at low temperature can also be improved by addition of special molecules such as methyl formate (MF), methyl acetate (MA), ethyl propionate (EP) and ethyl butyrate (EB) along with carbonate electrolytes as described in [9-14]. It is important to judiciously select the co-solvents to improve the performance of LIBs at low temperature. It was observed that higher molecular weight esters showed better ionic conductivity as compared to lower molecular weight esters at -60^{0}C. LIBs delivered better performance by using cells having electrolytes blended with low viscosity additive as compared to that of all carbonate blended electrolyte [9-14]. At low temperature EA electrolyte has low freezing point (-84ºC) and ultra-high conductivity. In [9] it was shown that electrolytes with EA and vinyl carbonate (VC) used in LIBs at low temperature delivered superior rate capability at 3C and at -14ºC as compared to carbonate based electrolytes. It was also shown that use of such hybrid electrolyte improved interfacial compatibility of LIBs. Based on the results obtained in [14], researchers [6] developed an organic electrode based rechargeable LIB which used electrolyte consisting of EA. It was observed that this developed battery delivered almost 100% of the room temperature at -40ºC. As the temperature decreased further the discharge capacity of LIB decreased by 10% but still it performed well till -70ºC. The sluggish solvation/desolation of Li ion, intercalation compounds degraded the performance of LIB at -70ºC. Further it was shown that by adding inert Di chloromethane (DCM) to EA, resulted in electrolyte which possessed high ionic conductivity, low viscosity and wide electrochemical window from 0 to 4.85V at ultra-low temperature. However DCM has low boiling point and high volatility which limits its application in electrolytes at high temperatures. Methyl Pivalate (MP) based electrolyte having low melting point (-85ºC) and high dielectric constant was used as the primary solvent in LIB [15-23]. However it was observed that MP based electrolyte suffered from poor reductive stability and irreversible Li plating.

(c) **Use of additives**. Discharge capacity of LIB at low temperature can be further improved by adding special additives like EA along with EC. It was observed that a $LiCo_{1/3}Ni_{1/3}Mn_{1/3}O_2$ (NMC111) graphite cell at -40ºC with EA delivered 59.33% capacity retention to that at room temperature, thus proving that EA is an effective additive of low temperature electrolytes. To stabilize negative electrode and positive electrode interfaces along with building a SEI, fluroethylene carbonate (FEC) was added in MP based electrolyte. Graphite||Graphite dual ion batteries (DIB) showed better retentivity as compared to LIB, retaining 93.1% of its room temperature at -40ºC and 63.2% of its room temperature at -60ºC and at 0.1C. One of the component of liquid electrolyte for LIBs is Lithium bis(oxalate)borate (LiBOB) because it is fluorine free, environmental friendly salt and it possesses a superior thermal stability upto 300ºC and favours the formation of robust SEI on graphite anode and cathode electrolyte interphase (CEI) on high voltage cathodes. It can effectively passivate aluminium current collector [21]. The poor ionic conductivity of LiBOB/ carbonate solvents can be improved by using Gamma Butyrolactone (GBL) as it effectively dissolved LiBOB. LIBs using GBL based electrolytes can be operated in wide temperature ranges as it has low melting point (-43ºC), high flash point (98ºC) and high boiling point (204ºC). However LIBs using GBL/LiBOB has irreversible capacity which deteriorates its cycle performance. Therefor [8] introduced addition of Tetrafluropropyl ether (F-EPE) as an additive of GBL based electrolyte which has several diverting characteristics such as low surface tension, low melting point and high oxidation stability. [24-26] suggested use of ether electrolytes in LIBs because of their superior compatibility with lithium metal. Dioxolane (DOL) and dimethoxy ethane (DME) which are representative of ether are used as solvents for high energy Lithium Sulphur (Li-S) batteries as they have low freezing points of -95ºC and -58ºC respectively. However ether based electrolytes cannot be used in lithium cells as they cannot support high voltage. The LIB performance at low temperature can be significantly improved with Flurosulfonyl isocyanate (FI) additive as it prevents the crystallization of electrolyte at -20ºC.It was observed that at very low temperature Li ion cells using FI additive displayed higher capacities at different current rates as compared to carbonate based electrolytes. Thus it is proved that graphite/Li battery using 2wt% FI exhibits outstanding rate capability of 20mAh/g at 0.2C at -20ºC. The only drawback of FI is that it slightly decreases the initial columbic efficiency of graphite electrodes

(e) **Change in electrode chemistry**. Low ionic conductivity and sluggish diffusion in electrodes hinders the operation of LIBs at temperatures below -20ºC. At -60ºC, it was demonstrated in [28-30] that Fluorinated Ester Carbonate (FEC) Electrolytes altered solvation structure and formed thin SEI in DOL/DME based electrolytes. The morphological investigations carried out in [26-28] showed that the electrodeposited Li particles were larger in size as compared to that in pure ethers at temperatures below 0ºC, thus LiFePO4/Li metal batteries exhibiting high reversible capacity at -40ºC. One of the methods to improve the low temperature performance of LIBs is to apply surface coating on the cathode. This applied surface coating on cathode reduces the cell impedance. It also impedes side reactions between the electrode and electrolyte, thus improving the charge/discharge capacity and energy density of LIBs at low temperature. Several carbon based materials have been applied [41] to various cathodes which enhanced the Li ion transport property thereby improving the performance of LIB at low temperature.

(f) **Generic methods**.

(i) Use Battery Heaters. Battery heaters can be used to warm up the battery before use. This can help to reduce the internal resistance of the battery and increase its capacity. Basically the performance of LIB at low temperature decreases because at low temperature the impedance of LIB increases, discharging the battery terminal voltage which further reduces the capacity and power of battery. Thus the battery is heated to a certain temperature before charging the LIB. The various types of methods of heating applied to LIBs include internal core heating, internal resistive heating, convective heating, mutual pulse heating and external heating strategies such as those air, liquid or by using phase change materials. The charging time of the LIB used in vehicles can be reduced by turning on the motion mode which in turn will turn on the warm air, thus heating the battery and also increasing the power capacity of the battery. Zhang et al.17 developed a method to internally preheat LIBs at low temperature using a sinusoidal alternating current. They found that the battery subjected to an alternating current with an amplitude of 7 A and a frequency of 1 Hz could be heated from -20℃ to 5℃ within 15 min, and the temperature distribution remained essentially uniform.[42]

(ii) Charge Batteries Indoors. Charging batteries indoors can help to maintain a warmer temperature, which can improve their charging efficiency.

(iii) Use High-Quality Batteries. High-quality batteries are often better equipped to handle extreme temperatures. They may have a wider temperature range or be specifically designed for use in cold temperatures.

(iv) Proper Storage. Proper storage is critical to ensure that batteries are not exposed to extreme cold conditions for extended periods. Batteries should be stored in a dry and cool location, but not exposed to freezing temperatures.

(v) Battery Insulation. Battery insulation can be used to protect batteries from extreme cold. Insulation can be placed around the battery to keep it warm and maintain its performance. Insulating materials are also used in LIB to improve its performance at low temperature. Commercially available insulating materials made of rubber, plastic, cotton, having low thermal conductivity; low thermal diffusivity and being porous are used to maintain the temperature of LIB so as to enhance its performance at low temperature.

(vi) Battery Maintenance. Regular battery maintenance can help to prevent the effects of extreme cold on batteries. This includes checking the battery regularly for damage, cleaning the battery terminals, and replacing any damaged parts.

Other Factors Affecting The Low Temperature Performance Of LIB

10. The performance of LIB is directly affected by the size of diaphragm vent. If the diaphragm vent is too small it will increase the internal resistance of the battery. If the diaphragm vent is too large, the positive and negative electrode will make the contact easily or in other words it will be pierced by lithium dendrite causing short circuit of battery. Smaller the porosity of diaphragm, lesser will be the permeability of electrolyte, thus reducing the conductivity and vice versa

II. Conclusion

11. Low-temperature lithium batteries have received tremendous attention from both academia and industry recently. Electrolyte, an indispensably fundamental component, plays a critical role in achieving high ionic conductivity and fast kinetics of charge transfer of lithium batteries at low temperatures . In the context of the limited low-temperature performance of conventional electrolytes due to their intrinsic molecular structural properties, many attempts have been devoted to exploit unconventional electrolytes (fluorinated ester, EA, GBL, liquefied gas, ether, plastic crystal, and aqueous electrolytes) with low melting point and low viscosity to fulfil the harsh requirement of low-temperature lithium batteries up to now. Herein, in this Review, we have elaborated fundamental knowledge (solvation structure modification and SEI optimization) of unconventional electrolytes of lithium batteries from various new perspectives. Although the emerging unconventional electrolytes possessing some distinct functions alleviated unique problems presented in low-temperature lithium batteries, there are still grand challenges and plenty of rooms for the rational investigation in low-temperature unconventional carbonate electrolytes. In addition, perspectives on future development and research opportunities of low-temperature unconventional electrolytes are also included. Based on the MD' simulations, it has been found that the electrolytes have to exhibit unique solvation structures to accelerate the ionic conduction kinetics for low temperature batteries. Most decomposition products of lithium salts participate in the formation of SEI, including LiF, Li2O, Li2CO3, which is crispy during the process of charging and discharging. Combining with the reaction products of solvent, it forms stable SEI on the electrodes. The continuous decomposition of electrolytes results in the increased resistance of SEI. As a result, the electrolytes need to form stable SEI, but it also need to suppress the continuous decomposition. Overall, in spite of great achievements in unconventional electrolytes for low-temperature lithium batteries, many challenges and opportunities need to be explored. Finding good low-temperature electrolytes remains a significant challenge

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