

Review

Examining the Benefits of Using Boron Compounds in Lithium Batteries: A Comprehensive Review of Literature

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Abstract: Boron and boron compounds have been extensively studied together in the history and development of lithium batteries, which are crucial to decarbonization in the automotive industry and beyond. With a wide examination of battery components, but a boron-centric approach to raw materials, this review attempts to summarize past and recent studies on the following: which boron compounds are studied in a lithium battery, in which parts of lithium batteries are they studied, what improvements are offered for battery performance, and what improvement mechanisms can be explained. The uniqueness of boron and its extensive application beyond batteries contextualizes the interesting similarity with some studies on batteries. At the end, the article aims to predict prospective trends for future studies that may lead to a more extensive use of boron compounds on a commercial scale.

Keywords: boron; boron compounds; boric acid; lithium battery; lithium-ion batteries; cathode; anode; electrolyte; battery thermal management system (BTMS); battery performance



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1. Introduction to Boron and Lithium

During decarbonization, the trend of electrification continues to intensify and will drive the demand for batteries, especially for electric vehicles (EVs), where lithium plays a critical role. The challenge to improve battery performance has made boron, in various forms of compounds, a research topic in relation to lithium-ion batteries (LIBs) for decades.

Boron and lithium are similar elements in some ways. They are both considered light elements and less abundant in both present crustal concentrations and, indeed, in the universe. Their formation requires a further enrichment process that involves continental crust growth [1,2]. Furthermore, the extraction of lithium and boron together is common in existing mining operations and some prospective greenfield projects. Application wise, both elements could be used as flux in the making of vitreous materials, such as glass.

Why is boron used in batteries? Boron is a unique element in many respects. Firstly, boron is lightest element of the “metalloids”, which separates metals and non-metals in the periodic table. The fact that many boron compounds are electron deficient means that they have Lewis acidic characteristics. The four covalent bonds and the variety in the molecular structures of its compounds result in a wide range of chemical properties [3]. Consequently, its applications in urbanization are extremely diverse: glass and ceramics, oil and gases, polymer fire retardants, wood preservatives, fertilizers in agriculture, and many more. The benefit of using boron is often multifaceted. For instance, when sodium borate pentahydrate is used in fiberglass, it brings a unique combination of benefits to both the melting process and fiber properties; when disodium octaborate tetrahydrate (DOT) is used as an indoor wood preservative, it offers deep penetration into wood and residential benefits (such as low mammal toxicity).

Since energy storage is one of key enablers for decarbonation, the researching effort of LIBs is continuously fueled by a variety of needs for challenging improvement, such as safer ratings, longer life, more powerful output, higher capacity, lower cost, and less waste generation, etc. The abovementioned uniqueness of boron has therefore made boron

compounds the rational candidate of research, in every component of LIBs. With a boron-centric angle, this review could be the first review in lithium batteries that allows readers to conveniently understand extensive prior studies on boron compounds, to draw similarities and to facilitate research ideas.

2. Boron Used in Electrolytes

Battery mainly consists of four major components, which are cathodes, anodes, the electrolyte, and separators. Figure 1 shows the schematics of these components in conventional Li ion batteries (LIBs) and the movement of electrons, ions, and current flow under charging and discharging condition.

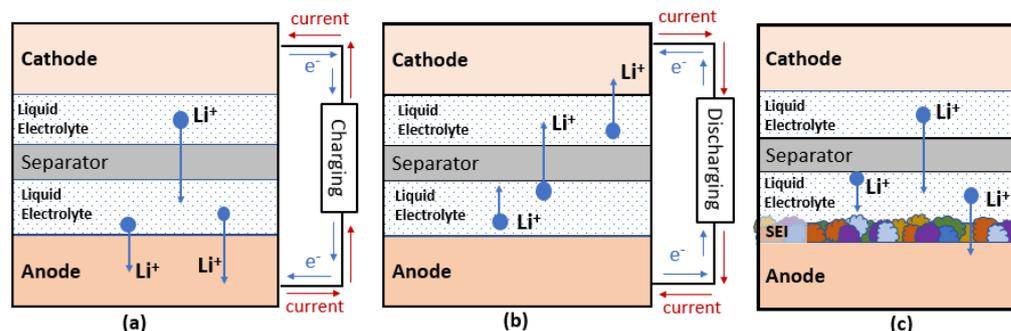


Figure 1. Schematic diagram of LIBs for (a) charging (b) discharging and (c) formation of SEI.

2.1. Electrolyte Additives—LiBOB

As early as the year 2000, lithium bis(oxalato)borate, also known as LiBOB, was first reported for synthesis and use in electrolytes of LIBs [4]. The presence of bis(oxalato)borate (BOB) anions helps the formation of a solid electrolyte interface (SEI) (Figure 1c), which is critical for the stability and long-term cyclability of graphitic anodes [5]. Because of the merits of being halide-free and more thermally stable than LiPF_6 , LiBOB has been proven beneficial either as a direct replacement for LiPF_6 or as an electrolyte additive (2 wt%) in a nickel cobalt aluminum (NCA)—graphite system [4,6]. Moreover, LiBOB was studied as an enabler for using propylene carbonate (PC) as a solvent [6]. Although it was beneficial to the NCA cathode system, research revealed that Co could play a role in catalyzing the decompositions of BOB anions on cathode surfaces, which explained the inferior performance of LiBOB in lithium cobalt oxide (LCO) cathodes and nickel manganese cobalt (NMC) cathodes. Another study found that one disadvantage of using LiBOB was that SEI could become too resistive to perform well at sub-zero temperatures [7].

2.2. Electrolyte Additives—TPFPB and TB

Regarding the interface on the cathode side, tris(pentafluorophenyl)borane (TPFPB) can be added into the electrolyte as a film stabilizer on the cathode, which improves the power capability in lithium manganese nickel (LMN), NMC333, and LiFePO_4 (LFP) cathodes [8–10]. Similarly, adding <0.1 M of trimethyl borate (TB) or 0.1 M TPFPB to LiPF_6 was found to be effective at suppressing the thermal breakdown of electrolytes and promoting the establishment of a protective film on LFP, hence improving the cyclic stability at 55–60 °C [11,12]. TB with 10 wt% was also found to be beneficial to the cyclic stability and coulombic efficiency in NMC333 [13].

2.3. Electrolyte Additives— LiBF_4

Around the time of the first report of LiBOB, LiBF_4 was found to have lower conductivity but better low-temperature performance than LiPF_6 [14]. Later in LIB development, LiBF_4 salt in carbonate electrolytes was tested as a direct replacement for LiPF_6 , which improved capacity retention in NMC622 and NMC442 systems, due to a more stable B-F bond compared to the P-F bond [15–17]. When only used as an additive to LiPF_6 , 1 to 2 wt% of LiBF_4 significantly enhanced capacity retention in the NMC532 system, suggesting

the likely mechanism that LiBF_4 contributed to the formation of SEI on both electrodes [18]. It is worth noting that one study suggested that boron trifluoride (BF_3), which is the decomposed product from LiBF_4 , was a typical Lewis acid and could dissolve the decomposition product LiF from LiPF_6 [19]. Harnessing the BF_3 functionality mechanism, pyridine-boron trifluoride (PBF) was tested as an additive. A 3 wt% dosage was able to enhance capacity retention and preserve low impedance in NMC/graphite systems, such as NMC111 and NMC442 [19]. Similar effectiveness was also found in a pouch cell experiment on NMC532 and NMC622 [20]. A combination of using PBF as additives and LiBF_4 as electrolytes for NMC442 provided incremental benefits compared to using LiBF_4 as electrolytes [15]. Figure 2 shows the molecular structure of boron compounds mentioned in this review.

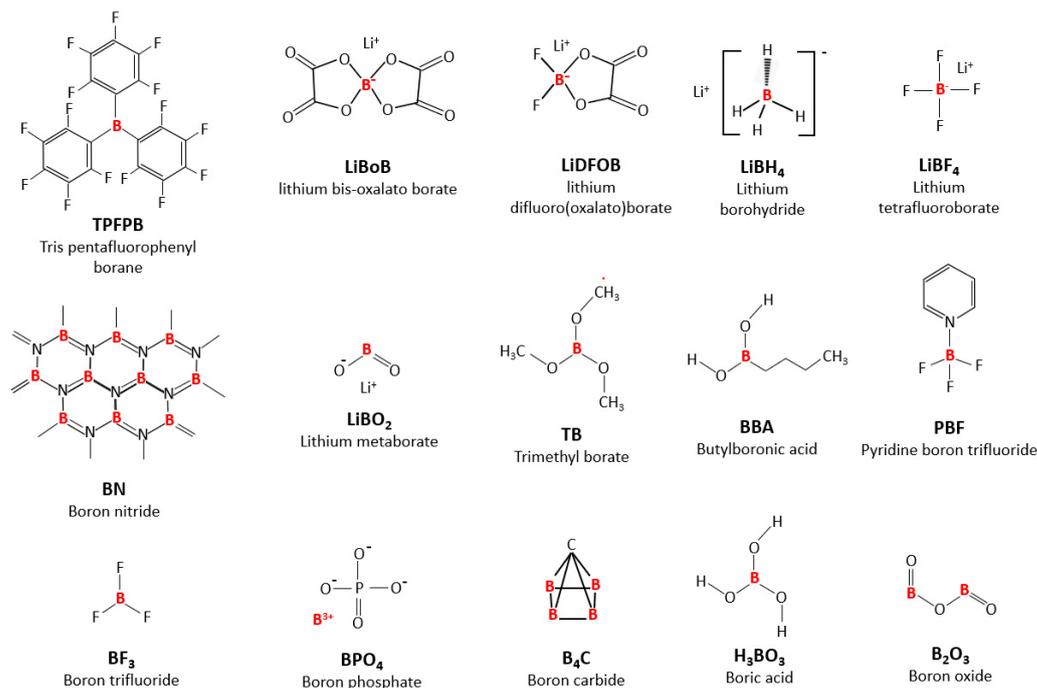


Figure 2. Molecular structure of boron compounds mentioned in the review.

2.4. Solid-State Electrolyte—Boron Nitride & Boric Acid

To advance the safety aspect of LIBs, solid electrolytes were also extensively studied for their mechanical stability and characteristic of lower flammability. Several combinations for solid electrolytes exist, but they usually contain inorganic and polymer materials. The typical challenge of using solid electrolytes, however, relates to the relatively inferior ionic conductivity and high interface resistance with electrodes [21]. Boron nitride has been commonly studied because of its physical, thermal, and electrical properties [22].

Studies revealed that boron nitride (BN) could be made into a sheet-form hosting structure for ionic liquids (ILs) containing solid electrolytes; as a result, low-temperature conductivity was increased [21]. Other studies showed that adding 1% BN as additives could improve the cycling performance in polymer/salt hybrid electrolytes [23]. Hexagonal boron nitride (h-BN) was also studied to improve cycling performance at a temperature of up to 175 °C [24]. It is also worth noting that, even in non-solid-state electrolytes, adding h-BN was proven to be effective at improving thermal stability and cycling performance [25]. It is also interesting to note that light elements similar to boron, such as fluorine, were also commonly studied as doping elements in solid state batteries to improve stability [26,27].

Li metal batteries (LMBs), that utilize lithium as anodes, have the advantage of a higher energy density. Their use has been commonly studied together with solid-state electrolytes. The challenges of using Li metal concern Li reactivity and the formation of Li dendrites [28]. A study showed that BN could be used as a coating on solid-state electrolytes of LMBs to stabilize the electrolyte–anode interface and hence improve cycling

performance in LFP systems [29]. Other studies showed that cycling performance could be improved by adding 1% BN to ceramic electrolytes [30]; adding hexagonal boron nitride (h-BN) to polymer composite electrolytes of LFP [31]; adding amine-functionalized boron nitride nanosheets to gel polymer electrolytes of LFP and LCO [28]; or adding hexagonal boron nitride nanosheets (h-BNNS) to a composite polymer on LFP [32]. An alternative route to electrolyte additives was found to use a boric acid treatment on a Li anode to form a lithium borate layer that suppressed dendrite growth and enhanced cycling performance in LMO/Li systems [33]. Using boric acids as additives was also found to successfully suppress dendrite growth, stabilizing interfacial film on the Li-metal anode in various LMBs systems [34,35].

3. Boron Used in Anodes

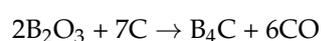
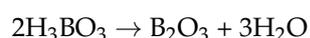
3.1. Graphite Anode—SEI

Notwithstanding extensive and intensive research and development (R&D) efforts for alternative materials, graphite, a key component of LIBs, has been successfully used for over three decades on a commercial scale in the LIB industry. Like typical cathodes in LIBs, graphite functions by acting as an intercalation compound, which means inserting Li^+ into the crystal structure to store energy [36].

Due to the active nature of graphite in LIBs and the issue around exfoliation because of solvent interaction and repeated volume change during the discharge/charge cycle, SEI remains critical to sustain the performance of graphite anodes. The boron surface coating on graphite was found to benefit the formation and stability of SEI by contributing to formation chemistry or morphology changes [37–39]. The essential mechanism was identical to some boron-based electrolyte additives. When the surface of graphite was treated with borate acid, results showed an improved rate capability, life cycle, and capacity [40–43]. U.S. Borax Inc. led similar in-house research projects and found a similar outcome to these findings in the literature.

3.2. Graphite Anode—Graphitization

As well as the graphite coating, borate addition during the high-temperature treatment process of synthetic graphite (SG) could lower the required temperature for graphitization [44]. When placing boron in a lattice structure, it was found to act as an electron acceptor; hence, it increased the specific capacity to 437 mAh/g, which is superior to the theoretical maximum of pure graphite at 372 mAh/g [45]. Regarding improved graphitization, the degree of graphitization was increased with boron substitution, which was found to exist in the form of B_4C [46]. As B_2O_3 can be decomposed from boric acid at the pitch synthesis temperature, adding either B_2O_3 or boric acid will form B_4C during the graphitization process through the reaction of carbon and B_2O_3 [44]. A study in 2001 used boric acid, boron oxide, and B_4C as the graphitization catalyst, suggesting that 1–2 wt% boron atoms enhanced anode performance [47]. A recent study (2021) also found that the preparation of pitch and boric acid (max. 5 wt%) to make SG resulted in enhanced graphitization and a higher C-rate, the speed where the battery becomes completely charged or discharged. It was further reported that excessive boric acid dosage (7 wt%), however, was detrimental to graphitization and capacity [44]. Hence optimization was required for the use of boric acid.



3.3. Graphite Anode—Upcycling

While the recycling of LIBs is becoming increasingly topical, the industry does not practice graphite recycling, presumably due to a relatively lower value of recycling anodes compared to recycling cathodes.

A recent study (2020) by the University of California, San Diego, revealed that using a boric acid solution was considered a green and upcycling approach to rejuvenate spent graphite anodes, providing significant advantages over the use of HCl and H₂SO₄. Firstly, the process of boric acid treatment and an annealing step afterward was considered relatively simple and low-cost compared to high-temperature sintering. Secondly, using boric acid alleviated concerns over secondary pollution caused by using volatile and corrosive acids. Thirdly, using boric acid completely recovered Li⁺ trapped not only in SEI, but also in graphite bulk. Finally, the boric acid treatment modified the graphite surface with functional B-doping afterward. The outcome was that the rejuvenated graphite anodes were found to have improved electrochemical activity and excellent cycling stability [48]. The schematic in Figure 3, which is reproduced from Gaines [49], shows the routes where boric acid could be introduced in the typical lifecycles of EVs.

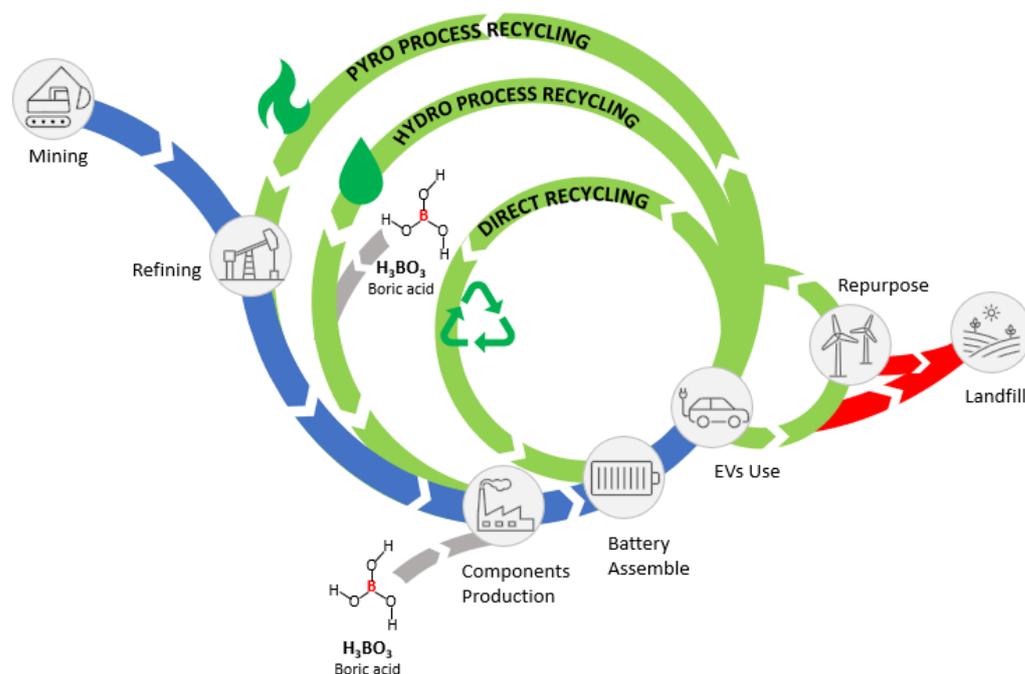


Figure 3. Introduction routes of boric acid in lithium battery lifecycles [49].

3.4. Graphene Anode—Li Adsorption

Graphene is a two-dimensional monolayer structure that was first studied and obtained in 2004 by mechanical exfoliation from graphite materials [50]. Because of its specific area and physical strength, graphene was investigated as an alternative anode material to enhance the capacity retention and cycling performance of graphite [51,52]. Graphene anodes could help to overcome the challenge of LIBs, that is, having the advantage of a high-power density (energy discharge rate), but the relative disadvantage of low energy density (amount of energy stored) [53]. A lithium iron phosphate (LFP) system could be a classic example of that.

Boron doping in graphene could provide higher capacity because introducing lattice defects allows for more active sites for Li⁺ intercalation [54]. It could increase capacity and suppress the decomposition of electrode materials to some degree [55]. It was later discovered that a green, large-scale synthesis was successful by using boric acid as a boron precursor to make boron-doped graphene. This method was reported to achieve a doping level of 5.93% and improve the discharge capacity by 66% from pristine graphene [56].

3.5. Other Anodes—Miscellaneous

While there is also extensive R&D for other anode materials, boron has commonly been studied jointly. For example, boric acid was studied as a catalyst to synthesize hard carbon microfiber for LIB anodes from cotton [57].

Another example is a silicon anode, which was revealed to have a significantly larger specific capacity [58], with a theoretical value of approximately 3580 mAh/g. To optimize the binder system for silicon-based anodes, boric acid was studied as an effective chemical to crosslink polyvinyl alcohol (PVA) and chemically connect binders with the Si surface. Furthermore, elemental boron was also found to promote the formation of SEI, which resulted in improving initial coulombic efficiency (ICE) [59]. This application draws a comparison with the current commercial use of boric acid to crosslink PVA polarizer film in the liquid crystal display industry. Boric acid was also evaluated in carbon-coated porous silica macroparticles (SiO₂@C). The formation of a borosilicate (Si-O-B) phase and B-O bonding helped to improve charge capacity and stability [60]. However, a big volume change throughout the intercalation is one of the main challenges for silicon-based anodes in LIBs [61].

TiO₂ have been studied as anodes in LIBs due to their outstanding merits of low volume change during the Li⁺ intercalation process, a faster diffusion rate, and more lithium hosting channels [62]. By applying boron doping (up to 10.4 wt%) to the TiO₂ anodes, the results demonstrated that TiO₂ lattices were enlarged, and the specific surface area was increased. The application was through a sol-gel method with the use of boric acid (H₃BO₃). The further increased Li⁺ diffusion rate therefore improved the cycle capacity of TiO₂ [63].

Spinel Li₄Ti₅O₁₂ (LTO) was also widely studied as an anode due to its durability and safety [64]. To overcome the main challenges for its wide applications in EVs, extensive studies were carried out to enhance the intrinsic electronic conductivity and specific capacity [64–66]. To improve conductivity, using boric acid to form a B-doped carbon coating was found to be effective [67]. Capacity performance could also be improved by using a B-doped carbon layer on LTO [68]; h-BN composite with LTO [66]; or B-doped carbon nano tube composite with LTO [69]. Another study found that using 2 wt% of Li₂O-2B₂O₃ (LBO) to form an interface between LTO anode and electrolyte could improve the rate capability without changing the specific capacity [70].

4. Boron Used in Cathodes

The appearance and dominance of LIBs has originated from the nature of high energy density. The design and development of cathode materials are critical and remain a key focus of LIBs because cathode materials not only dictate energy density, but also represent the bulk of battery costs [71,72]. Boron, like its applications in other parts of LIBs, has been studied throughout the development history of cathode materials, mainly through either doping or coating (Figure 4).

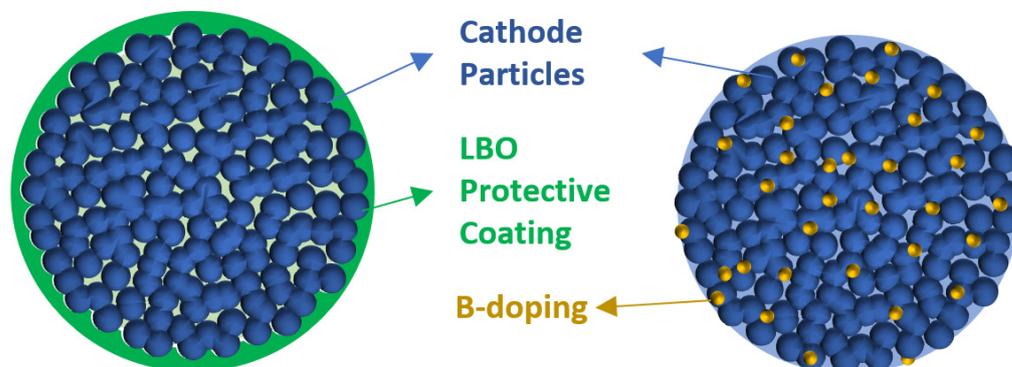


Figure 4. Schematic of two improvement approaches of using boron compounds in cathode.

4.1. LCO, LNO, and LMO

Since LIBs were commercialized in 1991 [73], lithium cobalt oxide, or LiCoO₂ (LCO), has been used extensively in portable electronics due to high volumetric energy densities. However, the irreversible and significant structural changes during the charge/discharge

cycle resulted in rapid capacity fading [74]. To improve cycling performance, lithium metaborate LiBO_2 was studied to modify cathode composition to form $\text{LiCo}_{0.85}\text{B}_{0.15}\text{O}_2$ (2011) [75]. Another cycling performance improvement strategy involved modifying the cathode surface. For example, LiBO_2 was used to form a layer over LCO in all-solid-state LIBs, enhancing cycling performance [76]; the use of boric acid to form a glassy B_2O_3 coating on the interface with LCO helped to suppress electrolyte decomposition [77]. A recent study (2021) demonstrated the value of using a lithium borohydride (LiBH_4)-modified LCO surface to increase cycling stability [78].

Later in the development, lithium nickel oxide (LNO) was studied as an alternative to LCO because (1) they share comparable specific capacities of 274 and 275 mAh/g, respectively; (2) nickel was more abundant than cobalt, hence more cost effective; and (3) LNO had a similar lattice structure, allowing Li^+ intercalation [72]. A study showed that using 2 wt% lithium difluoro(oxalato)borate (LIDFOB) was effective at improving cycling performance. Although the application was through the route of an electrolyte additive rather than a direction application on the cathode, the mechanism was essentially about forming a boron-rich layer on the cathode [79].

Further to LNO, lithium manganese oxide (LMO) was considered as an alternative material, given its merits of low cost, greater natural abundance, and low toxicity. However, LMO also faced similar issues of capacity fading [80]. Single-crystal boron-doped $\text{LiMn}_{1.96}\text{B}_{0.04}\text{O}_4$ could be synthesized by a facile approach with no changes to the spinel structure. The result exhibited a high initial discharge and improved cycling performance [81]. A glassy coating of lithium boron oxide (LBO) $\text{Li}_2\text{O}-2\text{B}_2\text{O}_3$, which was a commonly studied boron compound, was another strategy to improve LMO cyclability. Compared to uncoated LMO, a few studies examined how beneficial impacts varied by different application methods of LBO and by various particle sizes of LMO, such as powder or nanoparticles [82–86]. The reasons for extensive studies of glassy LBO as a surface treatment were as follows: (1) good conductivity; (2) good wettability; and (3) low viscosity for processing ease [87,88]. It is interesting to note that other boron-in-glass applications, such as enamel glaze coating and insulation fiberglass, share similar benefit from the use of boron compounds.

4.2. LFP

LiFePO_4 , or Lithium iron phosphate (LFP), is one of the most widely produced mainstream battery systems in EVs and even utility-scale stationary applications. LFP is widely used and produced in China [89]. LFP cathodes do not contain nickel or cobalt, both of which are considered in short supply and have high price volatility. Therefore, cost is the first key advantage of LFP. The second advantage is safety because LFP is non-toxic and more thermally stable than other LiB cathode systems [90], and hence less susceptible to fire or explosion. The disadvantage of LFP is that it has lower energy density and lower operating voltage because of the inferior electronic conductivity and diffusion rate of Li^+ through the $\text{LiFePO}_4/\text{FePO}_4$ interface [91,92].

To overcome this challenge, the direct substitution of $(\text{PO}_4)^{3-}$ in LiFePO_4 in the form of $(\text{BO}_3)^{3-}$ at 6 mol% could advance capacity retention of LFP cathodes [93]. Another similar B-doping experiment used 3–9 wt% of boron phosphate BPO_4 as a precursor and reported that the rate capability was enhanced due to the improved diffusivity of Li^+ [94]. In addition to direct doping, a B-doped carbon coating was adopted to improve rate performance. The doping on the carbon layer could be achieved either by boron alone [95] or jointly with other elements, such as N or P [96,97]. The rate performance was enhanced due to improved conductivity.

4.3. NMC

The development history of LCO, LNO, and subsequent LMO led to studying a combination of the three, commonly named nickel manganese cobalt (NMC). Apart from LFP, NMC is currently the most used category of battery cathode for EVs. By forming

a tertiary system, the individual benefits of LCO, LNO, and LMO (capacity, rate performance, and safety, respectively) can be combined. Accordingly, the overall performance can be tailored by relative composition. Figure 5 shows in a ternary plot the location of relative NMC compositions and the part of the battery in which boron's study have been mentioned in this paper. As battery capacity remains the focus of development for EVs due to so-called range anxiety, the clusters of commercially used systems are currently situated toward the vertex of LiNiO_2 , with NMC333 having the lowest LNO ratio. The major challenge of high nickel was that it was believed to have low thermal stability due to the high reactivity of Ni^{4+} [72].

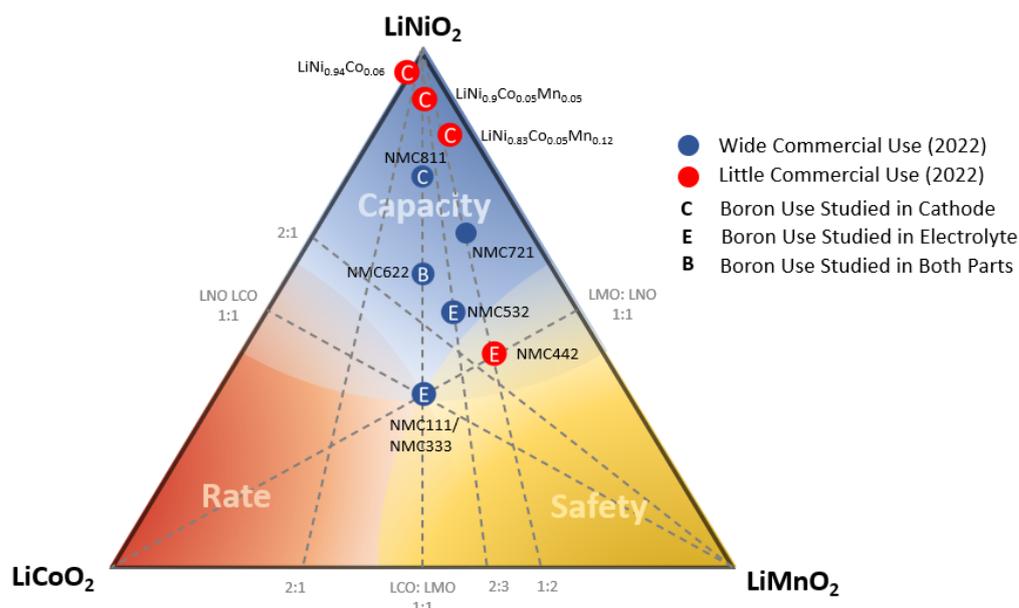


Figure 5. Ternary plot of NMC systems.

NMC811 is currently (2022) the highest commercial NMC cathode in terms of LNO ratio. A 2020 study showed that using boric acid through a wet-chemical coating treatment, followed by a sintering step, was able to improve the lifetime and high-temperature performance of NMC811. The formed glassy borate lithium film successfully prevented microcracks [98]. Apart from wet-chemical treatment, solid boric acid could also be mixed dry with an NMC811 cathode, followed by a 300 °C heat treatment to form a lithium borate coating on the NMC cathode. This coating improved cycling performance of the NMC811/sulfide-based solid electrolyte system [99].

Regarding B-doping, a 2021 study utilized boric acid as a co-sintering agent to achieve boron doping (3 at%). This not only improved rate performance and cyclability but also improved structural stability [100]. The mechanism behind the B-doping benefit is not fully understood, but one study suggested that B-doping affected crystal structure and cation disorder [101]. In NMC622, B-doping was found to change the microstructure of primary particles significantly and doping alone provided a marginal benefit to structural and electrochemical stability. When 0.5% B was co-doped with 1% Mg, there was a synergistic effect that improved the capacity retention noticeably due to a reduction in impedance growth and cation mixing [102].

Other high-nickel NMC, such as $\text{LiNi}_{0.90}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$ and $\text{LiNi}_{0.83}\text{Co}_{0.05}\text{Mn}_{0.12}\text{O}_2$, despite having little commercial use currently (2022), could increase their capacity retention and cycling performance with 1 mol% and 0.6 mol% B-doping, respectively. The mechanism was believed to be related to a stronger B-O bond and the release of the internal lattice strain [103,104]. The research of $\text{LiNi}_{0.83}\text{Co}_{0.05}\text{Mn}_{0.12}\text{O}_2$ was also demonstrated on pouch cells [104]. A similar beneficial impact and mechanism were also found in high-nickel systems such as $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Ti}_{0.05}\text{O}_2$ and $\text{LiNi}_{0.94}\text{Co}_{0.06}\text{O}_2$ [105,106]. Another less common

approach was adding a bi-boron additive to electrolytes, which could stabilize the interface of $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$, and hence the cycling life [107].

Li-rich layer oxides (LLOs) could be considered a variant of NMC since NMC can turn into LLO, with Li ions partially replacing Ni, Co, and Mn in NMC. LLO has the advantage of high capacity but the disadvantage of capacity fading [108]. One example of LLO is $\text{Li}_{1.2}\text{Mn}_{0.6}\text{Ni}_{0.2}\text{O}_2$. A study in 2015 showed that B-doping improved rate capability and cycling performance [109]. Another example of LLO is $\text{Li}_{1.2}\text{Ni}_{0.13}\text{Co}_{0.13}\text{Mn}_{0.54}\text{O}_2$ [110]. Studies showed that B-doping improved cycling performance in both half cells and pouch cell experiments. The abovementioned advantageous mechanism of B-doping was very similar to that in NMC systems [110,111].

4.4. NCA and NCMA

The focus on capacity increases the LNO content, but at the expense of safety. This is because, as the LNO ratio increases, the structural and thermal stability decrease, thus also decreasing cycling ability [112]. The role of aluminum substitution for nickel is to provide thermal stability and safety owing to the inertness of Al [113]. Aluminum can be found in lithium nickel cobalt aluminum (NCA) and NCMA (nickel, cobalt, manganese, aluminum), both of which have wide commercial use in cathode systems of EVs (2022).

In NCA of $\text{LiNi}_{0.885}\text{Co}_{0.1}\text{Al}_{0.015}\text{O}_2$, using B-doping enabled the modification of the microstructure and helped alleviate internal strains, which then enhanced cycling performance [114]. A gradient B-doping method, which allowed the concentration to change with depth, was applied to $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$. Even at high temperature, the research demonstrated that structural stability and cycling performance were improved because doping could encumber impedance increase during cycling [115]. In relation to coating on NCA, an ultrathin LBO layer was constructed on the interface that enhanced structural stability and the cycling performance of $\text{LiNi}_{0.87}\text{Co}_{0.10}\text{Al}_{0.03}\text{O}_2$ [116].

In the NMCA system of $\text{LiNi}_{0.88}\text{Co}_{0.06}\text{Mn}_{0.03}\text{Al}_{0.03}\text{O}_2$, an LBO coating was applied for chemical stability and structural stability during the charge/discharge cycle, which reduces capacity loss [117].

5. Boron Used to Assist Manufacturing

Beyond batteries, boron compounds are often used in manufacturing and chemical processes. For instance, borates have been used as glass melting flux since the 17th century. Sodium borates are also widely used in various metal cleaning processes. The cleaning application is attributed to the known capability of degreasing and the pH stability at various concentrations and temperatures. Again, it is interesting to observe similarities in how boron could help with the manufacturing process of LIBs.

In relation to the abovementioned NCMA cathode, one study showed that boron could assist fluxing agents and make uniform single crystals of $\text{LiNi}_{0.92}\text{Co}_{0.06}\text{Mn}_{0.01}\text{Al}_{0.01}\text{O}_2$, which exhibited an outstanding rate capacity and thermal stability [118].

Regarding cleaning, there has been increasing focus recently (2020–2022) on studying boric acid as a washing agent for residual lithium compounds (RLCs). Nickel-rich cathodes are prone to reacting with H_2O and CO_2 in the air, forming RLCs, such as LiOH and Li_2CO_3 . RLCs are highly undesirable because they (1) deteriorate cathode performance as well as (2) creating manufacturing challenges, and (3) potential safety hazards [119].

The efficacy has been demonstrated on Ni-rich cathode systems, such as the NCM811 [120] and NCMA system of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.09}\text{Al}_{0.01}\text{O}_2$ [121] and $\text{LiNi}_{0.90}\text{Co}_{0.06}\text{Mn}_{0.04}\text{O}_2$ [122]. In an NCM811 study, boric acid was used not only to clean surfaces, but also to transform RLCs into uniform boron-containing lithium coatings, resulting in a high discharge capacity and better cycling performance compared to pristine NCM811 and ethanol-washed samples [120]. This dual benefit was also tested in a study of NCMA, simultaneously improving structural stability and electrochemical performance [121]. When using a system of $\text{LiNi}_{0.90}\text{Co}_{0.06}\text{Mn}_{0.04}\text{O}_2$, optimal parameters were established at a boric acid concentration of 1 g/L; wash time of 5 min; and solid–liquid ratio of 1 g/mL. The benefits found

in the study included effective washing and suppressing the formation of RLCs during storage [122].

6. Boron Used in Separators

In internal combustion engine (ICE) vehicles, lead-acid start-stop batteries commonly use borosilicate glass microfiber as an absorbent glass mat (AGM) that not only functions as separators, but also as electrolyte absorbents. For LIBs in electric vehicles (EVs), a battery separator is a critical and complex component. On the one hand, it functions like a physical barrier, preventing short circuiting between the cathode and anode. On the other hand, it also acts as a channel to allow for the passage of ions during charge/discharge cycles. Therefore, proper functioning demands a complex suite of parameters. Table 1 lists typical required parameters for separators used in LIBs [123,124]. The current conventional commercial mainstream materials are polyolefin-based, such as polypropylene (PP) and polyethylene (PE), although other fibers, such as polyester, cotton, and glass, are also available in the nonwoven types of separators.

Table 1. Parameters and requirements for separators [123,124].

Parameters	Requirement
Chemical and Electrochemical Stability	Yes. Long Duration
Wettability	Quick and Complete Wetting
Mechanical Property	>100 kg/cm
Thickness	20–25 μm
Permeability	<0.025 s/ μm
Porosity	40–60%
Pore Size	<1 μm
Dimensional Stability	Yes
Thermal Stability	<5% Shrinkage

The typical challenge of using polymer-based separators was believed to be limited thermal stability, which could potentially result in thermal runaway (an uncontrolled and exothermal chain reaction) and subsequent catastrophic failure [125]. Extensive studies were carried out on the synergetic benefit of boron nitride (BN). BN has a unique combination of high thermal conductivity and high electrical insulation and, as mentioned in Section 2.3, has been commonly studied as an additive for solid-state electrolytes.

A 2019 paper studied the use of boron nitride nanotubes (BNNTs). When synthetic long and fine BNNTs were incorporated into polyolefin separators, thermal stability was improved by up to 150 °C. The incorporation assisted heat absorption and distribution during the cycling process, hence improving battery safety [126].

There are also studies on the applications of hexagonal boron nitride (h-BN), which was mentioned in Section 2.3, being studied as an electrolyte additive. A composite material made of carbon-coated hexagonal boron nitride (h-BN) and polyvinylidene fluoride (PDVF) exhibited a superior electrochemical performance compared to polyolefin, allowing for the safe operation of LIBs up to 120 °C [125]. Another study showed that coating conventional PP with a layer made by h-BN and polyimide helped effectively suppress Li dendrite growth and consequentially improved NCM811 performance compared to PP separators without a coating [127].

For separators used in Li metal batteries (LMBs), there was a study on using a layer of boron nitride graphene coating on one side of a PP membrane to enhance the stability of the LFP/Li system [128].

7. Boron Used in BTMS

When LIBs or other high-energy-intensity batteries operate, there are different types of heat generated in various parts of the battery pack, such as reaction heat, entropy-change heat, and electrical resistance heat. The battery temperature can then affect both

kinetic and thermodynamics aspects of the battery, including the power rate and capacity fading [129]. A battery thermal management system (BTMS) is therefore essential to provide an optimal working temperature range for batteries. If the BTMS fails to provide timely heat distribution and dissipation, localized acute heat generation could lead to thermal runaway [130].

Among conventional BTMSs, such as water-cooled or phase-change material cooling, thermal interface materials (TIMs) play a critical role in establishing seamless physical contact on a microlevel between a heat source and dissipation media because of surface roughness (Figure 6). Boron nitride, which has a unique combination of high heat conductivity and high electrical insulation, is often used in experiments as a filler in TIMs. One experiment showed that using surface-modified spherical boron nitrides (SBNs) reduced maximum temperature [131]. The silicone rubber matrix was also studied to incorporate hexagonal boron nitride (h-BN) as a filler to increase thermal conductivity. One paper concluded that 30 wt% of h-BN made the thermal conductivity over three times greater than that of original silicone rubber [132]. Another paper showed that the thermal conductivity would be four times higher if using hybrid fillers of h-BN carbon fillers [133].

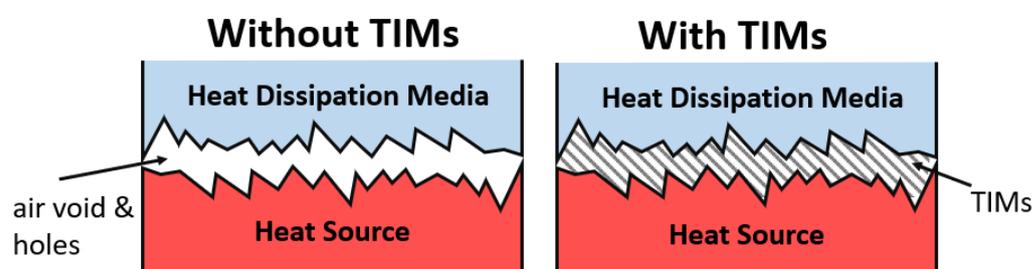


Figure 6. Schematics of thermal interface materials.

Apart from conventional systems, there are also studies on using non-conventional systems along with BN. For example, using a silicone sealant as an immersion medium with 10 wt% BN loading reduced the maximum temperature compared to an air-cool module or a silicone sealant module [134]. There was also a study on solid cooling media by using h-BN together with paraffin wax, which has a high heat capacity and good electrical insulation. The results show that, with 20 wt% loading, an orderly interconnected h-BN frame inside a paraffin wax exhibited eight times more thermal conductivity than pristine paraffin wax, and four times more than random dispersed h-BN [135]. Finally, BN was also studied for use as a coating on the external casing of LIBs. With the Taguchi method, the parameters of coating thickness and casing surface roughness were optimized, with the use of BN to improve thermal performance [136].

8. Boron Used in Lithium Sulfur Batteries

This paper has so far predominately focused on a category of LIBs that have been proven successful in commercialization. Lithium sulfur batteries (LSBs), although not yet commercially available, have received great research attention because of the merits: low material costs (i.e., free of cobalt and nickel) and higher energy density. In terms of mechanism, LSBs differ remarkably from LIBs in the following areas. Firstly, LSBs utilize lithium metal as an anode, which is similar to lithium metal batteries (LMBs). Secondly, the charge-holding mechanism of LSBs is based on Li^+ reactions on the electrodes rather than Li^+ intercalation [137,138]. As shown in Figure 7, these reactions during the charge/discharge cycle are part of a complex multi-step process with lithium polysulfides that range from solid to liquid [139]. Thirdly, with some soluble polysulfides, LSBs pose a major challenge that LIBs do not; that is, the “shuttle effect”, a mechanism related to the diffusion of soluble polysulfide through separators and the subsequent undesired reactions with the lithium anode surface. The shuttle effect directly results in self-discharging [138].

The combined effects with anode corrosion and dendrite growth lead to impaired cycling stability and capacity [140–142].

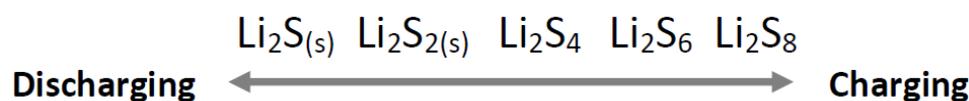


Figure 7. Lithium polysulfides in LSBs during charge/discharge cycle.

To alleviate the shuttle effect, one research direction of interest was to make separators hinder the passage of polysulfides. Hexagonal-boron nitride (h-BN) and BNNT separators were reported to lessen the shuttle effect preventing polysulfide passage [142–144]. These reports further suggested that the mechanism was due to the presence of boron atoms and the availability of a vacant p-orbital, which enabled the Lewis-acid characteristics of boron nitride [142,143]. A similar research direction was pursued by studying a boron-doped functional layer on separators to improve cycling stability, such as using B-doped graphene oxide [145,146] or B-doped anatase TiO₂ [147].

Another research direction of interest to alleviate the shuttle effect was to better trap polyamide on cathodes. Using the nanostructure of B₄C inside the cathode of LSBs was reported to increase cycling performance [148–150]. The improvement was due to the following: (1) B₄C improved the kinetics of polysulfide conversion; and (2) nano B₄C structures provided entrapment for polysulfides [148].

9. Summary and Prospect

This paper has extensively reviewed the literature studying boron compounds in various parts of lithium batteries. With all of the mentioned benefits and studied boron compounds, Figure 8 attempts to generalize the similarity in this review. The dotted line means that some of these benefits are common even when the application is through the adjacent components.

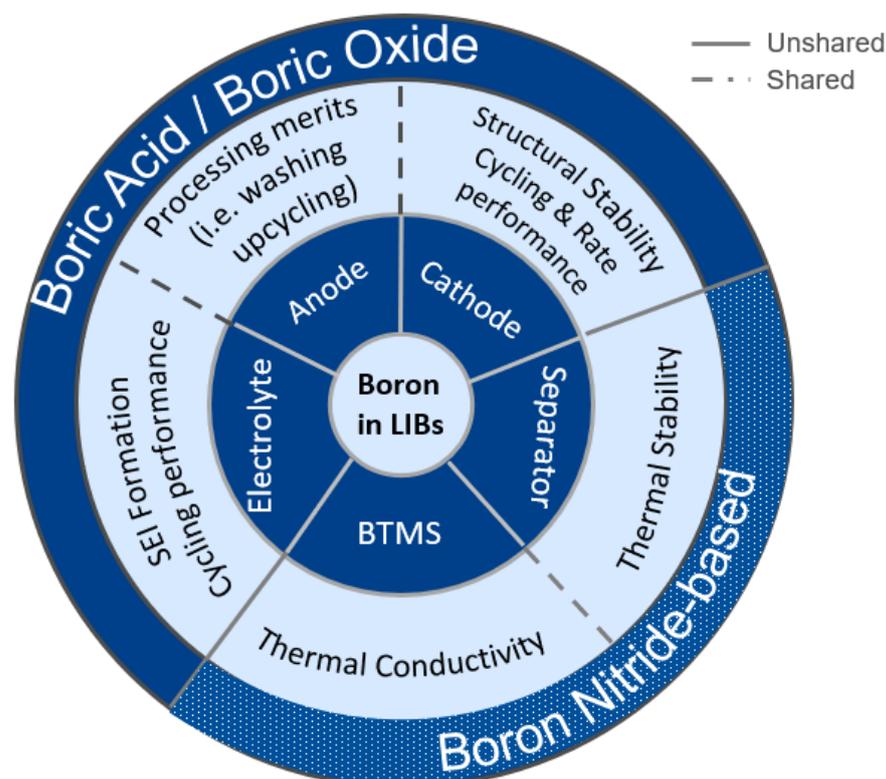


Figure 8. Generalized benefit and studied boron compounds in five LIBs components.

In closing, an additional summary and reflections are offered to assist discussion for future study that might lead to commercialization.

9.1. Summary

- Various boron lithium compounds were studied and proven effective in assisting the formation and stability of SEI. These can be introduced through electrolyte additives or through direct application to the anode.
- For the direct application of boron compounds to the anode, using boric acid was reported to provide diverse benefits.
- In the cathode, boron doping and forming an LBO layer were of particular interest. Cathode doping and cathode coating are the two conventional approaches to improving the cycling performance.
- The use of boron compounds has been widely employed in experiments on those cathode systems that currently have wide commercial applications in EVs, such as NCMA, NMC, NCA, and LFP.
- Using boron compounds to assist manufacturing could be a way of achieving the “one stone, two birds” outcome. Using boron compounds to achieve multifaced benefits is common beyond batteries.
- Particularly, using boric acid was considered an effective approach in anode upcycling and cathode washing for residual lithium compounds (RLCs).
- For applications in separators and BTMS, boron nitride and its derivatives have been the primary focus of research, given their unique electrical insulation and thermal conductivity.
- For novel lithium systems such as LSBs, boron nitride in separators and boron carbide in cathodes were reported to be effective at alleviating shuttle effects through different mechanisms.

9.2. Prospect

- Boron compounds will continue to be of interest in battery research and development, in lithium batteries and others. This can be evidenced by the boron studies on other novel battery systems, such as sodium-ion batteries and magnesium rechargeable batteries [151,152].
- While there are extensive studies on coin cells, more research on pouch cells or cylindrical cells is required to narrow the significant gap towards commercialization. Gaps include but are not limited to processing difficulty and raw material costs.
- In particular, the study of using more commercially available boron sources as precursors, such as boric acid and boron oxide, will be useful to achieve a fine balance between cost and efficacy.
- With a closer link to commercialization, studies on the use of boron compounds (e.g., boric acid) to assist the actual production environment will likely attract more interest.
- The topic of battery recycling is important and currently topical in the industry. Therefore, more studies on recycling or upcycling batteries with the use of boron will be likely.
- Given the diversity of available boron compounds, using a machine learning approach and leveraging existing extensive studies on boron compound might be of interest in guiding research [153].
- Leveraging machine learning may also help with the growing need for performance benchmarking among various types of battery systems.

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