



# The next generation of fast charging methods for Lithium-ion batteries: The natural current-absorption methods

T.G. Thusitha Asela Bandara<sup>\*</sup>, J.C. Viera, M. González

Department of Electrical Engineering, University of Oviedo, Gijón, Spain

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## ABSTRACT

The fast charging of Lithium-Ion Batteries (LIBs) is an active ongoing area of research over three decades in industry and academics. The objective is to design optimal charging strategies that minimize charging time while maintaining battery performance, safety, and charger practicality. The main problem is that the LIB technology depends on multi-disciplinary engineering factors that form rapidly varying intrinsic states in the cell during the charging process. These factors take the form of interdependent electrochemical, structural, and thermo-kinetic perspectives. Here, the list can grow as electrochemical changes; charge transfer, ionic conductivity, structural transformations; mass/particle transfer, migration, diffusion, and thermo-kinetic exchanges; phase transitions, heat effects, and collectively their inter-dependencies. Fast charging intensifies this varying nature making it very difficult to achieve an optimal process. In fact, many charging strategies fail to adhere to such rapid variations and are based on predefined/fixed parameters such as voltage, current, and temperature, individually or collectively, that enforce and aggregate stress on the LIBs. Consequently, fast charging accelerates battery degradation and reduces battery life. In order to facilitate the design of optimal fast charging strategies, this paper analyzes the literature around the influences of intrinsic factors on the LIB charging process under electrochemical, structural, and thermo-kinetic perspectives. Then, it examines the existing charging strategies with a new categorical analogy of; 1) memory-based, 2) memory-less, and 3) short-cache, showing their efforts to achieve the optimal charging targets and challenges in adapting to the demanded intrinsic variations. Accordingly, a potential paradigm shift for the next generation of LIBs' fast charging strategies has been identified in the new area of short-cache-based natural current-absorption-driven charging strategies. Importantly, this new approach is competent in bringing the practical intelligence necessary to adapt the control of LIB fast charging over rapid intrinsic variations.

## 1. Introduction

The design of optimal charging strategies for Lithium-ion (Li-ion) batteries has become extremely important for electronic devices ranging from portable electronics (smartphones [1], biomedical applications [2], power tools [3,4]), battery-powered electric vehicles (e-bikes [5], EVs [6,7], e-busses [8], e-trains [9] & e-airbuses [10,11]) and battery energy storage systems (BESS) [12–14] for uninterrupted and more stable operation of smart-grids. Over three decades of research, since the invention of the commercial LIB by Sony in 1991 [15], the fast-charging solutions for LIBs have dispersed in various directions seeking an optimal balance between battery performances, safety, and the practicality of chargers. Firstly, these directions include the charging strategies [16–22] to improve charging performance. Secondly, different

alternatives for fast charging demands; the new battery materials [23, 24] to enable high energy and fast charging capabilities, and chemical/structural advancements [25,26] in battery elements (electrode, electrolyte, separator) [27] to enhance the tolerance against charging effects. However, as these attempts face issues in battery performance, health, and safety that hinder the sustainability of LIB electrification, the need for optimal fast charging has intensified.

At the battery cell level, the multi-disciplinary engineering factors of LIB technology influence its charging process over the mass and charge transfer of particles/ions. These factors include a) electrochemical reactions/changes [28,29], b) structural behavior/transformations [30–32], and c) thermo-kinetic exchanges [33,35]. The electrochemical reactions change the factors like charge transfer resistance, ionic conductivity, cell voltage, current, and cell capacity. The structural

<sup>\*</sup> Corresponding author.

E-mail addresses: [bandarathannehene@uniovi.es](mailto:bandarathannehene@uniovi.es) (T.G.T.A. Bandara), [viera@uniovi.es](mailto:viera@uniovi.es) (J.C. Viera), [mgonzalez@uniovi.es](mailto:mgonzalez@uniovi.es) (M. González).

behavior in cell components (electrode, electrolyte, SEI-solid electrolyte interphase, and separator) and interfaces (electrode/SEI/electrolyte) impacts Li-ion mass/particle transfer, migration & diffusion, and bonding phenomena. The energy exchanges and kinetic variations over the electrochemical and structural transformations influence the thermo-kinetic effects such as phase transitions [32,36] and heat effects. Altogether, their inter-dependencies collectively contribute to rapidly varying intrinsic states. Moreover, as the high rates of current aggravate the charge and mass transfer during fast charging, across and within the battery components, the varying intrinsic nature gets intensified and makes it even more difficult to measure.

Due to the rapidly varying intrinsic, the real-time measures of multi-disciplinary phenomena or their antagonistic effects on charging are essential to proper control of the charging process, but it is quite challenging. For example, the electrochemical characterization techniques such as X-ray diffraction (XRD) and absorption [34], electrochemical impedance spectroscopy (EIS) [29], potential step chrono-amperometry (PSCA) [35,37] are such measurement techniques that are far from reality in practical/commercial chargers. Challenges in effectively incorporating these techniques in a practical charger limit the possible types of measurements mainly to voltage, current, and temperature of LIBs for charging controls.

With such limitations, the fast charging can be realized by different control strategies such as design patterns [38,39], responsive [26, 40–42] and interactive regulations [43], battery model-based [44] and statistical/mathematical [16,17,45–47] controls, and maintaining electrochemical relationships [40,48]. In most cases, these strategies employ a set of high current [49] and/or voltages [50,51] continuously, which oppose the demanded proper control over the rapid intrinsic variation of the cell components. This hostility to the intrinsic variation influences numerous potential adverse effects such as Li-plating/Li-deposition [52], Li-dissolution, loss of active materials, blocked pores in interface structures, micro-cracks in electrode structures, and mechanical stress. They lead to critical failures in battery performance, health, and safety when propagated and accumulated [53, 54] over the battery life. These include capacity loss, energy loss, premature aging, thermal instability, unexpected battery drains, thermal runaways, and sometimes even causing fires and explosions when such adversary conditions are aggravated suddenly. Consequently, the fast charging strategies grow uncertainty in the process that damages the reversibility of electrochemical reactions and promotes battery degradation. Furthermore, despite the extensive research on fast charging in both the academic and commercial contexts, the commercial fast chargers still take about 1–2 h [55,56] for smartphone batteries which drives the users to alternatives such as power banks, and in the range of about 4–10 h for general-purpose chargers of EVs & PHEVs [7,57,58] that cause the range anxiety among their drivers.

As alternatives for fast charging, the new battery materials [23,24] and chemical/structural advancements [25,26] add another layer of complexity to the charging problem. Here, the enhancements in the battery production processes such as doping, coating [24,59], layering, and new chemistry [60,61] can be identified as pathways toward new materials that increase the storage capacity, energy, cycle life, and power capabilities [62]. In terms of chemistry/structural improvements, the usage of thinner/wider electrodes [63], nano-structural advancements in electrodes (nano-crystallization [64,65], spinal structures [66], exfoliating edges [25]) can be identified. Inevitably, these new LIB technologies need advanced experimental validations, aging analysis, and reliability evidence which takes a considerable time before they mature enough to be effectively commercialized.

As described, the incorporation of battery intrinsic variations within the charging process would make it possible to achieve optimal fast charging benefits. Hence, it is important to investigate possible approaches to overcome the implementation problems. Although some reviews are available on fast charging LIBs, there is still a substantial need for constructive analysis on the impact of intrinsic variations on

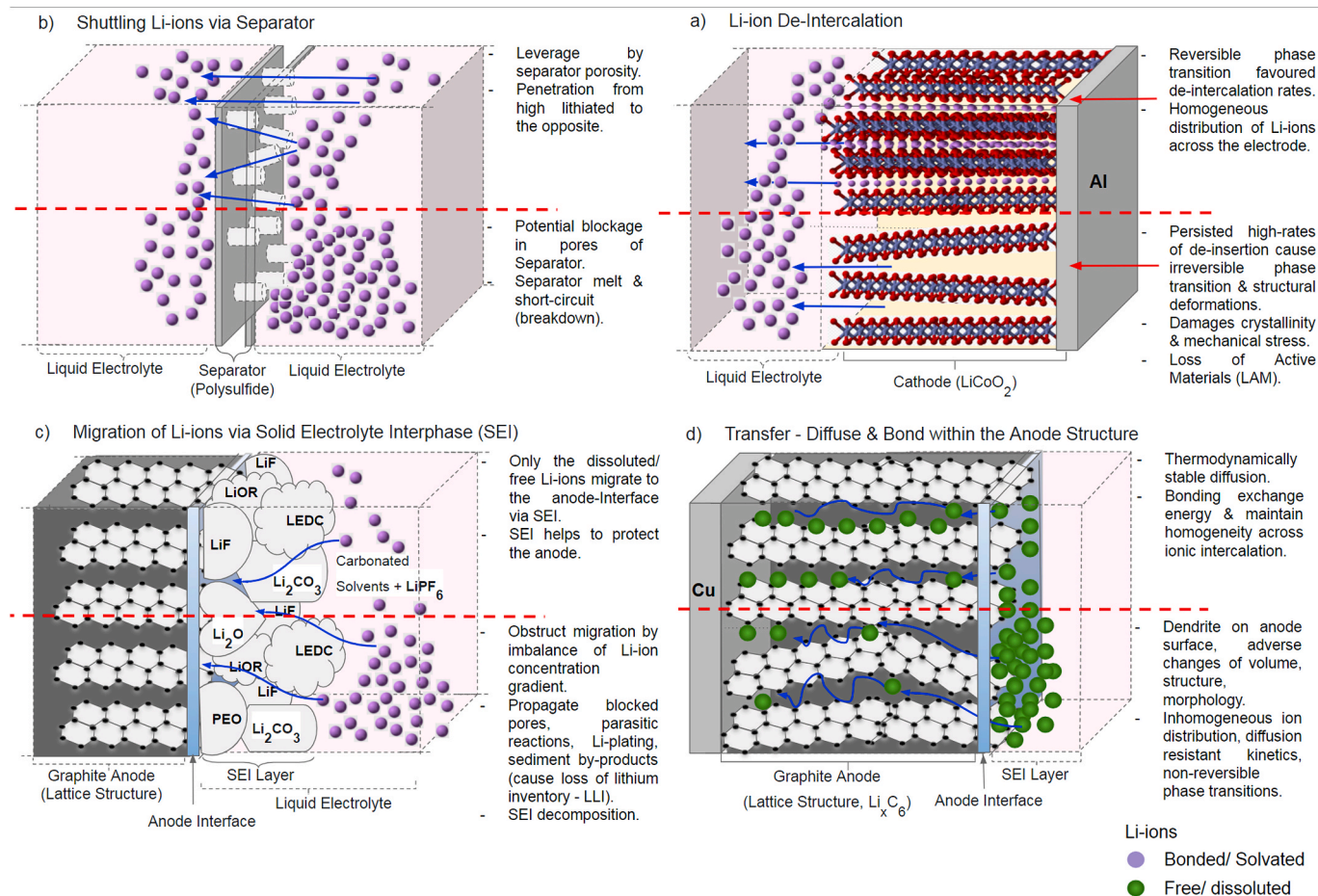
charging. This paper provides a review of the influences of intrinsic factors on the LIB charging process in terms of electrochemical, structural, and thermo-kinetic disciplines. Then, the main charging strategies are classified under a new analogy considering the use of battery data memory on their controls. Here, the charging is divided into two branches as memory-based and memory-less which provide impressive insights about the challenges and limitations in incorporating the intrinsic variations. Meanwhile, short-cache-based charging has been identified as capable of adapting the charging process over the variation demands. It helps in avoiding unnecessary stress on the cell and enables the use of closely real-time charge favorable variations, showing the path for a potential paradigm shift in fast charging. Accordingly, the short-cache controlled natural current-absorption-based charging pattern “iterated set voltage with zero current (ISVZC)” has been recognized as the next generation of fast charging. Finally, the conclusion discusses the potential advancements, limitations, and suitability of the method and future work.

## 2. LIB charging process from electrochemical and structural perspectives

The effective fast charging strategies should ensure less adverse impacts on their reversible properties to assure safe and stable charge-discharge operations over long cycle life. Understanding the deterministic factors of the LIB charging process is vital to cell design and the development of safe and effective fast charging methods. Hierarchically, these factors include the power capability of the external charger [75], the battery pack balancing [76], and the charge relative cell design factors [7,41,52,77]. The cell level atomic/particle transformations [78, 79] and their impacts on the internal structures are considered the most significant rate-deterministic factors [80,81] on charging. For this reason, a close analysis of the battery reactions over mass transport, charge transfer, formation/de-formation of byproducts, phase transitions, and thermal exchanges will help to understand the importance of cell-level sub-processes for healthy charging.

As it is shown in Fig. 1, the movement of  $\text{Li}^+$  from one electrode to the other has both the physicochemical and electrochemical challenges, as both the mass and charge particle transfer. During the charging process, the  $\text{Li}^+$  trajectory can be presented through a series of intermediary sub-processes [67–74]. Fig. 1 shows this trajectory in terms of 4 subprocesses, each of which is divided into Top and Bottom by a red horizontal line; Top - showing the ideal sub-processes, and Bottom - showing the potential adverse deviations from the ideal processes [82]. Here, as the widely used cathode material in LIB [67,68], the  $\text{LiCoO}_2$  has been used to explain the charging process. In order to clarify, the  $\text{Li}^+$  trajectory can be listed as: 1) de-intercalation (extraction/delithiation) from the cathode (positive electrode) [69,83], 2) shuttle through electrolyte passing the separator [70], 3) penetrating/migrating through the solid electrolyte interphases (SEI) [71] layer near the anode (negative electrode) [72], 4) intercalation (insertion/lithiation) into electrolyte/anode interface [73], and 5) diffusion [74] into the anode structure. Finally, 6) the intercalated Li-ions, once diffused through the bulk anode, get bonded within its layers [84], ideally at a ratio of one Li-ion per six carbons in the case of graphite anode. Meanwhile, the composition of Li-ion varies at the interfaces of electrode/electrolyte aggregating high ionic concentration gradients [85] near the negative electrode. Growing high ionic density near the anode interface and the low diffusion rates of anode materials oppose each other and obstruct the Li-ion diffusion process. These obstructions cause an imbalance of ionic composition near the anode/SEI surfaces forming  $\text{Li}^+$  congested/blocked SEI-sites and inhomogeneous distribution of bonded Li-ions in the electrode structure. This potentially results in numerous intermediate phase transitions [86,87] (yet reversible in general) collectively influenced by the energy exchange and kinetics of the processes [88].

Hence, an evaluation of electrochemical, structural, and thermo-



**Fig. 1.** The cell-level realization of LIB charging process (considering LCO): The  $\text{Li}^+$  trajectory via 4 sub-processes, each divided into Top and Bottom by a red horizontal line; Top - the ideal sub-processes, and Bottom - the potential adverse deviations from the ideal processes. a) Li-ion de-intercalation, b) Shuttling Li-ions via separator, c) Migration of Li-ions via solid electrolyte interphase (SEI), and d) Transfer - diffuse and bond within anode structure.

kinetic intrinsic perspectives is essential in understanding the charging process.

#### A. Electrochemical Intrinsic Perspectives

From an electrochemical standpoint, the charging process starts from oxidation at the cathode (positive-electrode) that purges/donate an electron at each extraction of a Li-ion particle de-intercalating  $\text{Li}^+$ s from the cathode structure [89] Fig. 1 a). It ends with reduction at the anode (negative electrode) where these  $\text{Li}^+$ s settle as Li particles after bonding with electrons, Fig. 1 d), transferred via the external load, which was donated earlier at the cathode and stabilized within the anode lattice structure. As a result, the voltage difference of the full cell increases due to the variation of electric potentials in electrodes; increment in positive and decrement in negative electrodes. The key for the reversibility of this process depends on the reversible reaction properties of chemical materials of the cell elements. Hence, the specific materials will differentiate the electrochemical states of a cell.

The generic chemical reaction of a positive electrode can be shown in the equation. (1) being the  $\text{MO}_2$  is a metal oxide such as  $\text{LiCoO}_2$  (LCO) or  $\text{LiFePO}_4$  (LFP).



and the negative electrode adheres the reaction in the equation. (2), especially for the graphite-anode:



As shown in Fig. 2, during charging, the electromotive force sparks the de-intercalation of  $\text{Li}^+$ s from the cathode. Then they shuttle through the electrolyte (against both the  $R_{ION}$ ; ionic resistance &  $R_S$ ; solution resistance), via the separator (through  $R_{PEN}$ ; penetration resistance), to pass through the SEI (passing the  $R_{SEI}$ ; SEI resistance) layer near the anode. After that, the charge-transfer resistance ( $R_{CT}$ ) plays a major role in transferring the charged ions to the surface of the anode. Finally, these  $\text{Li}^+$ s get intercalated by diffusing (against the Warburg Resistance:  $Z_w$ ) into the anode and bonding within the interspaces of this lattice structure. In this way, the anode structure is transformed from dilute stages to highly lithiated stages. The electrolyte is an ionic solvent to accelerate ion transfer [90], and the separator is a microporous interface that allows only the ions to travel through the porous holes. The SEI is a special layer formulated during the initial charging cycles of a LIB and serves as a protective layer allowing it to pass through only the  $\text{Li}^+$ s [71]. During the charging supported by an external power supply, the copper (Cu) current collector absorbs the electrons at the anode, while the aluminum (Al) current collector allows them to return to the external circuit. These metal collectors also function as safety elements to prevent the cell electrodes from potentially undesired external forces.

Briefly, at an open-circuit mode, a rechargeable LIB maintains an equilibrium between its intrinsic potentials and resistances inside the cell that forms a throughput voltage between the two electrodes. This is supported by the healthy operations and stability of the cell's internal components ranging from electrodes, electrolyte, separator, SEI, and their intrinsic properties. In the case of charging/discharging in closed



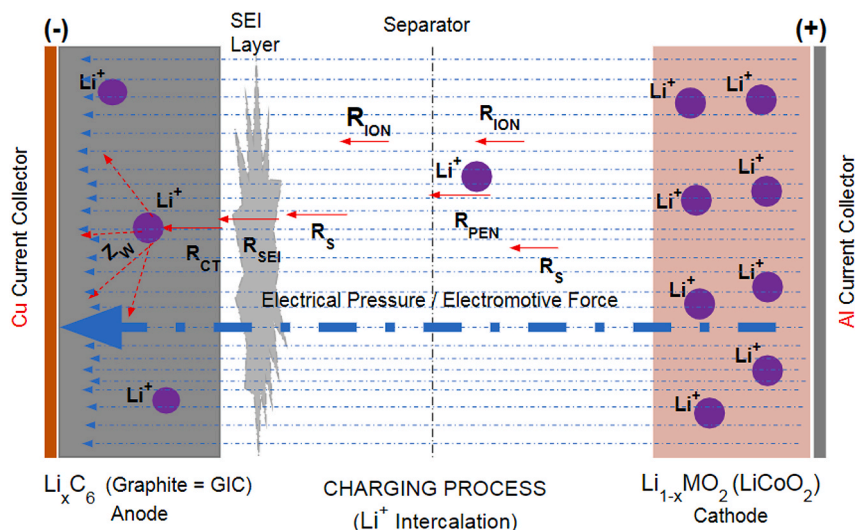


Fig. 2. Electrochemical realization of LIB charging process (in LCO cell): affected resistances against the shuttling of  $\text{Li}^+$  from cathode to, separator, SEI, anode/SEI interface, and diffusion at the anode.

circuits by the external electric supply/loads, it gets pushed through certain perturbations from equilibrium (potentially realized from polarization and non-equilibrium thermodynamics [91]) which sometimes results in rapid deterioration of the cells if not controlled properly. As the overall measures of these electrochemical variations, most charging methods use the voltage and current to regulate the charging process. Mainly, such measures related to the shuttle of  $\text{Li}^+$  rising voltage and the flow of electrons leading current. Although the electrochemical reactions, forces, and resistances drive the  $\text{Li}^+$  trajectory in this perspective, its inherent mass transfer gets cross-dependent influences from structural and thermo-kinetics.

### B. Structural Intrinsic Perspectives

Physicochemically, the mass transfer of  $\text{Li}^+$  triggered by the extraction of the Li-ion particles at the cathode and insertion of them at the anode impacts the stabilization of the electrode lattice structures during the charging process. The imbalances in this process lead to inhomogeneity of particle distribution across the electrodes. It is dependent on numerous structural properties such as the diffusion rate/length/path of electrodes [92], the surface area of the electrode-electrolyte interface, the morphology of edge sites (exfoliated edges), and tortuosity of anode pore structures. Limitations of such properties cause adverse effects on the charging performances. To provide a list; the poor diffusion results in low-rate capability [93], agglomeration of particles near anode interface leads to obstructing active sites and side-reactions that can potentially result in loss of active materials and mechanical stress. The structural properties combined with kinetics drive phase transitions and potentially transform the electrodes from reversible to non-reversible states leading to losing intercalation properties. These issues raise the need to measure and mitigate them before reaching their destructive levels during the charging process.

The techniques used to measure/examine such effects are complex and specific to certain laboratory conditions. Here, the advanced electrochemical characterization techniques such as X-ray diffraction (XDR), electrochemical impedance spectroscopy (EIS), potential step chronoamperometry (PSCA), galvanostatic intermittent titration technique (GITT), and potentiostatic intermittent titration technique (PITT) can be identified as helpful methods. Alternatively, complex mathematical/computational models which represent these intermediary processes, such as single-particle models [47,80], different orders of electric circuitry representations [94], polarization profiles [38,42,95], and

non-equilibrium thermodynamics [91] are used to predict the interior statuses and behavior in some advanced charging strategies.

As the main rate-limiting factor of charging [96], the diffusion coefficient has been examined in many research. In an evaluation for Li-ion diffusion coefficient in graphite, Yu et al. [81] employed an electrochemical impedance spectroscopy (EIS) model in terms of spherical particle operations versus the temperature and state of charge (SoC) of a cell. He compared EIS results against the PITT and Warburg impedance methods based on the following reaction (see Equation (3)) that represents the operations of particles at the graphite electrode surface.



Here,  $\text{S}_c$  denotes the Li-ion intercalation-possible vacant sites/inter-layer-spaces of graphite electrode/host. The  $\sigma$  and  $-\sigma$  respectively denotes the remaining charge of lithium and the accumulated charge of graphite sites/hosts after its intercalation. This EIS experiment reported that the diffusion coefficient varies from  $1.12 \times 10^{-10}$  to  $6.51 \times 10^{-11} \text{ cm}^2 \text{ s}^{-1}$  within the SoC range until 30% at  $25^\circ \text{C}$  and showed a slight increase at a high temperature of  $55^\circ \text{C}$ . Further, the Warburg impedance technique reported it as  $2.27 \times 10^{-13} \text{ cm}^2 \text{ s}^{-1}$  and the PITT technique showed an inversely proportional relationship with the SoC varying from  $1.03 \times 10^{-12}$  to  $9.30 \times 10^{-14} \text{ cm}^2 \text{ s}^{-1}$  over the rise of SoC of the cell. The experimental evidence shows the reliability of the diffusion coefficient is susceptible to the battery conditions and the measuring techniques. This makes it complex to measure them in a practical charger system and requires more research to explore possible ways to incorporate them within the charging controls.

### C. Thermo-Kinetic Intrinsic Perspectives

Collectively, the charge and mass transfer change the electrochemical and structural status of the cell. This intern leads to energy transitions influenced by chemical kinetics [97–99] and physical kinetics [100–103]. Additionally, the heat energy over the particle/mass movements causes thermal variations and becomes inter-dependent on the kinetics of the cell. Hence, the study should be focused on the combined perspective of thermo-kinetic on various effects such as the charge transfer kinetics, activation energy, phase transitions, and crystal structure of Li-occupations sites.

Jow et al. discussed the factors influencing the charge transfer kinetics of the LIB, from the  $\text{Li}^+$  de-solvation in a liquid electrolyte, its transfer towards anode/SEI interface, and until the acceptance at the anode-electrode structure during the charging process [98]. At a

microscale level, the charge-transfer process is triggered by the de-intercalation of  $\text{Li}^+$ s formed by a series of sub-processes. The sub-processes include the de-solvation of solvated  $\text{Li}^+$  in the electrolyte, transport of  $\text{Li}^+$  through the electrodes' SEI layers, and the acceptance of electrons (affinity) by the  $\text{Li}^+$  at the anode (stabilizing as Li-particles within the electrode structure). As such, the microscopic properties such as inhomogeneous concentration of ions/particles, resistive electrical fields, electrode morphology, and the nature of SEIs collectively impact the  $\text{Li}^+$  charge/discharge operations and their speed. In addition, the activation energy [99] contributes to leveraging the de-solvation as a dominating factor and is affected by the electric conductivity and ionic concentration of Li-ions throughout the process. Further, failing to achieve the timely specific/required activation energy leads to numerous interphase reactions (side-reactions/by-products) during the charge transfer. Consequently, it forms the agglomeration of by-products such as Li-plating, Li-dissolution, and Li-deposition increasing the loss of active materials (LAM) and the loss of lithium inventory (LLI) finally hindering the reversibility of the electrochemical reactions [104].

Hou et al. has provided an interesting review on phase transition in LIBs on  $\text{LiCoO}_2$  and  $\text{LiFeO}_4$  cathodes [100]. Here, two kinds of phase transitions, as first-order (in  $\text{LiFeO}_4$ ) and second-order (in  $\text{LiCoO}_2$ ) can usually be observed within the  $\text{Li}^+$  intercalation/deintercalation processes. In addition, the phase transition depends on the fulfillment of many factors. These include possible intermediary reactions (occurrences or nonoccurrences), their speed, their thermodynamics, the influences/limitations of kinetics, and their structural/material qualities. Based on the Li-ion diffusion trajectory and activation energy that sway their transport [101], the crystal structure of Li-insertion compounds makes further differentiation of the battery intrinsic. They are termed into three classes as olivine (1-D:  $\text{LiFePO}_4$ ), layered (2-D:  $\text{LiCoO}_2$ ) and spinel (3-D:  $\text{LiMn}_2\text{O}_4$ ) holding inherently different structural properties and kinetics which makes it complicated to realize/regulate the phase transitions. According to Ceder et al. [102], the kinetic factors of Li-ion strongly influence the possibility of the occurrence of phase transition in  $\text{Li}_x\text{CoO}_2$ . Such factors can be identified as the  $\text{Li}^+$  diffusion coefficient, its concentration (representative as  $x$  in  $\text{Li}_x\text{CoO}_2$ ), and the electric conductivity within the solution space. Defined by the structure of Li occupation sites and their oxygen stacking manner, different structural types can be identified as O1 (hexagonal form), O3, and H1-3 during their phase transitions in a broader Li concentration range ( $x \leq 0.4$ ). Amatucci et al. [103] showed that the O3 structure is unstable as it occurs when the electrode-sites are completely de-intercalated. Nonetheless, the reversibility properties of the constituents of Li-ion solution space are determined not just by the unstable phase transitions, but as a series of re-occurrences of them within a certain period. However, while the reversible phase transitions [142] are inevitable, the irreversible phase transitions will deteriorate the battery performance.

#### D. Multi-disciplinary impacts

The extent of the multi-disciplinary impacts on the charge/ion transfer makes the charging processes collectively interdependent with a number of phenomena. It makes the charge-motivated transitions (high rates) counter-productive by they are being the cause for accelerating the charge demotivated operations (Li-plating, etc.). For example, the rapid increase of a large volume of Li-ion intercalation, caused by a high rate of charging (in fast charging), hinders the proper diffusion of Li-ions within the electrode structures and agglomerate particles (by-products) near the electrolyte/electrode interfaces. It increases the chance for side-reactions, which finally risks the battery's performance and health.

In terms of mass transfer, the impact of the rate of particle diffusion within the anode surface, then in its bulk, and the smooth phase transition/migration at reversible boundaries (from the many intermediate processes) are considered the most crucial parts of this process. They determine the speed and the reversibility of reactions in the battery. The

electrochemical reaction of the charging process depends on a series of continuous intermediary sub-processes that are influenced by factors such as charge and mass transfer kinetics of particles, ionic conductivity and concentrations, and transitions in electrode-electrolyte interfaces. Accordingly, the overall speed of the charging process is highly subjective to the runtime internal cell state and, also, it is affected by all the subprocesses and their resistances. It causes a high possibility for incomplete intermediary reactions or even disappearances of some of them. The kinetic limitations and thermodynamics effects influence the occurrences, non-occurrences, and the speed of sub-processes. All these collectively drive the phase transitions and sometimes to irreversible levels. Phase transitions influence the battery characteristics such as power density, energy retention, and volumetric changes.

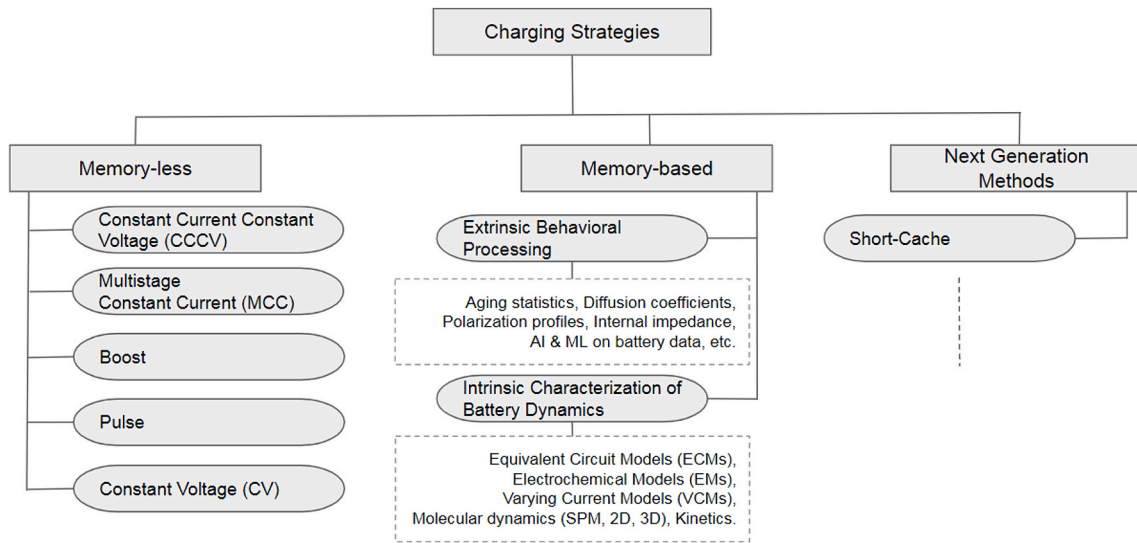
Inevitably, these complicate the control of LIB charging indicating the necessity of charging regulation to effectively incorporate the effects/measures of inter-dependent operations at electrochemical and physicochemical levels. Moreover, the rapid volatility of the internal cell conditions, the requirements of advanced technological methods to measure them, and the limitations in their precisions have made their presence far from practical commercial fast chargers.

### 3. Fast-charging methods: memory perspectives

Reviewing the existing charging strategies helps to gain a profound understanding of the challenges and limitations of the current charging methods in both research and practical charger implementations. Here, analyzing from the aspect of how far the existing charging strategies tend to address the aforementioned multi-disciplinary factors, a new logical and intuitive classification for charging methods can be introduced. The key for this classification considered whether the charging strategy incorporates any battery-specific memory of data in processing and regulating the charging process. Accordingly, it can classify the world of charging methods into two interesting branches as 1) Memory-less (with predefined/fixed parameters) [18,19,53,105,106], and 2) Memory-based (with some history of data/information of battery behavior) [16,17,45–47] strategies, Fig. 3. Evaluation of several relevant charging methods under each of these categories helps to understand their abilities, limitations, and struggle towards achieving the optimal charging targets. Consolidating the review from both the categories will be an eyeopener to the fact that none of these approaches addresses the demanded adaptation to the battery intrinsic variation during the charging process in real-time.

Memory-less charging strategies tend to control the charging process with a predefined and fixed set of parameters such as constant current (CC), constant voltage (CV), constant power (CP), and constant temperature (CT), irrespective of the changing battery dynamics. Mainly, the direct readings of current, voltage, and temperature of the cell have been used in controlling these charging methods. Further, the span of time and SoC have been used as additional parameters to support the controls of the charging process. These parameters set fixed thresholds individually or collectively to control the overall charging process of starting, continuing, and ending. In a way, it helps show positive acceptance in commercial chargers due to their simplicity in implementation.

Their charging control parameters are predetermined and fixed up to specific patterns, their charging decisions do not incorporate/adhere to the internal behavioral changes or the battery state of health (SoH) conditions. Consequently, they stress the battery causing an accumulation of adverse impacts/side reactions within the process and across its cyclic charging iterations. In most cases, due to the uncertainty caused by such potential aggregations of destructive effects, the memory-less methods always tend to enforce average rates and the lowest possible thresholds when fixing their parameters/controls. They mostly fail with aging and tend to accelerate cell degradation when used with high rates of current/voltage. Also, using the batteries only up to the average levels will hinder the opportunity of the full use of potential tendency in



**Fig. 3.** High-level classification of charging strategies for LIBs, based on the memory perspective. It considers whether the charging strategy employs a memory of data on battery analytics and/or prognostics and short-cache controls on the charging process.

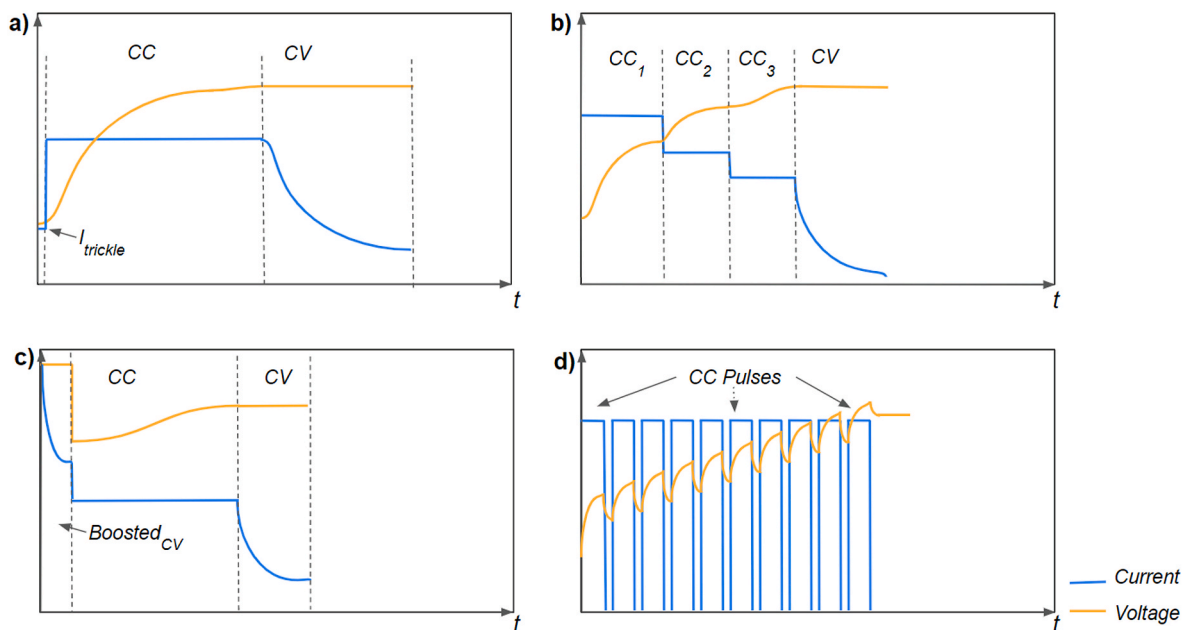
**A. Memory Less Charging Strategies**

parameters (e.g.: high voltage, high current rates) [87,107,108]. Particularly, it loses the chance of getting charged with higher current rates when the intrinsic state of cells holds charge-friendly internal kinetics. Further, it risks the battery enforcing fixed parameters when the internal kinetics demands lower thresholds.

To holistically understand this branch of charging, the most fundamental strategy of constant-current constant-voltage (CCCV) should be well understood. Then it will be helpful to explore other charging strategies that are holding the roots from the CCCV.

The CCCV, Fig. 4. a), is a two-stage charging method. It is considered constant current (CC) and constant voltage (CV). CCCV is also known as the conventional charging method for LIBs and could find in a wide range of LIB solutions. Firstly, it charges the battery at a constant current,  $I_{CC}$ , until it reaches a predefined cut-off voltage,  $V_{CV}$ , linearly incrementing the SoC. Secondly, it switches to a constant voltage,  $V_{CV}$ ,

during which the current decays exponentially as the battery gains charged. The values of both the  $I_{CC}$  and  $V_{CV}$  are generally defined as a range by the battery manufacturers and are typically kept low to avoid potential safety risks. Here, the end of the process is determined by satisfying one of two conditions, being first, if the drawn current drops below a predetermined value,  $I_{cutoff}$ , which is generally about 5%–10% of  $I_{CC}$ ; and the second, if the duration of CV based charging exceeds a predefined duration (usually 2hrs). These configurations make it easy to implement, simple and straightforward. Additionally, when the starting battery voltage is very low (below  $V_{trickle}$  usually 2.8 V/cell), a special trickle-charge mode is employed before the CC stage. During the trickle-charge scenario, a very low constant current,  $I_{trickle}$ , typically less than 10% of the rated-capacity value of the battery, is applied until the trickle-voltage ( $V_{trickle}$ ) is reached. Generally, the CCCV is known to be a slow charging method, especially with prolonged CV stages when



**Fig. 4.** The schematic representation of several Memory-less charging strategies: a) Constant-current constant-voltage (CCCV) charging, b) Multistage constant current (MCC) charging, c) Boost charging and d) Pulse charging.

attempting to fast charge with higher current rates. However, the continuous use of long-duration CV stages, contribute to adverse effects and result in side-reactions, the high rates of CC are detrimental to the cell, and all these will degrade the battery performance.

The multistage constant current (MCC) method can be considered an enhanced version of the CCCV strategy. This method combines some CC stages (in decrementing magnitudes) and, in the end, one CV stage, as seen in Fig. 4 b). In a way, these changing CCs to lower levels interrupt the phase/rate of continuous aggregation of heat and support reducing; the possible thermal instability,  $\text{Li}^+$  plating, and mechanical stress that would be obvious in a longer-continuous CC stage like in CCCV. Ikeya et al. has introduced a five-step charging method partitioning its time-space into five consecutive sections [105] where each of it charges with a different current and gradually lowers the current magnitude with incrementing steps. Selecting higher current for the initial steps in such methods is sometimes supported by the findings of less internal resistances in lower SoC ranges [48] of LIB. With regard to the evolution of internal resistance as a function of SoC, it depends on cell type and the charging current. In general, the resistance is lower in the middle SoC stages, but it increases in higher SoC stages. Deciding the charging current values and the width of the time frame at each step of MCC is predetermined, but sometimes optimized using different algorithms [109,110] such as particle swarm optimization [109]. However, the cell stress made by the fixed parameters and high-current over cyclic use may lead to premature aging and hinder the charging performance.

Boost charging, Fig. 4. c), is another fast charging strategy modeled with a short initial boost-of-current which seems to take advantage of the less internal resistances at the lower SoCs, and hand over the rest of charging to the standard CCCV method. Notten et al. [19] has suggested a new, ultra-fast charging algorithm as an improved pattern of CCCV called boost-charging, which mainly consists of three steps, starting with an initial-CV which draws a high initial current, followed by 2-steps CCCV charging. This initial CV stage is limited to a short period, for about 5 min, where the voltage is set to a higher value (even at 4.2 V or 4.3 V) straight away, enabling the battery to draw very high currents such as about 4–5 C-rate (The nominal capacity of a battery denoted as 1C, whereas a fully charged battery discharge at 1C-rate takes 1 h to fully discharge). Afterward, a standard CCCV charging scheme is employed with a low CC (about 1C) and a general CV until the battery gets fully charged. Its experimental results on both the prismatic (LP423048) and cylindrical (US18500) LIB cells [19] accounting for around 300 cycles showed no significant deterioration in the charging performances. Nevertheless, as the initial boost-charging may draw unexpectedly high currents, the battery could experience sudden hikes in ionic concentration and result in high polarization and heat generations.

Pulse charging, Fig. 4. d), applies a train of CC pulses separated by periodic rest-steps with either zero current or discharge current-pulses. The purpose of the rest-steps is to influence the cells to self-control the ionic concentration. It targets to maintain an even distribution of ionic composition to reduce the mechanical stress due to possible uneven distribution during the charging process. Hence, it can also slow down the concentration polarization and secure the cell from rapid deterioration. In 2001 [106], an early-stage pulse charging protocol reported that it can achieve around 1 h fast charging with 1C pulses (with quick zero current rests), compared to around 3.5 h time requirement when charged with conventional CCCV (CC rates at 1C). More research can be found in optimizing the pulse charging parameters using various approaches such as electrochemical-systems-based models [111–113], SoC governed methods [48], Taguchi orthogonal arrays method [114], and online polarization profile-based models [42]. However, their distinctive pulse load (also high rates) may counter impact with increased heat generation in cells leading to uneven heat in battery packs, and these must be extensively studied [115].

Constant Voltage (CV) based charging is not popular as a complete charging method. It is well known as a part/inner stage of charging

methods and recognized as a relaxation step. Normally, the CV is necessary for charging controls, as it shouldn't continue the CC charging when the cell voltage reaches the maximum value. While the voltage is kept constant, it has a distinct nature of decaying current that allows the cell to gain only a particular amount of charge-current that is related and peculiar to the internal statuses of the cell at that very moment. Hence, the CV-based charging profile is used for numerous estimations in battery interior statuses during a charging process. Wang et al. suggested a CV charging profile-based battery capacity estimation technique [116]. Akarma et al. have investigated the CV charge phase of a CCCV based charging as an indicator of the SoH of a battery for electric vehicles [117]. Nonetheless, due to its potential rise of electrolyte oxidation [118,119], it has been found as highly detrimental to the battery if the CV charging is continuously used for a long-time. Contrary to the rapid deterioration of battery, when used for a prolonged time, interestingly, its use in short-charging durations tends to show remarkably opposite behavior in favor of relaxing the deterioration, due to the prominence of naturally decreasing current based on its battery intrinsic. It open-up new avenues to explore possible utilization of charge-friendly intrinsic status.

## B. Memory Based Charging Strategies

Memory-based charging strategies have arisen as advancements of the memory-less branch injecting memory-specific controls (decisions made using memory about battery analytics and prognostics) within the charging process. It has expanded into a wide range of areas depending on the sources of data that substantiate the memory. This memory is formed by the use of some history of data, battery modeling techniques/laws, or a combination of all. These memory sources can mainly be classified under two types of battery analysis: 1) Extrinsic behavioral processing and 2) Intrinsic characterization of battery dynamics. Extrinsic behavioral processing can include a wide range of analyses such as aging (charge/discharge cycles) behavior/statistics, pre-recorded diffusion coefficients, thermodynamic analysis (entropy/enthalpy), polarization profiles, and patterns of internal impedance, history of battery behavior, and AI & ML [120] on battery data. The Intrinsic characterization of battery dynamics (experimental/empirical/predicted/heuristics) can include the processing of mathematical/statistical/electrochemical circuitry models at the cell level, representation of physical/particle/molecular level dynamics (single-particle model - SPM, spatial molecular view - 2D/3D [23,121]), kinetics/thermal/electrical model [122] designs and prognostics. All these approaches attempt to produce effective models that can closely represent the internal dynamics of the cell, and subsequently in controlling the charging process.

Undoubtedly, these methods need high computational and dedicated experimental setups [123]. Also, the limitations in power electronics of commercial charger applications [124,125] challenge their implementation making this branch of charging methods far from practicality. Extrinsic behavioral processing depends on indirect measures to perform quantitative/qualitative analysis on intrinsic effects (e.g.: diffusion, polarization). Such measures require long-term experiments and become inaccurate due to the very present status of the cell and deviations in measures. The intrinsic characterization techniques struggle in achieving due accuracy and get compromised due to the rapid volatility during the charging of the battery. They are highly dependent on numerous implications such as the technique of experiment, micro/mesoscale variations of battery constituents, and unexpected aggregation of side reactions. To substantiate these findings, several memory-based charging strategies should be discussed.

Extrinsic behavior-based analysis (type 1) driven memory provides important insights to substantiate the charge regulations. A list of such insights includes SoC estimation, identifying favorable SoC regions for fast charging, the tendency of polarization/ohmic resistance during charging, and aging effects over ionic concentrations. At another level,



the contextual and statistical representation of the internal electrochemical properties such as polarization, internal impedance, ohmic resistance, warbung resistance,  $\text{Li}^+$  concentration gradient, and SoC estimation can be seen as the main factors which attempt to minimize the battery degradation effects. Here, some mathematical and statistical analysis methods based on both the real-time and offline data can also be seen in deriving the approximations about the internal electrochemical statuses. Some neural networks and fuzzy logic [46] based approaches can be seen controlling the charging patterns along with certain unknown or directly immeasurable (e.g.: aging effect on ionic concentration or diffusion) properties of the battery. In a simpler electrochemical perspective, the cell voltage consists of two contributive components as interface and volumic [126]. The interface component corresponds to the voltage difference between electrode/electrolyte interfaces at the open circuit stage. The volumic component, also called the ohmic drop, is induced by the charge transport across the volumetric elements of the cell (current collectors, electrolytes, and connections). As an electrochemical status indicator by external measures on the internal resistance of the cells, Fleury et al. [40] employed an ohmic-drop compensation principle-based fast charging method. With such indicator-based charge favorable insight of using an extended constant current period at higher limit voltages, it managed to use high charging rates to fast charge the cell. In another work [127], the open-circuit voltage and temperature have been used with SoC to evaluate the entropy change of the cell reactions during charging. It identified a fluctuation of entropy on various SoC regions (range between 70% and 90%) led by the cathode phase transition which in turn influenced by varying temperatures and excess lithium in the cathode. Such indirect measures can use to regulate the charging to avoid non-reversible phase transitions in the cell electrodes.

In terms of representations of intrinsic (type 2) dynamics of the battery, a wide range of ongoing research can be found in different areas such as electrochemistry, mesoscale physics, molecular/atomistic models, particles, spatial, kinetics, and thermodynamics. As a striving area, the single-particle model (SPM) [80,128] considers that the electrodes are collections of spherical particles (Fig. 5) carrying the same physical properties with uniform distributions and approximate them by a single spherical particle in both electrodes. Further, the 2D and 3D [129] representations of the spatial stability at molecular levels are advancing with interesting new research in LIB space since recently. Such representations are underpinned by the heuristics of cell particles

and scientific principles. Several interesting principles about spatial stability can be considered as the Fick's law [130] on balancing the mass of li-ions in intercalated particles, and the Butler-Volmer kinetics [75, 121] in explaining the rate of the intercalation/deintercalation of  $\text{Li}^+$  of the electrochemical reaction at the electrode/electrolyte interface. However, it is still challenging for LIB technology to handle collective inter-dependencies of kinetics, forces, and transformations at molecular and particles levels. They influence the effects such as charge transfers, volumic/morphological changes, gas propagation, energy dissipation, and heat transfer, which will eventually define the specific health of the reversible reactions of the cells.

Patnaik et al. employed cell temperature degradation metrics proportional to the controller parameters in implementing a constant-temperature, constant-voltage (CT-CV) charging method [131]. This controller modulates the charging current whilst keeping the cell temperature at a constant set-point. In a comparison with the standard CCCV method which draws a similar rise in net-temperature increment as in CTCV, the latter shows a 20% faster charging performance. Here, the following discrete domain model controller was utilized.

$$E_r(n) = T_{set}(n) - T_{fb}(n), \quad (4)$$

$$I_p(n) = K_p E_r(n), \quad (5)$$

$$I_i(n) = I_i(n-1) + K_i E_r(n), \quad (6)$$

$$I_d(n) = K_d [E_r(n) - E_r(n-1)], \quad (7)$$

$$I_{pid}(n) = I_p(n) + I_i(n) + I_d(n) \quad (8)$$

Here, the  $E_r(n)$  denotes the controller error,  $T_{set}$  and  $T_{fb}$  are the cell temperature values representing the set-point and feedback values. The  $K_p$ ,  $K_i$ , and  $K_d$  represent the proportional, integral, and derivative terms of the controller. The  $I_{ff}$  denotes the current feed-forward term and given as below:

$$I_{ff} = \begin{cases} 2C, & 0 \leq t < tpk \\ \left(1 + e^{-\frac{(t - tpk)}{\tau}}\right) C, & tpk \leq t < tcv \\ X, & t \geq tcv \end{cases} \quad (9)$$

Memory-based charging methods are likely to employ numerous battery modeling techniques, mathematical and statistical models to

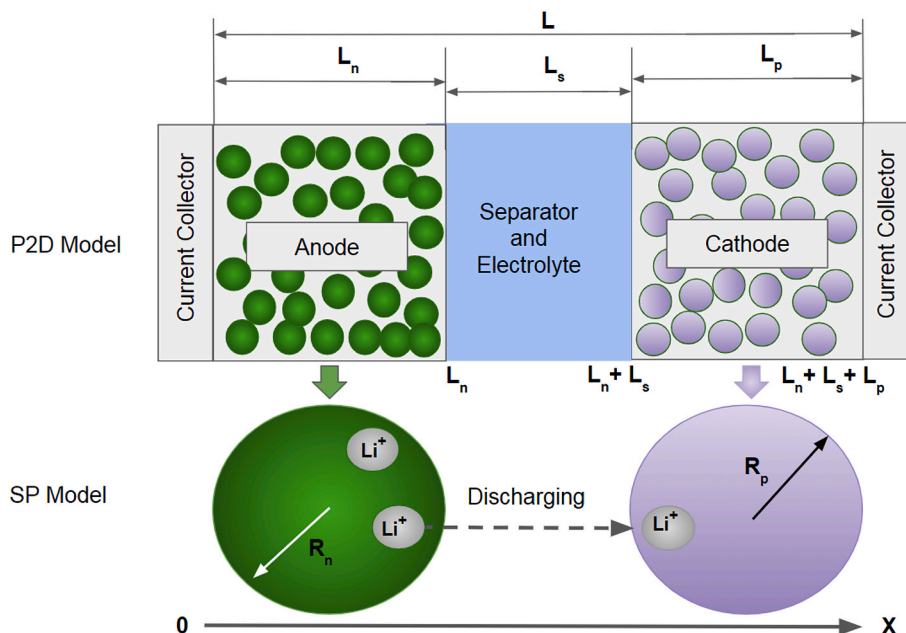


Fig. 5. Three-zone schematic representation of li-ion cell; Single Particle (SP) Model and Pseudo-2D (P2D) electrochemical model [80].



regulate the charging process. Here, the equivalent circuit models (ECMs) [132] and electrochemical models (EMs) [80,115] are widely discussed candidates in the LIB literature. The ECMs classified mainly as 1st order [133] circuits and 2nd order [134] circuits while EMs can vary within a wider scope, depending on what representative models are injected into them, such as single particles behavior [80], diffusion resistances [81,135], thermal particles [95], electrothermal [136] and electrolyte concentrations [137].

The temperature increases beyond the allowable limits can accelerate battery degradation and result in safety issues, especially as they affect the stability of the cathode lattice structures. As a solution, Caiping et al. [95] proposed a thermal behavior model enhanced with genetic algorithms in finding the optimal trajectories of charging current, addressing the relationship of polarization voltage variations vs charging current, to control the temperature within affordable limits. Here, the entire battery has been treated as a particle cell for the thermal model without considering its inter-particle dependencies. It has experimented with the battery temperature at time  $k$ , as below, with the sampling intervals of 1s:

$$T_{cell, k} = T_{cell, k-1} + \frac{\left[ I^2 R_{\Omega} + I U_p + T_{cell, k-1} \left( I \frac{\partial E}{\partial T_{cell}} \right) - Ah (T_{cell, k-1} - T_{amb}) \right]}{m C_{cell}} \quad (10)$$

where  $U_p$  is the polarization voltage; expressed as  $U_p = kI + b$ , where  $m$  is the weight of the battery. This experiment was conducted in an adiabatic box to prevent the heat exchange/dissipation, and it exhibited reduced charging time within a reasonable temperature rise. Nevertheless, it was found that the weighting component affects battery charging performance and that estimation error is large at resistance calculation in higher charge currents.

### C.Short-Cache based Charging Strategies

Notedly, a new charging branch is growing with adaptive charging that addresses the intrinsic variations in real-time or closely real-time. Although it inherits control properties from both the memory-less and memory-based strategies, it employs peculiar regulations in embracing the changing battery dynamics. They can be considered short-cache charging methods having roots from both the above. Being a short-cache, it intends to avoid outdated-data-driven errors that deviate insights from the very presence of cell behavior. It helps to understand the real-time/closely real-time variations of the cell intrinsic and adjust the charging process. It makes the charging control free from pre-set memory/model-centric charge enforcements that are not coined perfectly with the present cell dynamics. Also, it avoids stressing the battery for long periods enforcing fixed parameters while the intrinsic is highly volatile.

Bandara et al. provided an adaptive charging protocol based on non-linear voltammetry (NLV) fast charging mechanism, decomposing the charging-time space into a series of short durations and applying a non-linear relationship to decide the next charging voltage after each charging step [138]. It has also mixed with frequent relaxation of zero-current steps in between each charging step. This work roots from an initial heuristic defined by Rachid et al. [139] which claims a relationship of the changing-rate-of-current ( $dI/dt$ ) versus the changing rate-of-voltage ( $dv/dt$ ) is following a K-constant throughout the charging process of a LIB. However, this work [138] experimentally verified that it is not possible to follow a constant throughout the entire charging process, and such relationships may only be viable within certain SoC regions. Accordingly, it uses a series of different K-values, initially based on an empirically-selected-set and later improved with adaptation cycles in the charging process. At its best, this NLV based strategy reported a full charge of a LIR13450 cylindrical cell [140] within around 23 min, Fig. 6. It has also reported a charge efficiency of 99.7% compared to its nominal capacity. With promising results of more than 1000 cycles in fully charging, a pouch-cell with a nominal capacity of 2100 mAh, in around 35 min, it claims that both the SoC and SoH of a battery will likely affect the magnitude of the current and the pattern of its depletion during the short NLV based charging steps. As a short-cache charging, it doesn't enforce the battery to charge with any fixed current. Instead of that, it aggregates the charge from naturally drawn current flows within non-linearly changing voltage steps. It relates to the charge-favor-ability of the intrinsic status of the cell given by the control at short voltage steps.

### D. Charging Strategies over changing cell-intrinsic

The above section reviewed a number of charging strategies under the memory perspectives. In a nutshell, none of the memory-less and memory-based strategies meet the overall expectations of optimal charging performances in terms of achieving both the fast charging time and guaranteeing the safe and healthy usage of the battery. Although the memory-less charging methods look more commercial friendlier, their fixed-parameter controls fail to incorporate the intrinsic changes of the cell (structurally, electrochemically, and thermodynamically). The memory-based methods tend to control their charging strategies based on some cell behavioral data and intrinsic representations. However, their lack of effective real-time monitoring of the cell-intrinsic still causes deterioration of battery health, degradation of performances, and eventually poses safety risks.

In both memory-less and memory-based strategies, the cell inevitably gets stressed during their charging processes as they are failing to self-adjust based on the changing demands in the cell-intrinsic [141, 142]. Over time, such stress aggregations may lead to adverse impacts such as hindering the reversibility of internal reactions. As an example,

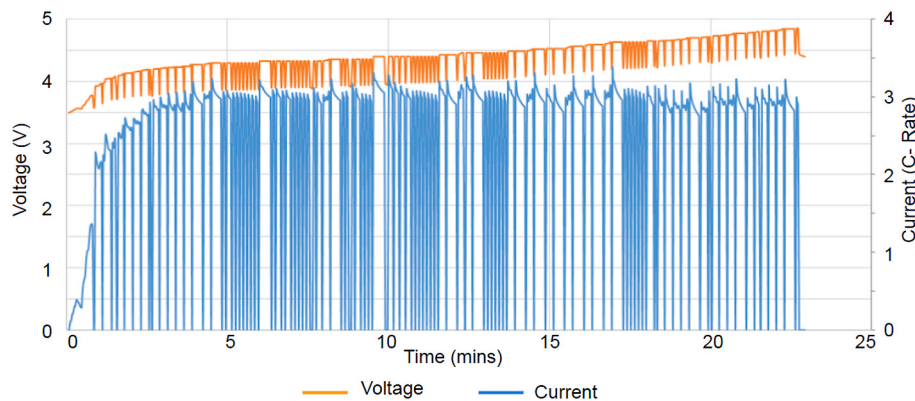


Fig. 6. The voltage and current profiles vs time of NLV based fast charging. Here, a cylindrical cell, LIR13450 with a nominal capacity of 650 mAh, has fully charged in 22 min using the NLV based method.

rapid agglomeration of Li-ions near the anode/electrolyte interface will interrupt proper ion diffusion, and in the long run, it led to issues such as Li-deposition and Li-plating. Further, the use of high current rates can undesirably produce a large-scale extraction of Li-ions from the cathode structure which in turn affect the crystallinity of the electrode. This leads to issues such as instability in electrodes, micro-crack in electrode structures, and depletion/non-reversibility of active materials. The use of high voltage and/or current continuously for prolonged periods draws severe vulnerability in thermal instability. The excessive levels of these can get aggravated towards thermal runaways causing explosions and/or fires. After all, these multidisciplinary effects are collectively influenced by the real SoC and SoH states, which precisely indicates the present intrinsic status of the cell. To gain optimal charging performances, it should remain the reversibility of the reactions without exceeding the thresholds (e.g.: minimum ionic concentration to maintain the crystallinity, reaction speeds & required kinetics to maintain reversible phase transition &, etc.) to adverse states, and hence the charging should be able to adjust according to the present statuses of the cell.

In a structural and electrochemical perspective of a charging process, the particle diffusion within the cell is considered the rate determinative sub-process and is affected by both the structural and electrochemical properties of its components. Most prominently, the surface area of particle penetrating interfaces, the thickness of the electrode, the tortuosity and exfoliated edges of electrode lattice, and the conductivity of solution space are all affects the rate of diffusion. Importantly, the relaxation [143] during the charging process seems to allow room for stabilizing the aggregated ion-particles into the bulk-electrode materials favoring smooth diffusion. On the other hand, as a key influential phenomenon that affects the reversibility of cell reactions, the phase transition depends on inter-dependent factors related to the cell dynamics. Particularly, these factors should meet several conditions in driving phase transitions. Such conditions can be listed as thermodynamics, kinetics, and kinetical limitations of active materials and high current/voltage rates. Essentially, it requires the presence of the collective combination of criteria occurring at a particular speed/rate over continuous reaction processes [144]. Notedly, intermittent relaxation could benefit in maintaining the phase transitions at reversible levels [144,145]. However, charging with high current and/or voltage continuously for longer periods drive into irreversible phase transitions [146] even in damaging the cell. In addition, the rapidly changing intrinsic state of the cell makes it harder to control/avoid such crucial thresholds to prevent forming adverse effects. Most importantly, as the cell-intrinsic status is rapidly changing during the charging process, it shouldn't be anticipated and not viable to be controlled by pre-defined/fixed/constant charging patterns. All these suggest that an ideal fast charging strategy for LIB should afford the demanding adaptability of the highly volatile properties of the cell-intrinsic states during the charging process.

Holistically, the optimal fast charging processes should instill a significantly high intake of electrons (current) and promote high amounts of faster  $\text{Li}^+$  intercalation (anode)/deintercalation (cathode), while on the other hand, the battery intrinsic dynamics tend to limit them from moving beyond respective thresholds of multi-factors (inter-dependent intermediary transformations) to retain the reversibility of cell reactions. However, the inability/complexity to accurately monitor the thresholds in real-time made many charging strategies to enforce limits to utilize parameters within average/low ranges. Hence, they settle at sub-optimal fast charging outcomes indirectly wasting the use of optimal capabilities of LIBs. In short, an ideal fast charging method should attempt to reach the best possible levels of these thresholds, to achieve maximum capabilities, and not to exceed them that decides the possibility of risks to the reversibility of cell reactions. Here, the relaxation coupled charging strategies can be considered a powerful technique to cope with such thresholds/turning points, and the short-cache-based charging methods tend to be beneficial in this aspect.

#### 4. Towards the future: natural current-absorption-based fast-charging methods

The above review shows that the existing charging approaches are considerably far from adjusting their charging methods towards the real-time/rapid changes that occur in the cell-intrinsic at material/particle levels. Parallel, considering the potential influences of electrochemical, physicochemical, and thermodynamic reactions/sub-processes/transfers on the charging outcomes during the charging process, it is vital to incorporate the internal variations timely, to adjust the charging control, and to expect optimal performances. Nonetheless, it is a complex process hindered by the technical limitations in measuring intrinsic phenomena in a practical charger context.

However, the adaptive charging protocols (short-cache-based) provide prominent evidence of changing their charging controls based on some response signals from the battery within the charging process. Importantly, the adaptive protocols with non-linear voltammetry tend to change the charging controls based on an analogy that helps to indirectly identify the charge-favorable and lazy-charge situations in real-time. Accordingly, a potential paradigm shift for the next generation of fast-charging LIBs can be identified in the new area of the natural current-absorption-based charging strategy. It expects the adaptability of the charging methods to respond to the intrinsic shuffles in the battery and achieve effective fast charging while providing practical feasibility for charger implementations. This section provides high-level recommendations and processes for next-generation fast charging.

##### A. Review Insights

Notedly, first, as per the literature of LIB, the constant voltage (CV) step can be considered as a way of indirectly keeping the battery at relaxation [145,147], without externally enforcing the intake of the current, and allowing the drawn/absorbed current to be decayed based on its internal states that also impacted by the SoC and SoH of the battery. Secondly, it cannot be justified to use the fixed parameters, such as CCCV, to regulate the charging of an ecosystem that has proven rapid volatility due to its multi-disciplinary engineering concerns which get further intensified by the fast charging attempts. Thirdly, compared to a few decades back, now the LIB is used in more computationally powerful electric devices [9–11] with miniature electronics, which has the potential to enhance its charging process with more intelligence to a new level of effective optimization. Further, the recent rapid proliferation of LIBs in human-affected service industries such as transportation, communication, and more recently in renewable energy integration into the electrical systems [148,149], and their adverse incidents due to charging issues have intensified the demand for more reliable charging strategies than earlier. However, it is still lacking a proper charging process that can actively respond to the cell's internal changes in real-time while ensuring safety, long cycle life, and short charging times.

Interestingly, it is an important fact that when the cell is set to a particular charging voltage, it will spontaneously absorb/flow a current, indirectly related to its internal characteristics at that time. In another way, a voltage step (especially a short step with enough relaxation) allows the cell to absorb a specific amount of current, which seems to be influenced by its internal states [40,48] and is not imposed externally. The magnitude of this flown current and its decay pattern over time has a strong correlation with its SoC, and potentially with the SoH of the battery [45,46]. As such, this very specific nature of natural current absorption within short voltage steps, if coupled with small incremental steps and zero-current relaxation, can be used to enable the natural current absorption for charging.

Consolidating all the findings and insights from this review, and the potential benefits of generally unused short-voltage-steps-based charging we can recommend a new control strategy for LIB chargers for the next generation of fast charging solutions. It introduces an iterated step-based approach as "iterated set-voltage with zero-current

(ISVZC)".

#### B. Natural Current-Absorption based Fast Charging (Iterated Set Voltage with Zero Current: ISVZC)

The identified next-generation fast charging method, shown in Fig. 7, mainly depends on the self-regulation of a series of nonlinearly incrementing set-voltage (SV) based short charge-steps, followed by dynamically controlled zero-current (ZC) based relaxation steps. It enables the natural current absorption-based fast charging in employing the iteration of SVZC steps at a finer granularity in time based on the real-time analysis of the flowed/absorbed current within each SV-based short charge-steps. Importantly, using the iteration at the correct finer granularity of time helps to avoid the possible stressing on the battery with potentially longer set-voltage steps. It ends the charging process based on the gained SoC and the analysis of the current response. The current response at the tail part of the charging process tends to signify the dislike/oppose to further charging.

The fundamental determinative factor of this novel ISVZC design is the consideration that the variation of the absorbed current within each SV-based charge-step indirectly implies how far the battery favors to charge at that step. Due to the nature of rapid volatility of the cell's internal state, the suitability and the pace to charge the battery at a particular set-voltage step may change very quickly. Further, it is also important to let the cell relax for a certain period to enable the stabilization from the incurred changes in its cell-intrinsic states (e.g.: proper diffusion of ions, homogenous concentration over the electrodes and avoid adverse aggregation of side-reactions/by-product/particles near the electrode interfaces) as a result of the charging step. Therefore, a certain level of controlled relaxation is required to ensure sufficient adaptation of the charging method to respond to such demands. The ideal frequency of such relaxations and their durations may also vary depending on many factors such as the internal kinetics, SoC, and SoH of the cell.

As per the ISVZC design in Fig. 7, six "control-processes" have been identified as deemed controls for this method. The "Set-Voltage" control-process is supposed to set the cell voltage precisely to the SV-step derived by the charging method. The "Set Zero-Current" is supposed to set the current to zero, making the battery fully open-circuit. The "Analyze Absorbed-Current Variation" control-process is the core

processing unit of this method. Mainly, it will monitor the variation of current during a short SV-charge step and regulate/adapt the charging strategy. If an increment or a less considerable decay of absorbed current is detected, this suggests the "Decide Next-Voltage-Increment" control-process to continue charging at the same set-voltage. When a strong decay of current is detected, it suggests a small voltage increment based on the gradient of the decay. This gradient is also taken as a measure to define the required relaxation period in between SV-steps.

Here, the "Control Relaxation" decides the time and duration of applying-rest (set zero-current) based on the above gradient and the tendency of charging given by the magnitude of flown current at the previous SV-step. This relaxation allows the cell-intrinsic elements to get settled from the incurred transitions by the preceding set-voltage charge step, and as a result, it helps to reduce the adverse propagation of sub-reactions/agglomeration of by-products which hinders the reversibility of charging reactions. Deciding the rest duration based on the magnitude of the drawn current enables longer relaxation upon the cell experience in absorbing more current than when it draws with a low. This charging design help to avoid/minimize the stress of externally-set fixed charging parameters, and adapt the charging control based on the closely real-time internal cell changes in response to its very own charging steps. Additionally, this method can also be improved by integrating other parameters such as temperature (nowadays practically measurable in large LIBs), SoH, and SoC.

In general, the charging ends once the battery gets fully charged. Here, the "Control Termination" decides the end of the charging based on accumulated SoC. It also recognizes the repetitive rapid decays of current in SV-steps as chargeability rejections and couples with SoC to determine the end of charging. This also benefits in ending the charging process in case of health issues in the cell, without always reaching an SoC count. As such the "Analyze Absorbed-Current Variation" should actively calculate the gained SoC during charging.

The main idea and the novelty of this strategy is the control of the entire charging process based on the natural current responses at each short set-voltage-charging step. This SV will be incremented in small non-linear values proportionate to the tendency for charging derived by the analysis of the absorbed current. Meanwhile, the SV steps are coupled with controlled relaxation steps. In this way, it enables the control of the charging process while actively incorporating the responses influenced by the effects of cell-intrinsic and intermediary

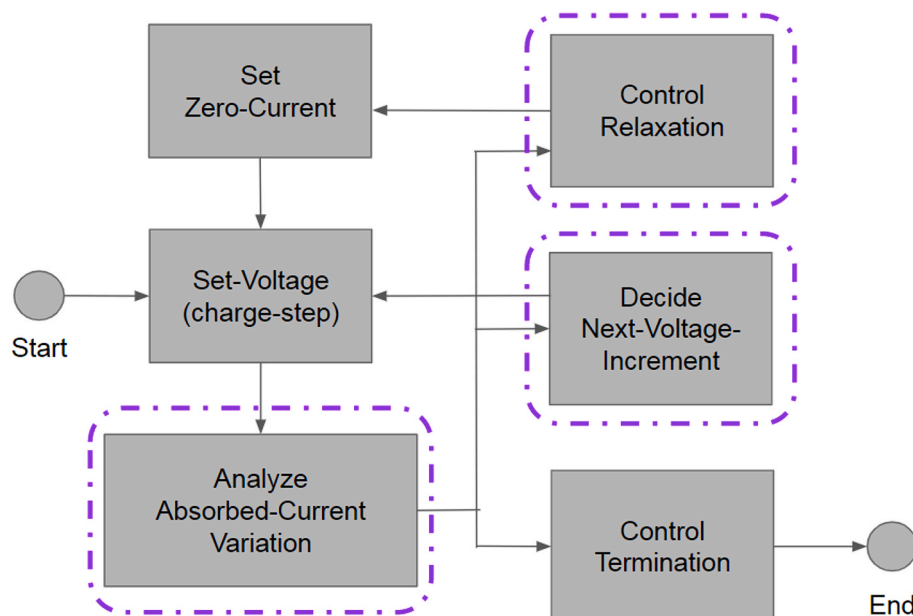


Fig. 7. Main control processes of the recommended next-generation fast charging strategy design for LIBs: Iterated Set Voltage with Zero Current (ISVZC).



reactions. Monitoring it through a series of short charging steps measured by the current, voltage, time, and possibly the temperature is within the affordable computational and electrical capabilities of today's practical chargers. In addition, this design enables incorporating advanced decisions for ending the charging process which can potentially help to avoid the possible destructive incidents caused by continuing charging without caring about the readiness/health of cell for chargeability. Further, this will also open new avenues for research in finding suitable combinations/relationships for each of its control-process to enable optimal regulations of the charging process. This ISVZC charging strategy leading towards natural current-absorption-based fast charging can be considered as a competitive and timely strategy for the next generation of fast charging.

## 5. Conclusion

At the first stage, this paper reviews the electrochemical, structural, and thermo-kinetic variations of LIB cell-intrinsic during the charging process. Then, the charging strategies are presented by a new classification as memory-based and memory-less, depending on whether the memory-based data processing (on battery analytics and prognostics) is used to change the charging parameters during the charging process, and short-cache as a new stream of charging. Lastly, the use of short-cache, short-voltage steps, and relaxations for optimal charging was discussed further and provided recommendations for a new charging strategy. In this new approach, the charging process is adapted in real-time based on cell-intrinsic variations implied by the charging tendency during short voltage steps. It promotes the natural current-absorption-based charging and is considered the next-generation fast charging.

Discussion on the battery intrinsic components, dynamics, and their significant impacts on the charging process provides important insights to the next generation of fast charging. Here, the mass transport, charge transfer, and thermodynamic effects are vital to be considered/monitored for the effective control of the use of LIB. The LIB characterization techniques that provide measures to help monitor such effects are very difficult to implement having account of commercial charger hardware limitations to enable accurate real-time monitoring. Moreover, the analysis on charging strategies provides a detailed understanding of the fast charging limitations in terms of the influences of both the memory factor and fixed controls in failing to adjust over cell-intrinsic variations.

Notably, it should be strongly discouraged to apply constant-current or constant-voltage for fast charging as it stresses the battery through external forces and defeats the purpose of effective performances in many aspects. On one side, the high charge current/voltage cannot be used continuously as they lead to aggregate adverse impacts due to the excessive/side reactions imposed by its opposing internal changes. On the other side, applying the charge current/voltage at a constant rate will imbalance the cell-intrinsic reactions as it ideally looks for varying current absorption (adhering to varying charge and mass transfer dynamics) to maintain their internal equilibrium. In both cases, cell degradation could be accelerated. Additionally, having account of the aforementioned effects an effective charging strategy should be able to autonomously adjust the parameters of the process, following the rapid variations in cell internals, with limited hardware requirements of chargers.

The recommended natural current-absorption-based fast charging design strategy enables adjusting the charging indirectly, through a controlled iteration of the short voltage step-based charging pulses coupled with dynamic relaxation intervals of zero-current. Although the CV charging is limited in most charging methods, its use in short-voltage steps tends to uncover impressive benefits in flowing a peculiar current indirectly driven by its cell intrinsic. This nature is considered an inherent relaxation of this step that promotes the natural current absorption. In addition, the immediate relaxation step allows the cell to settle down from the sudden intrinsic variations at its preceding voltage

step, maintaining good health (reducing the propagation/aggregation of adverse effects). As it uses only the voltage & relaxation steps, along with some computational processing, this new type of charging strategy strongly reduces the complexity for practical charger implementations. In a way, considering the computational overhead concerning small batteries, the high-capacity batteries such as those used in electric vehicles, and smart-grid energy storage systems should get more benefit from this design. Moreover, the possibility of closely real-time analysis of the behavior of this absorbed natural current opens a new window to the battery management systems (BMS) to anticipate battery incidents (before further charging). Therefore, it is highly recommended to intensify the research on this natural-current absorption-based charging design.

In particular, further research will be conducted on the recommended charging pattern, ISVZC - the next-generation fast charging method, to improve its control, expand it to different LIB chemistries, explore its effect on battery life over fast charging. As the paper [138] shows the application of such ISVZC methods only on smartphone battery fast charging, expanding it to larger size batteries would be essential to its adoption in transportation (EVs, e-Trains, e-Busses, e-Aircrafts) and energy renewable/grid storage markets. The demanding LIB fast charging reliability in such growing electrification markets substantiate further research and practical implementations of such next-generation fast charging methods.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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