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Review

Lithium-ion batteries under pulsed current operation to stabilize future grids

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SUMMARY

The large-scale utilization of renewable energy sources can lead to grid instability due to dynamic fluctuations in generation and load. Operating lithium-ion batteries (LIBs) under pulsed operation can effectively address these issues, owing to LIBs providing the rapid response and high energy density required. LIB deployment is also expected to reach 20 TWh from a vehicle-to-grid application by 2030. This review therefore highlights pulsed operation on LIBs for future grids, covering mechanisms, effects, and supporting hardware. Specific attention is paid to the fundamental mechanisms of pulsed operation on the stability of electric power system and micro-evolution in cells. The pulsed operation with appropriate parameters can provide superior effects for LIBs even under high-power charging and low-temperature operation. The hardware that supports bidirectional pulse is also introduced. This review presents the potential of LIBs participating in grid service via pulsed operation and may provide forward-looking guidance for the community.

INTRODUCTION

The world requires reaching net-zero carbon emissions by 2050 with a greener environment and considerable economic development.[1,2] This can be achieved only through a fundamental shift by fully using renewable energy sources (RESs).[3,4] Represented by wind, solar,[5] and hydro,[6] RESs contributed 11.7% to the 2020 annual electricity generation and will play an irreplaceable role in constructing a sustainable world.[7] Meanwhile, the generation of polytropic RESs changes with external conditions, such as climate and solar irradiance.[8,9] Both shifting power generation and load synergistically break the balance of supply and demand, endangering the stability and security of electricity supply (Figure 1A).[10,11] The conventional plants with high mechanical inertial response will gradually decrease under the pursuit of net-zero targets.[12,13] The inverter-based RESs will change the rotation-dominated grids to insufficiency of system inertia, inducing larger variations under the same imbalance power.[14] Therefore, supporting technologies, which input and output energy rapidly as required, are the bedrock of clean electricity with sufficient flexibility.[15–17]

Energy storage technologies are the key routes to provide flexibility for grids.[18–22] An overview of these energy storage technologies is highlighted in Figure 1B.[23,24] Of these technologies, capacitors, flywheels, and batteries have great potential for managing rapid power fluctuations in grids over short-to-medium

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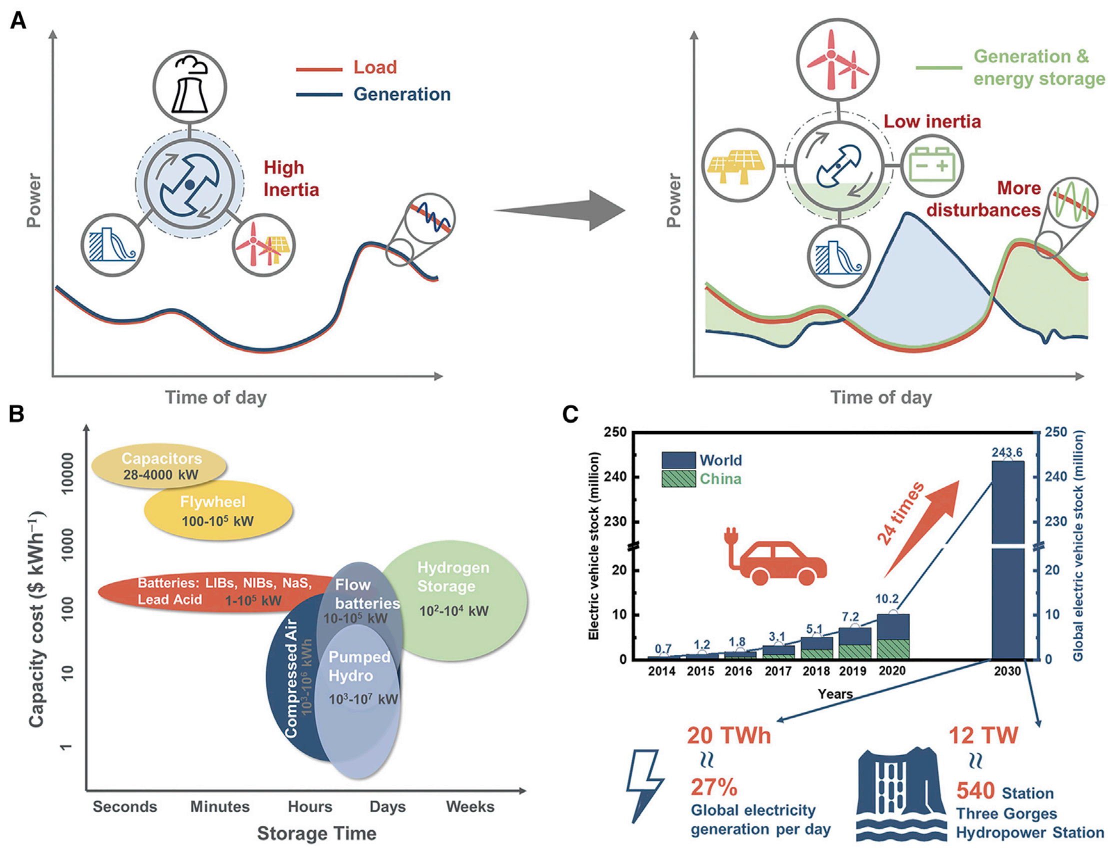

Figure 1. Potential of LIBs for future grids

(A) The challenges of lower inertia and more disturbances for future grids with large-scale RESs.

(B) Comparison between different storage technologies.

(C) Energy and power capacity from V2G regulation in 2030 all over the world.

timescales.[25,26] Both capacitors and flywheel technologies are limited by their low capacity, which challenges large-scale peak shaving and stability applications of grids.[27]

Battery technologies are one of the most suitable technologies for grid service within short-to-medium timescales. From BloombergNEF's prediction, we will need ~25 TW of wind, 20 TW of solar, and 7.7 TWh of battery power to achieve net-zero emissions.[28] Among the battery technologies, lithium-ion batteries (LIBs) possess a series of advantages, including low self-discharge rate, zero to low memory effect, long lifespan, high energy density, and portability.[29–33] They were installed at ~13.1 GW worldwide as of the end of 2020 and occupy >92% of the global battery energy storage systems, which is still far away from future demand.[34,35] However, the future stock of LIBs will dramatically increase with the growth of electric vehicles (EVs) and other electric appliances, which is an opportunity for distributed LIBs to apply in grid service. Specifically, only ~17,000 EVs were on the road in 2010. This number soared to 10.2 million in 2020, 44% of which came from China. According to the International Energy Agency, the global number of EVs will grow by >24 times, reaching 243.6 million EVs in 2030 (Figure 1C).[36] Such a vast amount of LIBs in EVs have a total capacity of ~20 TWh, equating to ~27% of the global electricity generation per day. Instant dispatch capability of 12 TW provided by LIBs in EVs in 2030 is equivalent to the installed capacity of the 540 largest hydropower stations in the world.[7] This large-scale deployment of LIBs in EVs will promote low carbon transport and improve the flexibility of the grid via vehicle-to-grid (V2G) solutions.[37,38] Therefore, the LIBs in EVs, LIB energy storage power stations, and utilization of 2nd life LIBs have the transformative potential to build a flexible and robust grid.[39]

These LIBs can contribute to stabilizing grids as an "energy sponge" due to their ability to rapidly absorb and release energy from short to medium timescales. Along with the variational demands of power systems, the needed power profiles vary significantly in terms of direction, amplitude, and duration, which can be considered a pulsed operation. Many requirements of grids can be satisfied by LIBs via pulsed operation, including active power balance, reactive power compensation, and accident recovery.[40] In addition, pulsed operations with appropriate parameters can improve the electrochemical performance of LIBs under high-power charging or low temperature working. For instance, Yin and coworkers[41] demonstrated that pulsed charging could significantly increase the charging speed, prolong the cycle life, and suppress lithium (Li) plating. Qin et al.[42] reported a rapid heating method through bidirectional pulsed currents to achieve a heating rate of 11°C min^{-1} and only 1% capacity decay after 170 h of continuous low-temperature heating. Power electronic to achieve pulsed operation on LIBs is also an emerging area, especially for devices with high-frequency switching and high energy efficiency. Specifically, excellent bidirectional pulse devices will provide greater flexibility for power systems and significantly improve the performances of pulsed charging and heating.[43]

In this article, we review the impact that pulsed operation has on LIBs for future grids, from the mechanisms and effects to the supporting hardware. Specific attention is paid to the fundamental mechanisms associated with pulsed operations on the stability of electric power systems and the (de)lithiation processes in LIBs. According to the initial understandings and related analysis, the influence of pulsed operation on LIBs is summarized, especially from the charging behavior at high-rate charging and the heating performance at low temperature. The supporting hardware for controllable input-output energy of LIBs is also introduced, which is the critical factor for practical applications. Although progress in pulsed operation on LIBs has been encouraging, it is insufficient to meet the various demands of future grids. The design principles of the aggregator are proposed based on the internal mechanism and latest research. This work, developing a deeper understanding of LIBs for grid service, will provide a roadmap for more robust future energy storage systems.

LIBs for grid stabilization

Power system stability is a condition of equilibrium between opposing forces. If a sustained imbalance is caused by a disturbance between a set of opposing forces, the system comes to an instability state.[44] This will lead to cascading outages and a shutdown of a significant portion of the power system. Disturbances in the form of load changes occur continually, and generators will also undergo more changes facing more RESs.[45,46] Furthermore, these inverter-based distributed RESs have the disadvantages of no grid-forming ability and lack of inertia.[47] Therefore, the future stable grid must use energy storage systems and suitable control methods to reach a new equilibrium state rapidly.[48,49]

The instability of the power system is a single problem, but it takes different forms, influenced by many factors.[50] Machine rotor speeds, system frequency, and network voltage are three main parts for stability.[51] Alternating current (AC) power systems traditionally require synchronous of each generator. The dynamics of relationships between power and generator rotor angle need to be controlled from this aspect

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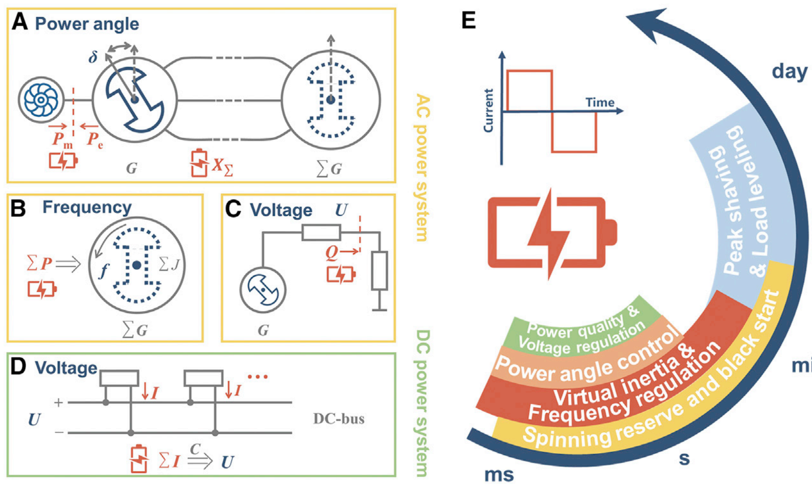

Figure 2. The applications and stabilizations for future grids through pulsed operation on LIBs
(A–E) The schematic stability of (A) power angle for AC grids, (B) frequency for AC grids, (C) voltage for AC grids. (D) voltage for DC grids, and (E) summary of different applications.

of stability.[52] Besides the fluctuation of load and power supply, various faults or cut-off transmission lines will induce the variation in the angular separation of generator rotors. Unstable generator rotor angle will finally cause generators to go out of service in the system. Forecasting load and planning dispatch of supply will often have unexpected short-term errors. Still, any such minor mismatch will cause the electrical frequency to deviate from the target value.[53] A constant frequency should be maintained throughout all of the connected systems. The frequency instability will decrease the quality of electric energy in the whole area. Furthermore, frequency deviation will affect the excitation of transformers and even bring about voltage fluctuation. The stability of synchronism is also a target for system operation in this case. A decrease in bus voltage is another typical unstable situation.[54] The voltage fluctuation is inevitable due to the variations of loads, network structure, and nodes of grids. Intermittent RESs will further aggravate voltage deviation. The collapse of voltage will cause the load device to become unstable and even affect the stability of generator rotors. The control of load voltage rather than the maintenance of synchronism will stabilize the power system in loads within an extensive area. In addition, direct current (DC) power systems mainly focus on voltage stability.[55]

LIBs can stabilize grids by providing a range of essential stabilizing and ancillary services (Figure 2).[56–58] Specifically, it is essential to ensure that all synchronous machines remain synchronous and connect with reliability.[59] LIBs with inverters can provide various synchronous services.[60] The position of the rotor axis of a certain generator G_x and the resultant magnetic field axis of infinite generators $\sum G$ in the whole system should be fixed while they are working normally (Figure 2A). The angle between these two axes is called power angle δ. Disturbances will accelerate or decelerate the power angle according to the swing equation.[61] Faults of the generator and related transmissions will bring about the instability of the power angle, causing the generator speed to go out of control. LIBs can act as additional power sources to regulate the accelerating power (the minus of mechanical power from prime mover P_m and electrical power P_e) on the generator side and therefore help stabilize the rotor in the transient stability problem.[62] LIBs can even adjust the transfer reactance $X_{\sum}$ to change the power angle at the transmission line side.[63]

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Traditionally, balancing authorities of grids are responsible for the management of the system frequency f. Short-term mismatches of supply and demand, which are called area control error (ACE), can be smoothed out within the time frame of seconds to minutes.[64] Frequency regulation follows the basic principle between the rate of change of frequency (ROCOF) $\frac{df}{dt}$ and the power gap $\sum P$, which is also determined by the total system inertia $\sum J$ (Figure 2B).[65] The governors of prime movers will regulate generator speed variations, called primary frequency regulation. The centralized backup power plants (e.g., hydropower, steam generators, combustion turbines) with automatic generation control (AGC) and some speed control units will participate in secondary frequency regulation and will also increase inertia. Distributed LIBs can take part in the primary frequency regulation and play the role of traditional governors via the droop control strategy based on the frequency derivation.[66–68] Moreover, aggregated LIBs can even supply secondary frequency regulation to match imbalance power in a relatively long period based on the ACE or even the AGC signal, which act as backup generators to provide isochronous control for power systems.[69] Virtual inertia gradually becomes mandatory for the direct current supplies, and LIBs with power electronic devices can also offer variable virtual inertial responses[70,71] through the ROCOF detection, compensating for the loss of conventional inertia.[72] Thus, the LIBs can improve frequency nadir and decrease the risk of system collapse.[73] In summary, LIBs will provide power and meet real-time misbalances via different control strategies to enhance frequency stability.

The equilibrium of reactive power Q is critical for the stability of voltage U (Figure 2C). Reactive power compensation and adequate reactive power supply under rated voltage will mainly sustain the voltage quality. Both the generator and transmission network voltage regulators are used for abnormal voltage. LIBs can also provide reactive power compensation through electronic power devices and then achieve voltage regulation for future grids.[51] For a DC power system, such as a DC microgrid, the bus voltage stability is determined by the sum of bus current $\sum I$ and bus capacitor C (Figure 2D). Distributed LIBs can directly regulate injected current via droop control.[74] Besides voltage regulation, power quality is a basic requirement for power systems with large-scale RESs. LIBs, with EV chargers or other power electronics, can act as harmonics filters and even inject current harmonics to power systems. Thus, the energy quality of power systems can be significantly enhanced.[75]

According to the above theoretical analysis, LIBs can stabilize the grid and supply various grid services to integrate RESs (Figure 2E). The peak shaving and load leveling provided by LIBs can support flattening the load curve of power systems over hours to days.[76] These energy and load regulations will obtain revenue from electricity markets by electricity price arbitrage. Furthermore, the integrity of the system is sustained without any tripping of generators or loads. Otherwise, some generators or loads may ultimately be tripped if stability is unable to be maintained. Power systems will emergently actuate relay protection, safety automatic, and splitting systems to avoid a system blackout in a large area. Aggregated LIBs can be constructed as a virtual power plant, providing spinning reserve service as a backup source for power support and a rapid response for black start after large area blackout.[77–79]

Fundamental understanding of pulse operation on LIBs

The specific input-output power demand can be allocated and implemented on the adjustable LIBs within a certain area, while for the single LIB, the applied signal is the

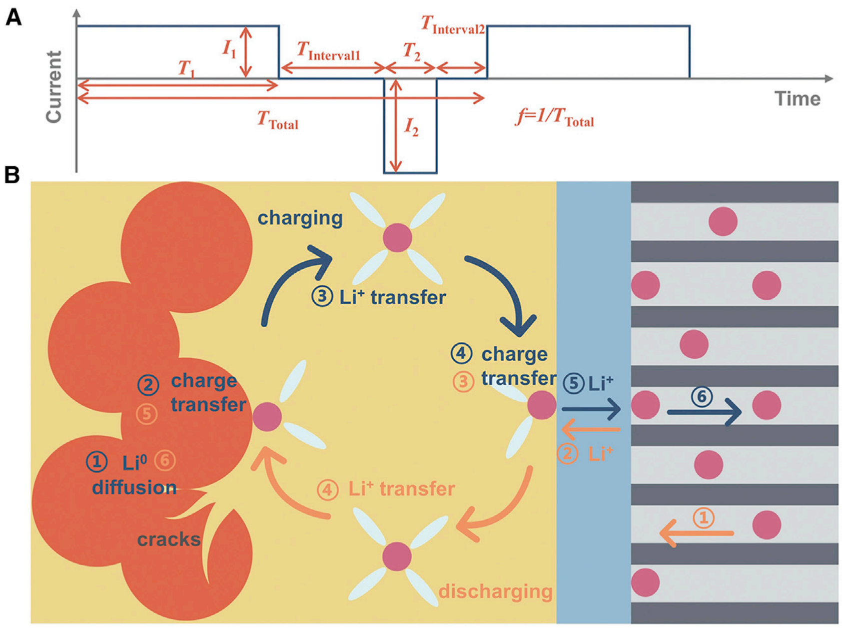

Figure 3. The mechanism effects of pulsed current for LIBs
(A) Definition of typical pulsed current.
(B) The Li^+/Li^0 (de)lithiation processes in LIBs.

pulsed operation. The controllable parameters of pulsed operation are limited, merely including current duration time. Regulating the above parameters should be based on the fundamental understanding of the internal evolution of applied LIBs.

The pulsed current operation is the basic unit of bidirectional interactions and grid applications, which periodically consist of at least two different levels. The recent literature often includes sinusoidal, triangular, squared, and other signal patterns belonging to pulsed waveforms.[80–82] The pulsed operations have been investigated widely, especially in the electroplating industry, in which pulsed profiles have been used to modify the composition, thickness, and morphology of the plated metal layers, reduce surface roughness, or achieve particular physical properties.[83,84] In the field of batteries, pulsed current profiles are often used for charging, discharging, or as a part of characterization techniques. A typical DC waveform is described in Figure 3A. $T_{Interval1}$ and $T_{Interval2}$ are the periods of the rest without current. T_1 and T_2 represent the length of the different pulsed currents of I_1 and I_2. T_{Total} is the whole duration of a pulse cycle. The frequency f specifies the number of complete cycles repeated in each second. The parameters of the pulsed waveform are of great importance for battery performance and the detailed statements as follows.

The profile of pulsed operations is decided synergistically by the grid demand and LIB characteristics. The (de)lithiation of LIBs spans multiple timescales, bringing about highly frequency-dependent behavior. Specifically, the reactions at the electrode/electrolyte interfaces are usually complete in a timescale of milliseconds. The reaction behavior depends greatly on the applied current density or overpotential.[85–88] The diffusion process (including Li^+ diffusion in electrolyte and interphase and Li^0 diffusion in active particles) proceeds over seconds to hours relying upon the diffusion coefficients and diffusion distance (Figure 3B).[89–91] The minimum pulse duration is limited by the electrical double layer (EDL) at the electrolyte/electrode interphase because the initial current contributes to the charging of the EDL rather than the lithiation of the electrode.[92] The time of non-zero current needs to be longer than the characteristic times of the transients, which are associated with the capacitive polarization of EDL.[93]

The current density of pulsed operation on a LIB determines the instantaneous power for the grid. The peak current levels in the pulsed operation are higher than in DC with the same average current. An increase in current density induces higher overpotential and faster reaction kinetics. The diffusion process rather than the reaction process becomes a rate-determined step.[94] Therefore, high Li^{+}/Li^{0} concentration gradients will form within electrode particles and electrolyte/electrode interphase during rapid (de)lithiation, which will result in particle cracks in cathode/anode and Li plating on the anode.[95,96] For example, Zhao and coworkers[97] simulated the effects of pulsed current operation on diffusion-induced stress in electrode particles. They found that pulsed current inevitably resulted in higher peak stress than DC charging with the same mean current and therefore the greater risk of cracking. Their model considered the diffusion in the particle but ignored the EDL charging, reaction kinetics, concentration gradient, and heat generation, which could significantly influence the Li^{+}/Li^{0} transport.[97] Pulsed operation engenders high current density but will minimize the concentration gradients during rest or reverse the current period in turn. Herein, opportune parameters of pulsed operation can meet the requirements for more stable grids and better battery performance. Why the pulsed operation within an improvement boundary can benefit LIBs is further systematically and fundamentally elaborated upon.

Pulsed operation is used widely in electroplating to obtain dense and uniform metal deposition.[98,99] Hence, pulsed operation technology has recently been proposed to prevent Li dendrite growth in Li-metal batteries.[100] Some previous work has focused on Li-dendrite inhibition via pulsed operation and concluded the mechanism from different views.[101,102] García and coworkers[103] adopted short periods of rest to reduce the gradient of Li^{+} concentration on the surface of the electrode and thus led to the uniform Li deposition and ultralong lifespan of full cells with >6,500 cycles at high charge rates. Aryanfar and coworkers[92] used Monte Carlo simulations. Furthermore, they demonstrated that the Li^{+} could replenish by diffusion in the rest period but converge on the dendrite tips due to fast Li^{+} migration in strong electric fields during the long-duration current. In addition to the Li^{+} distribution on the surface of the anode, Li et al.[104] presented the loose association between Li^{+} and anions in bulk electrolyte with square-wave pulsed current through molecular simulations. The pulsed current could improve the transportation of Li^{+} in electrolytes and therefore control the Li electrodeposition.

The (de)lithiation processes of LIBs mainly consist of Li^{+} transport in the electrolyte, Li^{+} diffusion through the solid electrolyte interphase (SEI), charge transfer on the surface of the electrode, and Li^{0} diffusion in the active particles (Figure 3B).[105–107] Although the above-mentioned studies focused mainly on Li-metal batteries, the fundamental understanding of pulsed operation on the transport behavior of Li^{+} in LIBs is also worth deepening our view. Furthermore, the mechanisms in LIBs are more complex due to the porous electrode and the competition between Li^{+} intercalation, Li deposition, and other side reaction.[108–111]

The SEI, which is formed via the electrolyte decomposition on the surface of graphite, aggravates the complication of electrode reaction of LIBs.[112,113] The formation of SEI consumes the active Li in the cathode at the first cycle. The cracks form in SEI during the following cycles due to the enormous volume expansion of anode, continuously consuming active Li.[114–116] However, the Li^+ needs to diffuse through SEI with a higher diffusion barrier than the electrolyte before or after the reaction.[117,118] The SEI plays a prominent role in LIBs. Some studies have been performed to explore the impact of pulsed current on the SEI.[119,120] Wang et al.[121] compared the SEI layers formed by 0.2–1 C constant current and pulsed current charging. The SEI constructed via pulsed charging exhibited more carbon and oxygen composition, indicating a higher percentage of organic compounds with alkyl and polycarbonate groups. Such composition of the SEI layer suggested a solvent-based rather than a salt-based SEI, displaying better thermal and kinetic stability. In their later work, Wang et al.[122] further assessed the effects of regular pulse and pulsed reverse waveforms on SEI formation. They found that using the reverse differential pulsed method led to a 58% reduction in the Li^+ diffusion activation energy of SEI and a 4.5% increase in accessible capacity at room temperature.

The charge transfer at the electrode surface is generally not a rate-determined step in LIBs, but the subsequent (previous) Li^0 diffusion through the electrode particle is prominent due to the low diffusion coefficients of common electrode materials. The diffusion coefficients of typical electrode material vary from 10^{-16} to 10^{-8} cm^2 s^{-1}.[116,119,123–125] Compared to the fast diffusion in liquid electrolyte with the diffusion coefficient of 10^{-6}–10^{-5} cm^2 s^{-1}, the sluggish diffusion behavior in electrode particles causes particle cracking under the diffusion-induced stress.[89,90] Pulsed waveforms have been proposed to release these concentration gradients and allow Li to diffuse through the particle during the (de)lithiation process, reducing diffusion-induced stress and hindering the cracks of particles. Kohl and coworkers[126] studied the structural changes in electrode particles, comparing 1 C DC and 0.5–1 C pulsed charging in a $LiCoO_2$ cell. The cycled graphite under pulsed operation demonstrated the thinner SEI layer but similar surface morphology of graphite compared with the DC charging. The improved structural integrity retention and the reduced extent of disordered cation were also achieved through the pulsed charging.[126] To summarize, when the pulsed operation is in the improvement boundary, the pulsed operation with different parameters will benefit other processes, including Li^+ transfer in electrolyte, SEI formation, and Li^0 diffusion in the cells. The better performance of LIBs even under high power and low temperature can also be obtained under the pulsed operation with appropriate parameters.

Pulsed charging effects

As the impacts on single cells, the pulsed operation displays extraordinary performance under higher power charging than traditional charging algorithms. Specifically, conventional charging algorithms have been developed, including constant trickle current (CTC), constant current (CC), and constant voltage (CV) approaches.[127] But both CC and CTC endure the safety risks of fast charging at high state of charge (SOC).[128] CV can hinder the hazards of high anode potential under the high SOC, while the high current during initial charging will impair the durability. Combining the advantages of CC and CV, the CC-CV approach is widely applied.[128–130] Nevertheless, the CV step at high SOC consumes longer. Fast charging with the continuous constant parameters is still hard to achieve due to the antagonism between the high current density and excellent battery performance.[131,132] When the battery undergoes high-power charging, Li^+/Li^0 diffusion rates through the different parts are diverse, easily causing the cluster of Li^+/Li^0 in interphase. Consequently, cracks in active particles, Li deposition, and other side reactions occur in the battery under high-power charging.[133] To consider the charging speed and battery performance, dynamic charging that breaks in the steady-state

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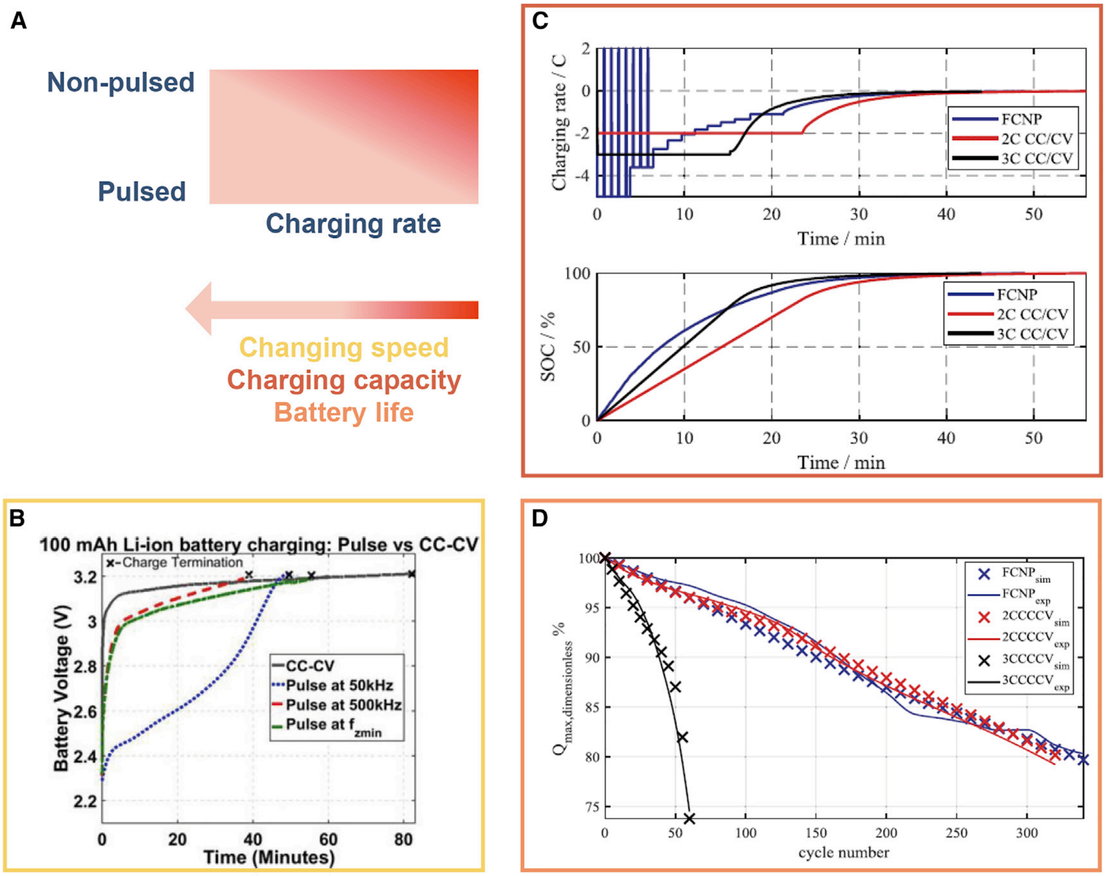

Figure 4. Improved battery performance at high charging rate by pulsed operation

(A) Outstanding performance under higher rate charging for appropriate pulsed charging methods.

(B) Unidirectional pulsed charging with a higher charging speed. From Amanor-Boadu et al.[138] Copyright 2021 IEEE.

(C) Bidirectional pulsed charging with a higher charging speed. From Song and Choe.[139] Copyright 2019 Elsevier.

(D) Bidirectional pulsed charging with a lower capacity degradation. From Song and Choe.[139] Copyright 2019 Elsevier.

Li^+/Li^0 diffusion behavior is an obvious idea to inhibit the local Li^+/Li^0 aggregation in the interphase. The multistage constant current charging (MCC) methods are one of the practical fast-charging algorithms.[134–137] Pulsed charging methods are a superior dynamic charging strategy by controlling more parameters than the current amplitude. Accordingly, the pulsed charging attributes to not only the better battery performance with a higher charging capacity, longer lifespan, and faster charging rate but also the potential function for grid-friendly charging. Moreover, the benefits of pulsed charging are more evident for high-power applications than other charging methods (Figure 4A).

Pulsed charging often consists of periodic steps of a low-rate current. The introduction of this step plays a vital role in controlling the Li^+/Li^0 diffusion behavior and eliminating local aggregation of mass transfer, especially under high-power charging. For example, Purushothaman et al.[131] constructed a model and then indicated that the pulsed charging with proper parameters may circumvent Li saturation at the graphite-electrolyte interface after CC charging and enable fast charging. Mayers et al.[140] developed a particle-based coarse-grained lattice model to reveal that the negative pulses will reduce the ion gradients and concentration overpotential on the surface of the anode. Yin and coworkers[41] explained that the lower-capacity decay of pulsed charging methods attributed to the lesser SEI formation, even under the fast charging speeds. Experimental results then further proved that pulsed charging could improve the reversibility of electrodes, maintain the stability of the $LiCoO_2$ cathode, inhibit the growth of the SEI, and enhance the active material utilization under a high power rate.[126] The experimental and calculational results synergistically provided reasons why pulsed charging operation could benefit charging performance.

Besides controlling mass transfer processes and affecting other reactions, pulsed charging can minimize impedance to achieve better performances comprehensively. The unipolar pulsed charging algorithms were explored to investigate the performance of V_2O_5|graphite 100 mAh LIBs. A pulsed frequency of 146.5 Hz with the lowest impedance of 4.18 Ω was selected. The LIBs demonstrated reduced charge time by 37% and enhanced charge efficiency by 3.2% compared with CC-CV (Figure 4B).[138] Unipolar pulsed charging with CV phase for 2,600-mAh NMC 18650 LIBs was also investigated. The proposed pulsed charging with 1 kHz was shown to reduce 17% charging time and achieve a comparable discharge capacity compared with CC-CV after 250 cycles.[141] Yin et al.[142] used a pulsed charging phase with variable unidirectional amplitude and variable frequency in place of the CV step in the traditional CC-CV. The results exhibited a fast charging speed nearly the same with 5 C CC-CV and a high cycle life the same with 1 C CC-CV. The bidirectional pulsed charging algorithm further contributed to better durability than the unipolar pulse, according to previous studies. Bidirectional pulsed charging can raise the temperature of LIBs and therefore inhibit Li plating.[143–145] Furthermore, Choe and coworkers proposed fast charging with negative pulses (FCNP), which take the place of the CV phase when LIBs are at high SOC to enhance the charging speed and the charging capacity. As a result, the charge time of FCNP is 43% shorter than 2 C CC-CV and 18% shorter than 3 C CC-CV due to the higher current adopted by the FCNP at low SOC (Figure 4C). Besides the faster charging speed, the FCNP also demonstrated a longer lifespan. The capacity decay of FCNP is 23% less than the 3 C CC-CV and almost the same as 2 C CC-CV after 60 cycles (Figure 4D). When fully charged, the energy consumption of FCNP and 3 C CC-CV were 593.9 and 591.4 kJ, respectively, suggesting the neglected energy cost of the negative pulse process during FCNP.[139] Beyond the benefits of faster charging and a longer lifespan, the LIBs also exhibit higher charging capacity. Majid et al.[146] analyzed the charging performance under pulsed and CC charging at a different frequency of ∼0.01–10 Hz. They tested 700 mAh LIBs, and the charged capacity of constant amplitude pulsed charging was improved compared to CC charging. Coincidentally, Ma et al.[147] used bidirectional pulsed current cycled with 0.9 s charging, 0.07 s discharging, and 0.03 s resting. When the amplitude of the current is raised from 1 C to 3 C, the additional charging capacity of pulsed charging compared with the CC is improved from 3.7% to 36.8% correspondingly, further demonstrating the significant advantages of pulsed operation under high-power charging.

Overall, the above-mentioned research has systematically revealed the detailed mechanisms and optimized performances and advantages of pulsed charging methods. From a practical point of view, many of the theories and experiments have further been converted into patents and potential applications. The pulsed charging methods have attracted a great deal of attention from industrial circles due to the excellent charging performance and ease of implementation, which will undoubtedly help develop future grid interactions.

Pulsed heating effects

The pulsed operation also can heat batteries themselves with superior performance and therefore is a promising method for cold environments. As is well known, the distance per charge of EVs and portable electronic devices will be significantly reduced at low temperatures due to the sluggish kinetics and transport properties.[148–150] When LIBs are charged/discharged at a low temperature, the degree of polarization will dramatically increase.[151] The capacity of LIBs correspondingly reduces because the LIB reaches the cutoff voltage early under high polarization. Li ions can plate on the graphite surface directly instead of intercalating into the graphite when the anode polarization continues to rise until the anode potential drops below 0 V versus Li^+/Li.[152] The Li deposition gradually deteriorates into Li dendrites, which may eventually pierce the separator, causing an internal short circuit and even thermal runaway.[153] Therefore, enabling LIBs at low temperatures with better performance is essential to meet the coming fossil fuel-free world. Preheating methods are proposed as part of the battery thermal management system and laid out by many companies. Through preheating to the room temperature before charging/discharging, the LIBs will exhibit increased power output, available capacity, as well as reduced risk of Li plating.[154–156]

Preheating methods can be divided into external and internal preheating according to the position of the heat source. Specifically, the external preheating refers to the heat that is generated from the outside of LIBs and transferred through different mediums, including hot fluid,[157–161] phase change material,[157,162–165] and electrothermal elements.[166–171] However, when adopting external heating, the different distances from the heat source and the loss of heat transfer synergistically induce the slow heating rate, low heating efficiency, and uneven temperature distribution of the battery and pack (Figure 5A).[158,161,170] Internal heating methods are related to the heat source inside the LIBs through tiny heating devices or the resistance of the LIBs themselves (Figure 5B). For instance, Wang and coworkers[172] demonstrated a self-heating Li battery (SHLB), which was constructed using an embedded metal foil and provided a fast heating rate through Joule heating Lei et al.[173] further settled the drawback of uneven temperature distribution inside SHLBs with the intermittent heating strategy. However, this method cannot precisely control the rate of temperature increase of each cell within a safe range due to the numerous switches in a battery pack. Nevertheless, the inserted heater will not only severely affect the energy density but also significantly aggravate the difficulty of the manufacturing process.

Another self-heating method uses the LIBs themselves as the heat source, overcoming the above-mentioned bottlenecks. This can be further divided into DC discharge preheating,[176] AC pulsed preheating,[177–182] and DC pulsed preheating,[174,183,184] according to the excitation waveform. DC discharge methods have excellent heating speed but will cause obvious capacity fade.[176,184] The pulsed heating methods, which typically consist of AC pulsed heating and DC pulsed heating, can also supply outstanding heating function and even reduce polarization and side reactions to achieve a negligible capacity decay.[42,174,181,182] Both the AC and DC pulsed heating methods rely on Joule heat and the reaction heat. Therefore, thermo-electrochemical coupling models will need to consider not only DC internal resistance characteristics but also frequency-dependent behavior.[42,174,177–182] Furthermore, lumped parameter models based on equivalent circuit models and three-dimensional finite element simulation can be used to describe the production and distribution of temperature in the single cell and battery packs.[178,182]

AC pulsed heating methods have been used on the cell or the battery pack and have shown a high heating rate of 1°–4°C min^{-1},[177–182] and slight temperature differences

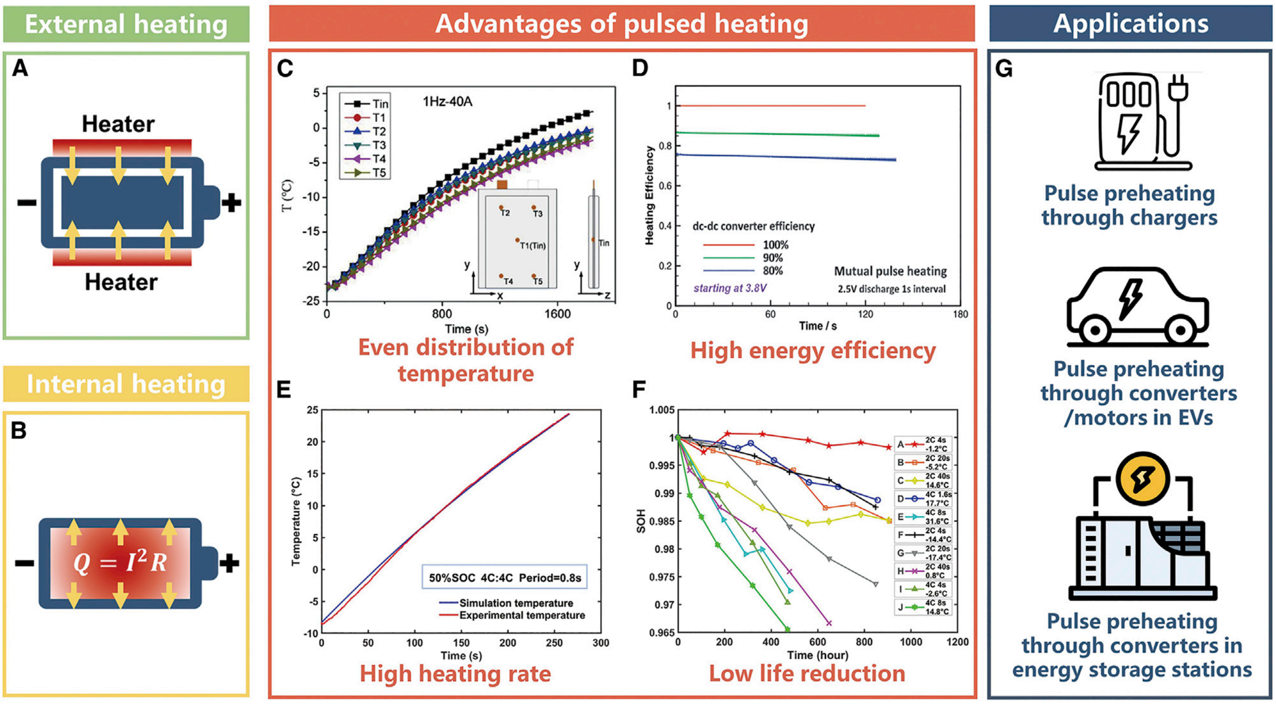

Figure 5. Enhanced heating performance by pulsed operation

(A) External heating.

(B) Internal heating.

(C) The uniform temperature distribution of pulsed heating. From Zhu et al.[174] Copyright 2017 Elsevier.

(D) The high heating efficiency of pulsed heating. From Ji and Wang.[175]. Copyright 2013 Elsevier.

(E) The outstanding heating rate of pulsed heating. From Qin et al.[42] Copyright 2020 Elsevier.

(F) The negligible degradation of LIBs after hundreds of hours of pulsed heating. From Qin et al.[42] Copyright 2020 Elsevier.

(G) Applications and scenarios for pulsed heating.

of 1.6°C on the surface of the cylindrical battery or different cells of a pack.[180,182] Such self-heating methods cannot exacerbate the inconsistency of battery packs. The effects of the heating current frequency and amplitude on the heating and battery performances were also investigated.[179–182] For example, to prevent Li deposition during heating, anode potential is used to design the maximum amplitude of the charging half-cycle of a sinusoidal wave.[181] Overall, AC pulsed heating has almost no impact on the health of LIBs according to designed parameters.

DC pulsed heating methods showed better heating performances than AC pulsed heating methods in many previous studies. Zhu and coworkers[174] reported an even temperature distribution with a lower temperature difference of 2°C on the surface of a pouch cell under a high current (Figure 5C). Ji and Wang[175] presented a high energy utilization and increased efficiency converter efficiency, ranging from 0.85% to 0.86% for 90% converter efficiency and from 0.75% to 0.73% for 80% converter efficiency using mutual pulsed heating through bidirectional DC/DC converters with different efficiencies (Figure 5D). Qin et al.[42] proposed a bipolar DC heating method with an enlarged voltage range and an ultrahigh heating rate of 11.3°C min^{-1} was achieved for both higher and lower SOCs (Figure 5E). The higher heating rate compared with the AC heating method is due to not only the voltage limitation but also the higher mean Joule heat of DC operation when the maximum amplitudes of DC and AC are the same. Besides the heating performance, the state of health (SOH) of the battery after hundreds of pulsed heating cycles were evaluated with critical metrics, including battery capacity, DC internal resistance, and electrochemical impedance spectroscopy (EIS), suggesting that the bipolar DC pulse current does not aggravate cell degradation.[174] Even under the exceeded voltage protection limitation, ~1,000 h of accelerating heating durability experiments revealed that thousands of heating cycles could be implemented with <1% SOH decay (Figure 5F).[42]

As for the practical application, no matter whether it is AC or DC pulsed heating, no modifications of the battery cells and modules are required and therefore no additional space inside vehicles and no increase in system complexity are needed. The cost of battery manufacturers and original equipment manufacturers can be minimized. Current studies on pulsed preheating methods mainly generated the pulsed excitation through external pulsed power or LC resonant circuit. For available applications, combined with bidirectional charging pile implementation, pulsed heating could be realized before or simultaneously with the charging process, solving the low-temperature charging problem.[145] Pulsed heating could combine with pulsed charging to heat and charge LIBs simultaneously. Pulsed heating before conventional CC-CV charging could cut the whole charging time by 23.4% and improve the capacity by 7.1%.[185] Besides external excitation-like chargers, mutual pulsed excitation was achieved with DC/DC converters[175] or vehicle motors[186] (Figure 5G), which will be discussed in the following sections.

In summary, compared with other heating methods, pulsed heating methods, including both AC and DC pulsed methods, exhibit superior heating performance, extraordinary battery performance, and great potential for large-scale application after parameter optimization. In addition, DC pulsed heating shows a higher heating rate than AC pulsed heating, and the DC waveforms are easier for power electronics implementation. Hence, pulsed heating methods, especially DC pulsed heating, are suitable for higher energy density applications.

Hardware for pulsed operation

Controllable bidirectional power interactions for LIBs should be achieved by power electronics. LIBs are passive storage and power devices whose input conditions (current or voltage) need to be regulated by power electronics. To accomplish all of the above-mentioned pulsed operation scenarios, power electronics with high safety, high dynamic performance, and high efficiency are urgently demanded in the field of energy storage stations and EVs for grid applications.[187]

LIBs cooperate with power converters to realize feasible charging or discharging with DC or AC sources.[188] In the topologies of converters, AC/DC acts as a rectifier and will function as the reactive power compensation device and the active power filter.[189] The power factor correction (PFC) and harmonic suppression are critical to generating high-quality energy and then injecting it into AC grids.[190] These functions are achieved mainly by controlling the capacity bus capacitor. In summary, the bidirectional AC/DC converter is the crucial part to interact with the AC power system directly and should have better performance, as mentioned above. However, DC/DC converters directly connect to LIBs and regulate their current and voltage according to the requirement of the whole system (Figures 6A and 6B).[191–193] For V2G interaction and pulsed charging or heating through charging infrastructures, bidirectional chargers, whether on-board or off-board chargers, are connected between the power sources and the LIBs of EVs, usually with a topology with bidirectional AC/DC and DC/DC (Figure 6B).[194,195] The core parts of the bidirectional chargers are also DC/DC converters, which are indispensable to match a wide voltage range

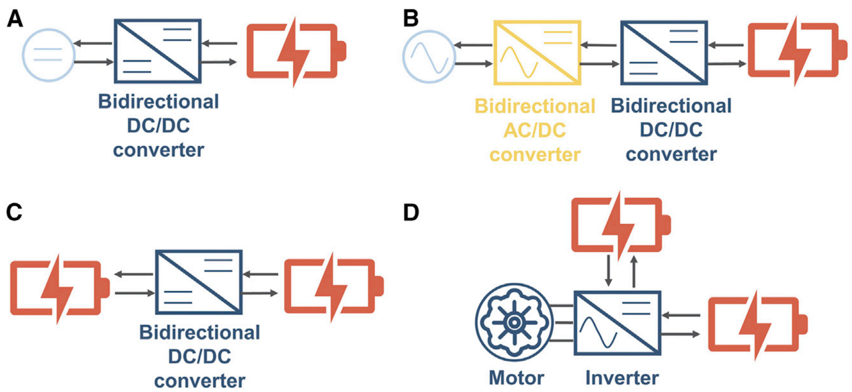

Figure 6. The topologies of supporting hardware for LIBs

(A) DC source and bidirectional DC/DC converter for pulsed operation.

(B) AC source, bidirectional AC/DC, and DC/DC converters for pulsed operation.

(C) Bidirectional DC/DC converter for mutual pulsed operation.

(D) Motor and new circuitry as bidirectional converter for mutual pulsed heating.

from ∼200 to 500 V for a battery pack of EVs, or even 800 V for the future high-voltage electrical architecture of fast charging EVs.[196] Beyond this, LIB swapping stations for EVs usually have chargers composed of unidirectional DC/DC for the energy supply of swapped battery packs.[197] These backup packs are the potential energy storage resources to participate in ancillary services and price arbitrage. Therefore, the same bidirectional charging topology can be applied for these smart swapping stations (Figures 6A and 6B). Moreover, if additional DC/DC converters are installed and connected to swapped LIB packs, they can also provide fast charging services to other EVs and thus compose a swapping-charging combined station. In addition, LIB packs in swapping stations, energy storage stations, and other battery groups can interact with one another to achieve mutual pulsed operation for heating via bidirectional DC/DC converters (Figure 6C).[175,186]

An isolated bidirectional DC/DC converter is a common device for practical application satisfying electrical safety requirements in the above-mentioned applications.[198,199] The dual active bridge (DAB) and the capacitor-inductor-inductor-capacitor (CLLC) are 2 kinds of mainstream topologies for isolated bidirectional power transmission.[200] The DAB is suitable for high-power applications exceeding hundreds of kilowatts, but it suffers from limited soft-switching regions,[201] along with complex control of performance improvement and bidirectional switching.[202] Compared with the DAB, CLLC demonstrates incredible bidirectional working performances with high switching frequency, high energy density, and high efficiency.[203] CLLC has excellent soft-switching ability for both forward and reverse operations,[204] which contributes high efficiency to the operation of full-load range.[205] Therefore, CLLC is a promising topology for high-frequency bidirectional pulse power implementation. Easy installation or integration in power electronic devices makes it suitable for pulsed applications on energy storage systems and vehicle chargers.[206,207]

Furthermore, pulsed heating can be realized in the vehicle through the motor and inverter in EVs, leading to innovation in on-board fast heating.[186,208] A new type of topology for the motor and inverter can work as a bidirectional DC/DC and transfer energy between 2 groups of LIBs (Figure 6D). This circuitry topology supports >4 C pulse currents on LIBs of passenger EVs, and therefore achieves a remarkable fast heating process. This new circuitry only adds a relay for operation mode switching.

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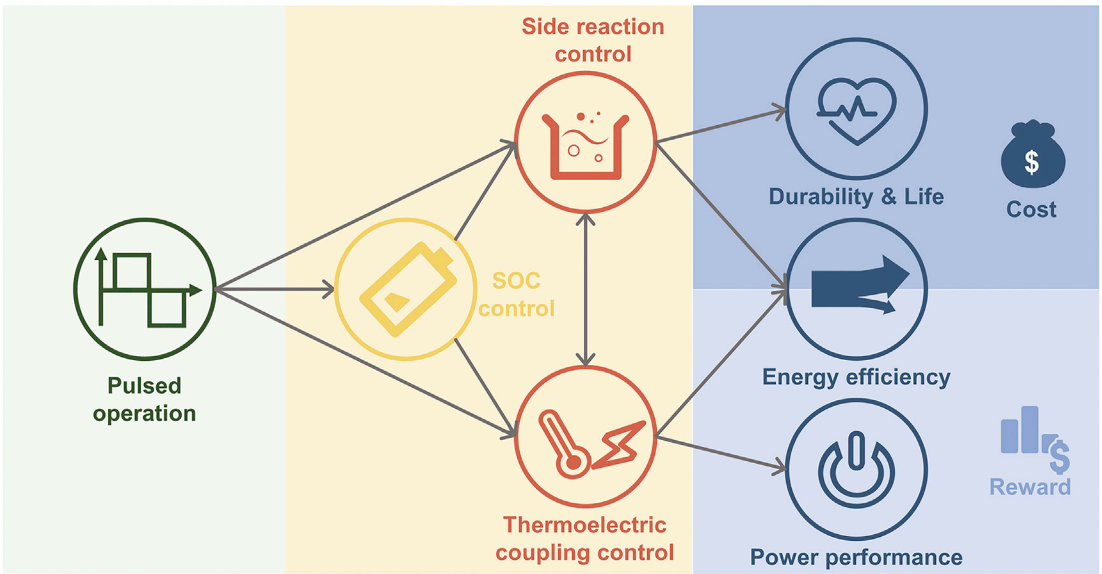

Figure 7. Parameter summary and design
The parameters of pulsed operation should consider SOC changes and then the side reaction and thermal effects; the durability and lifetime of LIBs, energy efficiency, and power performance of interaction together will determine the economy (cost and reward) of grid and other applications.

The control logic for the pulse width modulation of the motor will use a new voltage vector sequence. As a result, a heating efficiency >80%, a high heating speed of 7.2°C min^{-1}, and negligible degradation of LIBs can be easily achieved on-board. Furthermore, noise and vibration are hopefully reduced to realize comprehensive installation in EVs.

In summary, DC/DC converters are core parts for bidirectional power flow controls and pulse-curve regulations on behalf of power electronic devices. Specifically, CLLC topology and innovative motor drive circuitry contribute to pulsed currents with high frequency, high power, safety, high efficiency, and low cost.

Parameter summary and design

The parameter design is critical for different application objects. Applications for grids will passively depend on the imbalanced power demands of the grids. Nonetheless, each individual LIB cell or LIB subsystem has flexibility according to the aggregator, which connects different LIBs and gathers their power to interact with grids under a certain control strategy. Thus, each cell or subsystem can be optimized to a more economical working condition, and meanwhile, the whole system can provide better grid service. The basic rules of parameters and the behind mechanisms of performances are the critical parts for aggregator design (Figure 7). The ability of power response and the cost of LIBs will directly determine the total revenue of grid services. Specifically, the state of power (SOP), the energy efficiency, and the life of LIBs are the dominant factors. The capacity, maximum power, and duration for one direction operation of LIBs will determine the interaction performances. The current amplitude and pulsed duration will also affect the life of LIBs. Different temperatures will induce various energy efficiency and durability of LIBs. Therefore, a reasonable limitation of pulsed parameters and the all-time temperature control should be delicately designed to achieve a higher reward and a lower cost. Besides temperature, SOC control is critical for improving SOP, energy efficiency, and durability. Even though the comprehensive strategy has not been studied considering LIBs, the basic relationships between parameters and pulsed charging/heating can guide the aggregator design.

The detailed parameters of pulsed charging methods include amplitude, frequency, and pulse interval. The pulsed charging methods have various parameter ranges for benefit effects because effects are highly dependent on the type of battery, external environments, and battery states. Design rules do exist to guide the selection of parameters, but they are applied for different conditions. The literature has shown different effects of pulsed charging.[146,209,210] For pulsed charging without negative current, finding the frequency with the lowest impedance is the optimal strategy to achieve a faster charging speed, higher charging efficiency, and lower degradation, according to the recent literature.[138,141,146,211,212,213] However, the optimal frequency of LIBs dynamically changes during the process of charging. Dynamic parameter strategies with optimal parameter searching stages are introduced to further improve charging effects. The charging speed of the variable frequency strategy is 24% higher than the standard CC-CV and 10% higher than the fixed-frequency pulsed charging in the literature.[128] The duty ratio indicates the ratio of current duration and a whole cycle, and an additional optimal duty ratio searching stage is also added to the maximum charging speed for different SOCs.[212] This method with variable frequency and duty ratio achieves 46% higher than fixed pulsed charging in a 0%–80% SOC range. From a circuitry modeling analysis perspective, LIBs behaved like a low-pass filter due to the EDL. High-frequency pulse has a non-destructive effect but also is hard to charge energy into LIBs. The concentration gradients increase and anode potential becomes more negative when frequency decreases, thus leading to side reactions and decaying battery performance.[214]

Bidirectional pulsed charging is more durability oriented, as verified by the experimental and theoretical results. The relationship between parameters of bidirectional pulsed charging (temperature variation, frequency, and current rate) and side reactions was studied in detail.[41,215] Bidirectional pulsed charging can raise the temperature of LIBs and then improve mass transfer. Therefore, it can avoid side reactions in larger allowed current and frequency ranges.[143–145] Detail values of pulse also relate to ambient temperatures and the thermoelectric coupling features of LIBs. However, there is still a lack of systematic experimental research on the mechanisms of negative pulse, and bidirectional and unidirectional pulsed charging need to be compared for different LIBs and other conditions in future studies. As for the amplitude of current, it is well understood that the high amplitude will result in a slow kinetic of Li^+/Li^0 diffusion at the interphase and thus lead to various side reactions, while low amplitude will cut down the charging speed. In addition, SOC affects the feasible amplitude of charging current due to the characteristics (e.g., electrical conductivity, electrode potential, ionic conductivity) of electrode materials under different SOCs. Therefore, combining various amplitude operations for different SOCs is essential to balance high charging speed and battery performance for both bidirectional and unidirectional pulsed charging.[216–218] The on-board pulse charging algorithms and corresponding update methods will construct the pulsed charging algorithm, which adapts diverse cells under the different aging paths and SOC. This involves monitoring technologies based on multi-sensors with internal and external measurements, especially reference electrodes, to detect the anode potential.[219,220,221] In addition, if the differences in Coulombic efficiency in a small SOC range reach a specific value for some types of LIBs, then pulsed operation will have the potential ability to auto-equalize SOCs for SOC-inconsistent cells in the serial battery module.[222,223]

Much research has focused on the effects of pulsed parameters on heating and battery degradation, demonstrating the practical prospects of pulsed heating methods facing the dilemma of the LIB at low temperatures. The heating effects of bidirectional pulsed operation without time interval are better than unipolar current because of Joule law, especially when considering the durability of LIBs and the continuity of the heating process. The higher the amplitude, the larger the heat generation. The frequency has a slight effect on heating. Usually, the lower the frequency (lower than 100 Hz), the higher the heating speed, but this effect weakens when the duration is approximately tens of seconds, which is caused by the feature of polarization resistance. When the frequency is much higher, the heating effect is determined by the ohmic impedance in the frequency domain. However, there is no clear mechanism to elucidate why suitable pulsed heating methods do not impair the cells in a certain boundary, but the apparent relationship of parameters has been revealed. The parameters have opposite effects on heating and durability. The large duration and amplitude will cause a high risk of Li plating and other side reactions to accelerate the battery degradation. The parameter sensitivity analysis has shown that the frequency is more sensitive to battery SOH decay than heat generation, and amplitude is more effective in increasing heating speed in a modest boundary. Accordingly, higher amplitude and shorter periods are suggested to achieve an optimized pulsed heating strategy. SOC also plays a vital role in heating parameters. The internal ohmic resistance is normally much larger at lower SOCs (∼0%–20%). The resistance slightly changes when SOC increases. Some may have larger resistance at certain SOC ranges for different electrochemical systems. When SOC is lower, the discharge current should be mainly restricted due to the voltage limitation, while the anode potential should be focused on limiting excessive charging current. Therefore, the heating parameters should use a larger amplitude within the boundary of non-destructive bidirectional pulse and a higher frequency to balance durability and Joule heat.[181]

In summary, the general rules of parameter influence are summarized through the studies of pulsed heating and charging. Fast charging and low-temperature heating need to balance some different objectives. The purpose of pulsed heating is maximum heat generation, while pulsed charging aims to increase effective charging, and improving life is the same target for both.[224,225] For the grid application, the above-mentioned mechanisms are also applicative.[226] The thermoelectric coupling mechanism and the degradation mechanism are the principles for the parameter and logic design of real-time control (Figure 7).[227] SOC control is the core part, which then affects the temperature and side reaction control. Together, they control the power performance, energy efficiency, and degradation of LIBs during interactions. The hardware should be considered together with LIBs for the strategy design of different applications. The hardware performances will restrict the selection of parameters, and some non-ideal waveforms will affect control performance for both grids and LIBs. The hardware-LIB coupling features, more detailed relationships, and comparisons for the parameter impacts are still unclear.

Conclusions and perspectives

The green world urgently requires diverse energy storage technologies to integrate renewable energy. As the most popular energy storage devices used in consumer electronics and EVs, the LIBs operated under pulsed current are one of the most competitive technologies to provide flexibility for future grids within a short-to-medium timescale. This review summarizes the multi-scale considerations from microscopic effects (mass transfer and charge transfer) to macroscopic influences (at high charging rate or low temperature) of LIBs. To achieve grid-friendly utilizations

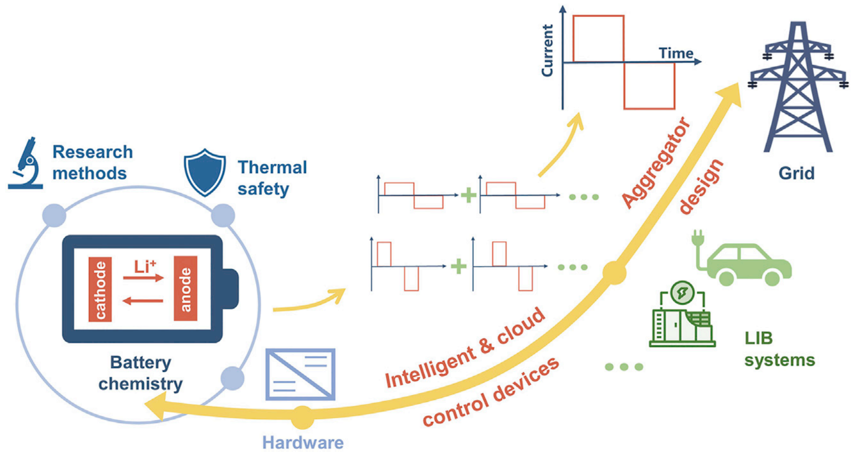

Figure 8. Gaps and perspectives

Deconstructing the variation of battery performances, safety, and the internal evolution under pulsed operation through advanced research methods; the intelligent cloud control devices and aggregators for effective charging, heating, and grid applications.

served by LIBs under the pulsed operations from fundamental understanding, aggregator design, to hardware support, we still have a long way to go. Many issues need to be considered, as follows (Figure 8):

1. Battery chemistry. Understanding the internal evaluation of LIBs under pulsed operation can guide the design principle from materials, battery configuration, and pulsed parameters (e.g., amplitude, frequency, waveform). LIB is a complicated system, in which the composition and structure of each part of the LIB are constantly changing during working conditions. The surface chemistry and the material evolution at the interphase between cathode/anode and electrolyte are also unclear but significant to construct a safe and long-lifespan LIB under pulsed operation.
2. Thermal safety. Ensuring safety is the extreme priority of LIBs to apply in energy storage systems. Enhancing the safety of the battery system primarily requires efforts in the following two aspects. First, it is essential to clearly understand the failure mechanism of LIBs under pulsed operation, including thermostability and heat generation rule of different materials. Second, figuring out the thermal runaway boundary is the last line of defense to ensure safety. Comprehensive considerations should include material design, battery manufacturing with zero defect, aging route detection, thermal management, and control boundary design to guarantee intrinsic security.
3. Research methods. Research characterizations of this system from a single battery to grid need further development. The inter evolutions of a single LIB are a black box for researchers. An *in situ* characterization on a working LIB can demonstrate the chemistry mechanism and evolution during the charging and discharging process. In addition, the system from LIBs to grid is complicated, involving many multi-dimensional parameters. Introducing and promoting artificial intelligence methods can effectively advance the all-around exploration, including material selection, battery design, capacity allocation of LIBs, optimized control, intelligent aggregator, and Internet of Things (IoT) for power electronics.
4. Aggregator design. When the concrete energy demands of grid service come over a period, allocating and controlling each LIB system in certain areas are

huge challenges. Besides the basic mechanisms of LIBs under various frequencies and different waveforms, future work should focus more on real-time optimization and dispatcher control. The revenue of interaction and the cost of LIBs are the key factors, but other opportunity costs and nonphysical factors will dramatically increase the complexity, especially the travel demand and individual preference of EV drivers in a V2G scenario. Therefore, the target of a well-designed aggregator is that diverse LIBs at different states and conditions can contribute better to both the demands of the grid and themselves. Grid services in all climate, grid-friendly charging, and heating algorithms may also be realized by considering different aspects synergistically.

5. Intelligent and cloud control devices. The controllable bidirectional electronic devices play a key role in interacting with aggregators, such as detecting states and actuating the control command of the corresponding pulsed operation on LIBs. Specifically, only with the DC/DC converters and other power electronic devices can LIBs act as virtual power plants, directly accepting scheduling and dispatching power interactions.[192,193] As for the converter itself, the isolated bidirectional DC/DC converter with greater single module power and the higher switching frequency is pursued urgently, thereby reducing the number of paralleled single converters and improving safety under distributed high-power scenarios. Meanwhile, bidirectional hardware can easily estimate or measure the states of LIBs and act as an active diagnosis device. For example, incremental capacity analysis can be achieved, and hybrid pulse power characterization, titrations, and EIS are analyzed through the pulsed operation. Therefore, power electronic devices with cloud control will become the future-oriented distributed energy Internet trend.

The pulsed operation of batteries has significant potential to transform the performances of LIBs for stable grids with high-penetration RESs and driver-convenience EV applications. However, the optimal selection of parameters requires a deep understanding of the underpinning principles and design of the system. Solving these problems will require a multidisciplinary approach, with cooperation from chemistry, physics, materials, engineering, and computer science.

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AUTHOR CONTRIBUTIONS

Y.Q. and X.C. conducted relevant literature research, co-wrote the manuscript, and revised the manuscript. A.T., H.C., and Y.W. helped write parts of the original draft. H.Z., Y.L., Z.C., and J.H. contributed to the literature collection. J.D., X.H., and L.L. provided supervision throughout the manuscript-writing process. B.W. and K.S. contributed to the conceptualization. M.O. and Q.Z. contributed to conceptualization and revision. All of the authors discussed the manuscript.

DECLARATION OF INTERESTS

Some of the authors (Y.Q., Y.L., L.L., J.D., and M.O.) hold patents in China (patent nos. 2019205376921, 2019108064362, and 2019205376921) related to this work. The other authors declare no competing interests.

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