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Review article

Comprehensive review of multi-scale Lithium-ion batteries modeling: From electro-chemical dynamics up to heat transfer in battery thermal management system

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ABSTRACT

The growing development of lithium-ion battery technology goes along with the new energy storage era across various sectors, e.g., mobility (electric vehicles), power generation and dispatching. The need for sophisticated modeling approaches has become a crucial tool to predict and optimize battery behavior given the demand of ever-higher performance, longevity, and safety. This review integrates the state-of-the-art in lithium-ion battery modeling, covering various scales, from particle-level simulations to pack-level thermal management systems, involving particle scale simplifications, microscale electrochemical models, and battery scale electrical models with thermal and heat generation prediction. Beyond that, authors highlight the growing trend in integrating highly accurate physics-based with thermal approaches such as the electrochemical-thermal coupled model to fully answer the multiscale challenges. Through capturing the electrochemical phenomena and thermal dynamics, and developing a comprehensive understanding of battery kinetics, safety risks such as thermal runaway can be thoroughly mitigated. Authors emphasize the trade-offs between computational efficiency and model complexity, explaining the limitations, strengths, and applications of diverse modeling approaches. This review illuminates the integration of battery management systems and cooling strategies.

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1. Introduction

An urgent paradigm shift is needed in the energy scenario to counter climate change and increasingly frequent energy crises [1]. Currently, government, academia and industry have been making huge efforts to sharply reduce the use of fossil fuels by encouraging the use of renewable and clean energy (wind, nuclear, and solar energy) [2]. In this transition from fossil fuel to renewable energy sources, a new energy storage era has been launched. Indeed, due to the lack of synchronism between energy production and demand, different ways to store excess energy are emerging. Among them, batteries represent a valuable solution. Their adaptability through diverse commercial sectors produced a massive growth of battery storage over the past few years, especially in 2023 [3]. The battery field presents different battery chemistries, such as lithium-ion batteries, Lead-Acid and Ni-MH [4,5]. In particular, lithium-ion batteries show exceptional and remarkable capabilities enabling them to emerge as practical technologies in various domains such as electric vehicles, electronics, and grid energy, as represented in Fig. 1, and to cover up to 90% of the total annual demand [3,6]. Their market has globally doubled its annual deployment compared to previous years. In particular, in the electric vehicle (EV) market, grows by 40%, showing an impressive increase in battery up to 42 GW [3].

Lithium-ion batteries provide high energy density by approximately 90 to 300 Wh/kg [3], surpassing the lead–acid ones that cover a range from 35 to 40 Wh/kg. Besides, due to their high specific energy, they represent the most enduring technology, see Fig. 2. Moreover, lithium-ion batteries show high thermal stability [7] and absence of memory effect [8]. Furthermore, their cost has been reduced in 2023 from around USD 800/kWh to less than USD 140/kWh according to the International Energy Agency special report,[3]. Eventually, the operational temperature range of the Li-ion batteries has also been widened, being typically between −20 °C and 60 °C [9,10]. However, it is important to note that the optimal temperature range for minimizing degradation and maximizing performance is narrower, usually within 15 °C and 35 °C. Maintaining the battery temperature in this range ensures safety and longevity, achieving ideal performance. All these characteristics make Li-ion batteries as the to-go solution in various applications [11].

Given these favorable features, electric vehicle manufacturers across the industry have widely adopted Li-ion batteries, leveraging their full potential. To mention a few in the sport EVs segment: the *Audi e-tron GT Quattro* showing high performance with boasting a 93 kWh gross battery pack and nominal power of 390 kW, and the *McLaren Speedtail*, a hybrid EV with a remarkable battery system storing 1647 kWh gross of energy. Moving to the widely used crossover, the *Tesla Model Y* is characterized by its 60 kWh gross capacity battery pack, supplied by a dual-motor all-wheel-drive system balancing between performance and traction. Furthermore, in the SUV market sector, the *Nissan Ariya* ensures space and capabilities with its two versions, B6 and B9, offering gross capacities of 66 kWh and 87 kWh, respectively. Finally, the *Volkswagen ID4* presents two versions, *Volkswagen ID4 Pure* and *Volkswagen ID4 Pro*, with available battery capacities of 55 kWh and 82 kWh [12].

However, as Li-ion technology continues to grow and the applications demand ever-higher safety and performance, the intricacies of Li-ion batteries evolve, meeting significant challenges. To enhance these technologies, improved numerical and experimental investigations are necessary with the aim to analyze the different aging mechanisms, mechanical stress, and thermal behavior under various operating conditions that are directly impacting the overall performance and longevity.

The multifaced degradation mechanisms, either chemical or mechanical, facing the Li-ion batteries ultimately lead to power and capacity fade caused by solid electrolyte interface (SEI) layer formation and lithium plating. In addition, the mechanical stresses due to volume changes, along with swelling phenomena and particle cracking, can entirely destroy the battery and lead to thermal failures. Operating temperature influences at large-scale the degradation processes. While low temperatures slow down chemical reactions, high temperatures accelerate chemical reactions causing structural changes in electrode materials and promoting electrolyte decomposition. Under high temperature conditions, capacity degradation is induced by lithium loss and active material reductions, while power loss is primarily due to an increase of the internal resistance [10,13].

The associated hazards of the uncontrolled temperature, either through electrical, thermal, or mechanical abuse, accelerate degradations, leading to the generation of more heat and potentially turning into thermal runaway [7]. Thermal runway is a complex risky phenomenon that usually occurs concurrently with internal short circuit (ISC), triggered by exothermic reactions and followed by smoke, gas release, fire, and explosions [14]. Ma et al. [10] and He et al. [15] examined the key factors impacting and fully mitigating thermal runaway, framing it as a safety risk that requires immediate attention and urgent investigation efforts. The challenge lies in balancing the output power requirements and the internal chemical phenomena while maintaining thermal stability.

Addressing this multiscale issue requires a deep understanding of the basics underlying chemistry and physics. Thus, sophisticated modeling approaches with detailed analysis have been developed over years, describing and focusing on the internal electrochemical process inside the battery. These go through the complex chemical reactions at the interface of the electrode–electrolyte to the control of the heat across various battery scales and thermal management strategies, enabling faster charging time and optimizing battery performance [16].

Computational modeling techniques are integrated as an effective and impressive tool in the quest to mitigate Li-ion battery failure. These finally perceive the industry requirements, saving time and cost. Numerical physics-based and thermal models in the fluid and solid domains have been developed with high accuracy [17]. Thoroughly studying the Li-ion batteries across various scales, a wide range of advanced modeling approaches have been developed. Electrochemical models describe chemical reactions occurring inside the battery and capture the Li-ion transport. On the other hand, electrical models use a range of electrical components to form a circuit network. Besides, thermal models have been introduced to analyze heat generation. Building on these models, studies as [18–21] showed improvement by balancing either chemical reactions and electrical aspects with the thermal distribution. They predicted the overall battery performance by developing electrochemical-thermal models as well as electro-thermal models under various hazardous scenarios.

At the end of the article, Table 9 stands as a conclusive summary of this review and presents a synthesis of these modeling approaches,

Nomenclature

Acronyms

AAM	Anode Active Material
AI	Artificial Intelligence
ARC	Accelerating Rate Calorimetry
BMS	Battery Management System
BTMS	Battery Thermal Management System
CC	Constant Current
CEI	Cathode Electrolyte Interphase
CFD	Computational Fluid Dynamics
CNT	Carbon Nanotube
CV	Constant Voltage
DC	Direct Current
DEC	Diethyl Carbonate
DFN	Doyle-Fuller-Newman
DFT	Density Functional Theory
DMC	Dimethyl Carbonate
EC	Ethylene Carbonate
ECM	Equivalent Circuit Model
ECTC	Electrochemical Thermal Coupled
EMC	Ethyl Methyl Carbonate
ESC	External Short Circuit
ETC	Electro-Thermal Coupled
EV	Electric Vehicle
FEA	Finite Element Analysis
FEC	Fluoroethylene Carbonate
HGR	Heat Generation Rate
HPPC	Hybrid Pulse Power characterization
IGF	Amorphous Intergranular Film
ISC	Internal Short Circuit
LFP	Lithium Iron Phosphate
LIB	lithium-ion Battery
LMO	Lithium Manganese Oxide
LTO	Lithium Titanate
MD	Molecular Dynamics
NCA	Nickel Cobalt Aluminum Oxide
NCM	Nickel Cobalt Manganese
OCV	Open Circuit Voltage
P2D	Pseudo-Two-Dimensional
P3D	Pseudo-Three-Dimensional
PC	Propylene Carbonate
PCM	Phase Change Material
PDE	Partial Differential Equation
PE	Polyethylene
PNGV	Partnership for a New Generation of Vehicles
PP	Polypropylene
PVDF	Polyvinylidene Fluoride
RC	Resistance-Capacitance
SEI	Solid Electrolyte Interface
SOC	State Of Charge
SOH	State Of Health
SPM	Single Particle Model
SPMe	Single Particle Model with Electrolyte Dynamics
TMS	Thermal Management System
TR	Thermal Runaway
VC	Vinylene Carbonate

Greek symbols

α	Charge transfer coefficient
δ	Thickness
ϵ	Volume fraction
η	Overpotential
κ	Ionic conductivity
λ	Thermal conductivity
ϕ	Current collector potential
ρ	Density
σ	Conductivity

Latin symbols

$\bar{R}$	Universal gaz constant
A	Active electrode area
a	Specific surface area of the particles
b_k	Correction factor
c	Concentration
C_b	Bulk capacitance
C_c	Surface capacitor
C_E	Energy storage capacity
C_p	Specific heat capacity
$c_{e,ref}$	Reference electrolyte phase lithium-ion concentration
$c_{s,max}$	Maximum concentration of intercalated lithium-ion in active material
$c_{s,surf}$	Surface concentration
C_{Th}	Equivalent capacitance reflecting transient response
D	Diffusion coefficient
E_a	The activation energy for the reaction
E_{OCV}	Open circuit potential
F	Faraday constant
$f_{\pm}$	Electrolyte molecular activity coefficient
I	Current
i_0	Exchange current density
i_e	Electrolyte current density
I_L	Load current
i_s	Solid phase current density
I_{PN}	Outflow current of C_{PN}
I_{Th}	Outflow current
j_r	Molar current flux
K	Reaction rate constant
k_0	Maximum reaction rate constant
R	Resistance
r	Radial coordinate
R_c	Capacitor resistance
R_E	Termination resistor or end resistance
R_o	Ohmic resistance or internal resistance

R_p	Polarisation resistance
R_s	Particle radius
R_T	Resistor terminal
R_{cc}	Current collector contact resistance
R_{SEI}	Resistance of solid electrolyte interface layer
R_{Th}	Polarisation resistance
$s_{i,100\%}$	Solid phase stoichiometry at 100% State of charge
$s_{i,0\%}$	Solid phase stoichiometry at 0% State of charge
T	Temperature
t	Time
t_0^+	Lithium-ion transfer number
U_b	Voltage across bulk capacitance
U_c	Voltage across capacitor C_c
U_d	Voltage across $1/U'_{OCV}$
U_L	Terminal voltage
U_{OCV}	Open circuit potential
U_{PN}	Voltage across C_{PN}
U_{Th}	Voltage across C_{Th}
V	Voltage
x	Spatial coordinate

Subscripts and superscripts

0	Initial
a	Anode
c	Cathode
D	Dimension
e	Electrolyte
eff	Effective
i	Species
Li	Lithium
$n, -$	Negative electrode
oc	Open circuit
$p, +$	Positive electrode
ref	Reference value
s	Solid phase
sep	Separator
tot	Total
x, y, z	Cartesian coordinates

providing a comprehensive overview of the strengths, limitations, and applications of each model.

Despite the huge development and implementation of the numerical modeling approaches, the field of battery modeling lacks a unified and detailed framework combining and comparing these diverse methods. This makes researchers blocked in the welter of models, experiencing difficulties in choosing an appropriate approach for a given scenario. Moreover, they miss the opportunity to couple different modeling approaches, restricting their knowledge about the internal degradation process. Thus, limiting further advancement in the optimization of particular segments.

This review aims to bring clarity to the multitude of models in the literature. Not only that, but this work provides a deep understanding of the fundamental electrochemistry of the Li-ion battery with a comprehensive analysis of the cutting-edge battery modeling techniques. Herein, the key challenges facing these models are also described. Exploring the principle operation and mechanism of thermal runaway, the reviewed models include an accurate temperature control and extend findings to prevent thermal runaway propagation.

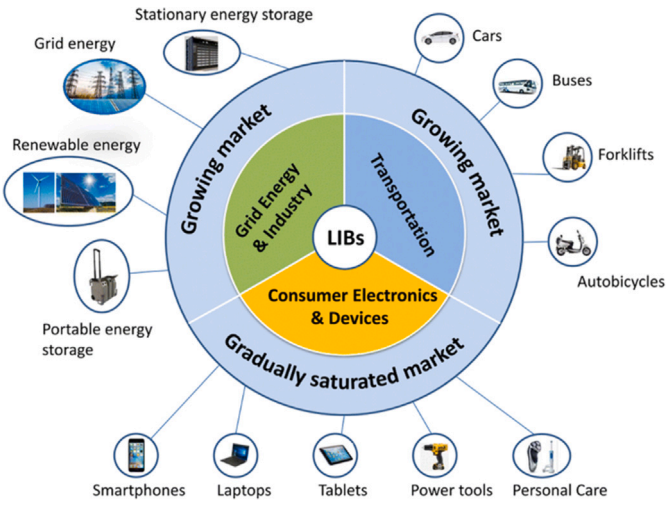


Fig. 1. Li-ion batteries applications across various market sections: from electronics to transportation and grid energy [22].

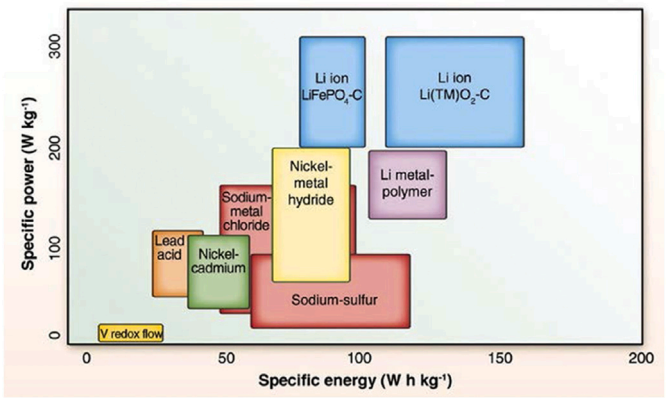


Fig. 2. Illustration of different battery chemistry and their performance in terms of their specific power and specific energy, where the Li-ion $Li(TM)O_2-C$ battery shows the highest combined specific power and energy [23].

This work offers a state-of-the-art overview of the Li-ion battery. It encompasses intricate ion transport phenomena, degradation processes, and heat generation dynamics. The review covers sophisticated multi-scale modeling approaches, ranging from particle-level simulations to pack-level battery thermal management systems. The authors provide a systematic comparison of the different physics-based and thermal models in a straightforward manner. They analyze each models limitations, strengths, and computational cost requirements.

The review is organized as follows: Section 2 provides an overview of the fundamental basis operations, key concepts, and degradation mechanisms of Li-ion batteries. Section 3 delves into the heat generation dynamics and effects, including thermal runaway phenomenon. Section 4 explores in-depth the different battery modeling approaches at various scales. Section 5 proposes some recommendations to orient future research on battery modeling towards most impactful and innovative directions. Eventually, Section 6 concludes with a summary of the key findings and recommendations for future research.

2. Lithium-ion battery: overview and general consideration

This section provides a detailed comprehensive understanding of Li-ion batteries. It examines their main fundamental components, operating mechanisms, and key performance parameters. The section also explores their degradation mechanisms and the complex chemical and physical interactions involved.

Table 1
List of materials commonly used in industry-scale production of LIBs.

Category	Materials
Electrode active materials	LFP, NCM, NCA, LMO, graphite, LTO
Electrode supportive materials	copper foil, aluminum foil, PVDF, NMP, CNT
Separator	PE diaphragm (DP), PP diaphragm (WP)
Electrolyte materials	LiPF6, EC, DEC, DMC
Housing	aluminum, steel

2.1. Components

Despite the diversity among the four types of Li-ion batteries, see Fig. 3, cylindrical, prismatic, coin, and pouch cells, all these cell structures share the same basic mechanism: the movement of Li-ions between the electrodes [24].

A schematic for Li-ion battery is presented in Fig. 4 highlighting its basic essential components, such as the cathode positive electrode, the anode negative electrode, the electrolyte, the positive and negative current collectors, and the separator.

The anode, negative electrode, active materials (AAM) include silicon based, nitrogen-doped materials [25] carbon-based materials. However, graphite is the preferred choice due to its electrical conductivity, structural stability and affordability [26,27].

On the other hand, metal oxides serve as cathode, positive electrode, active materials. These include layered, spinel, and olivine structures such as lithium cobalt oxide (LiCoO2), lithium manganese oxide (LiMn2o4) and lithium iron phosphate (LiFePO4). Each is chosen for their energy density, thermal stability, and cost-effectiveness [28].

Wang et al. [29] achieved an excellent performance by combining lithium cobalt oxide with graphite anodes, leveraging the high energy density of LiCoO2 and the stability and conductivity of graphite.

Large-scale manufacturing classifies Li-ion batteries materials into five main categories based on their components and functions [9,30], as shown in Table 1.

The electrolyte facilitates the essential Li-ion movement, promoting ion transport between the cathode and the anode [31]. It consist of a mix of organic solvents that have a high dielectric constant, such as ethylene carbonate (EC), dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), and propylene carbonate (PC) [32], with lithium salt, such as lithium hexafluorophosphate (LiPF6), lithium perchlorate (LiClO4), and lithium bis(trifluoro-methanesulfonyl)imide (LiTFSI) [1,33], ensuring high ionic conductivity and stability under various operating conditions [1,34]. Besides, some additives, including vinylene carbonate (VC) and fluoroethylene carbonate (FEC), help improve the solid electrolyte interphase (SEI) layer formation on the anode [35].

Battery electrolytes are classified into five types in terms of density, safety, and cost: ionic liquid, polymer, aqueous, non-aqueous, and hybrid electrolytes, as illustrated in Fig. 5.

2.2. Mechanism and operation

The principal mechanism of Li-ion batteries is based on the intercalation and deintercalation of Li-ions into and out of the electrode materials. During the charging process, Li-ions are extracted from the cathode and inserted into the anode. Conversely, during the discharge process, Li-ions migrate back to the cathode from the anode. The electrolyte facilitates the ion movement, acting as a medium and maintaining electrical insulation [38].

Taking lithium cobalt oxide (LiCoO2) and graphite as an example. Graphite serves as an intercalation host, forming binary/ternary graphite intercalation compounds [39], as well for the lithium cobalt oxide (LiCoO2).

The fundamental electrochemical reactions [40] occurring in Li-ion batteries during charging phase are summed up as follows:

Table 2
The capacity values for different battery types [48].

Battery Type	Typical Capacity
Lead-Acid Batteries	Small lead–acid batteries typically have a capacity of approximately 1 Ah, whereas huge deep-cycle batteries used in renewable energy systems have a capacity of over 200 Ah.
Nickel-Metal Hydride (NiMH) Batteries	For AA and AAA sizes, these batteries generally have capacities between 600 mAh and 2.5 Ah. The capacity of larger NiMH batteries used in electric cars can exceed 100 Ah.
Lithium-ion (Li-ion) Batteries	The capacity of a common Li-ion cell (such as in 18650 battery) ranges from 1.5 Ah to 3.5 Ah. Electric car batteries with larger pouch or prismatic cells can have capacities ranging from 20 Ah to more than 200 Ah.
Lithium Iron Phosphate (LiFePO4) Batteries	The capacity of these batteries is often a little bit lower than those of other lithium-ion chemistries. Depending on size and application, cells typically vary from 15 Ah to 200 Ah.
Nickel-Cadmium (NiCd) Batteries	These batteries typically have a lesser capacity than NiMH batteries, with an average capacity of 600 mAh for AA-size batteries and 50 Ah for bigger cells.

- Cathode Reaction:

$$LiCoO_2 \rightarrow Li^+ + CoO_2 + e^- \quad (1)$$

LiCoO2 releases Li-ions and electrons.

- Anode Reaction:

$$C + Li^+ + e^- \rightarrow LiC_6 \quad (2)$$

Graphite intercalates/adds Li-ions and electrons.

During the discharging process, electrodes have reversed reactions, moving the ions from the cathode and intercalating them into the anode structure.

2.2.1. Voltage and capacity of Li-ion batteries

Cycles directly affect battery voltage over time, impacting its performance. Lui et al. [41] illustrated in Fig. 6 how charging voltage increases gradually due to rising of Li-ion concentration. The optimal and safe operating range of the voltage in Li-ion batteries is between 2.5 and 4.2 V, ensuring performance and longevity [42].

One of the key parameters during these processes is the state of charge (SOC). The SOC indicates the battery leftover capacity based on its total capacity. In other words, it represents the ratio of the available charge to the maximum charge that the battery can hold in ampere-hours (Ah) [43,44]:

$$SOC = \frac{Q_{available}}{Q_{rated}} \quad (3)$$

where $Q_{available}$ is the current available charge amount in the battery and Q_{rated} is the maximum charge that the battery can hold, known as the battery capacity.

A fully charged battery is at 100% SOC, whereas at 0% SOC the battery is fully discharged. In order to optimize and accurately estimate charge level, it is required to understand the relationship between voltage and SOC [45,46].

Evaluating the battery performance necessitate a deep study of capacity — the total amount of energy a battery can store, which decreases with cycling [47]. As represented in Table 2, Li-ion batteries offer higher capacity than lead–acid and NiMH alternatives, enabling superior performance in advanced applications [48].

Therefore, from 2016 to 2023, electric vehicle manufacturers raised the demand for the Li-ion batteries, as illustrated in Fig. 7. The trend shows a significant increase, especially in regions like China and Europe, reaching nearly 900 GWh per year in 2023.

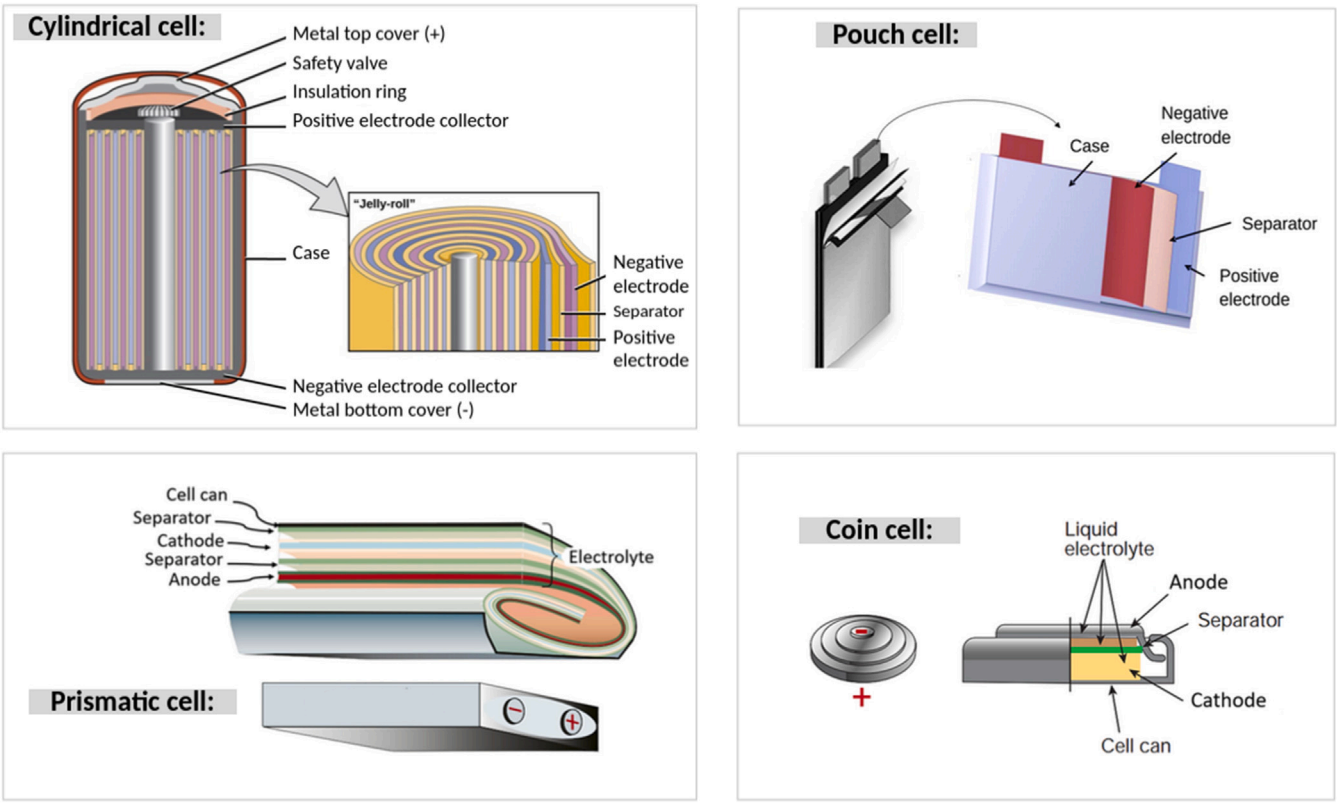


Fig. 3. Schematic representation of different cell shapes with focus on their internal components arrangement, cylindrical [36], pouch [37], prismatic [9], and coin cells [9] in Li-ion batteries.

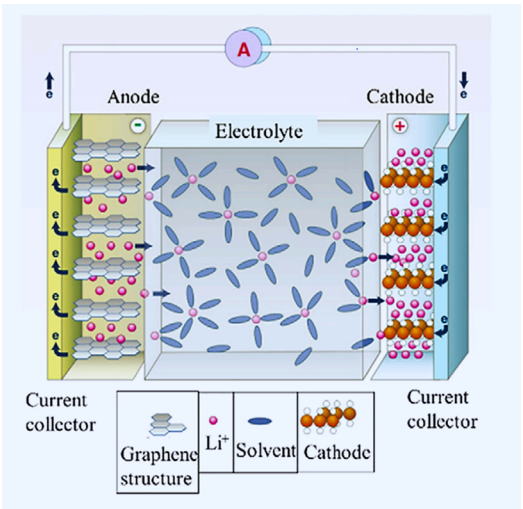


Fig. 4. Schematic representing the key components of a lithium-ion battery: anode, cathode, electrolyte, current collectors [9].

2.2.2. Charge rate

The production of stress and heat, under various conditions, can influence the battery capacity by impacting the charge rate [50]. The charge rate, also known as C-rate, quantifies the rate at which the battery is charged or discharged. It is defined as the ratio of the charging or discharging current (A) to the nominal capacity of the cell (Ah), it is expressed as follows:

$$\text{C-rate} = \frac{\text{Charging or discharging current}}{\text{Nominal cell capacity}} \quad (4)$$

Taking as example, charging a 2500 mAh battery with 5000 mA current results in having a 2C as C-rate, which means that the battery is fully charged in half an hour. Moreover, faster power delivery can be achieved with higher C-rates from 3C up to 30C, although over time, the overall battery capacity drops. On the other hand, C-rates lower than 1C, like C/20, C/3, and C/2, allow for more complete utilization of the active materials and potentially lead to increased degradation mechanisms [51].

The charge rate of the battery affects the rate of the Li-ions intercalation and deintercalation within the electrode materials. Higher rates can increase internal resistance and accelerate degradation [52].

2.2.3. Degradation mechanisms

During cycling, Li-ion batteries undergo intricate and multifaceted phenomena known as degradation mechanisms, as shown in Fig. 8(a). These chemical and mechanical processes ultimately lead to capacity loss and power fading, illustrated in Fig. 8(b).

Chemical degradation primarily occurs through electrolyte decomposition, solvent co-intercalation, material dissolution, gas evolution, SEI formation, and lithium plating [54,55].

The SEI layer formation and the lithium plating phenomena represent the most significant degradation mechanisms. They substantially impact cell internal resistance, capacity, and power. The SEI is a

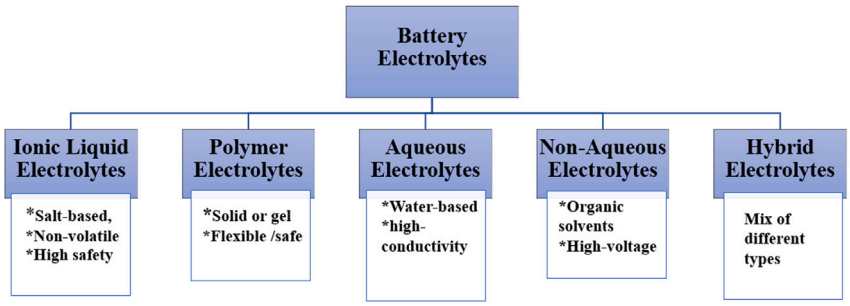


Fig. 5. Classification of battery electrolytes.

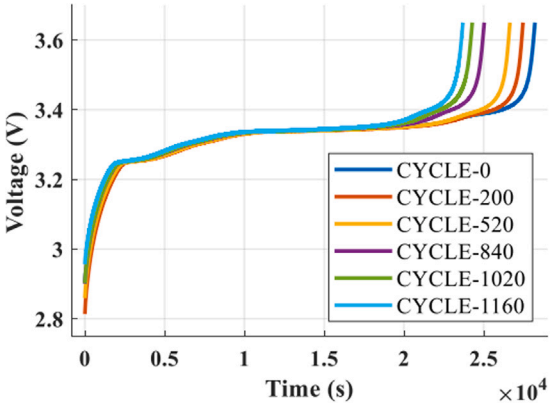


Fig. 6. Voltage response over multiple charge cycles [41].

passivation layer that occurs primarily during the first few cycles, as the electrolyte components decompose and settle on the anode surface. This process is established by the reduction of the electrolyte solvents, such as ethylene carbonate (EC), propylene carbonate (PC), diethyl carbonate (DEC), and dimethyl carbonate (DMC), that affects the formation of this layer and its stability. The SEI layer acts as a protective barrier, enabling lithium-ions to pass through while preventing further electrolyte decomposition. Further, this continuous decomposition can lead to more Li-ion and electrolyte loss. While the SEI stabilizes the electrode-electrolyte interface, the thickness of the SEI layer may block the diffusion pores and potentially lead to increased internal resistance, capacity, and power fade [53,56].

Lithium plating, on the other hand, occurs when metallic lithium deposits on the anode surface rather than intercalating, typically during fast charging or under low temperature conditions. This can lead to reduced capacity, altered SEI composition, volume expansion, and potentially hazardous dendrite formation leading to internal short circuits [57].

Mechanical degradation, on the other hand, occurs through repeated volume changes during lithium intercalation and deintercalation. These changes generate stresses that crack and fracture active materials and disrupt electrical contacts, particularly in the anode, where expansion cycles break and reform the SEI layer. In the cathode, excessive cycling causes particle fracture and active material dissolution, specifically in manganese-based materials, which accelerate the overall degradations [58].

Additionally, a key aspect of mechanical degradation is swelling, the volumetric expansion and contraction of electrode material. This alter the battery shape and, in extreme cases, it cracks the cell [59,60]. Li et al. [61] demonstrates that silicon anodes, despite their high lithium capacity, undergo volume changes up to 300% during cycling, with significant capacity fading over 500 cycles, as represented in Fig. 9. Ki-Yong Oh et al. [62] found that during a 0.4C charging, the anode-separator-cathode layer expands by 1.5%, due to swelling, causing mechanical stresses and SEI disruption through thermal expansion.

Furthermore, swelling can deform separator pores, increasing internal resistance and reducing ionic conductivity [59].

2.2.4. Progress in predicting the life of lithium-ion batteries

Building upon the understanding of degradation mechanisms and key performance in Li-ion batteries, significant advancements have been achieved in predicting battery lifespan. Predictive models have evolved from simple empirical approaches to sophisticated multi-physics models, that will be explained in details in Section 4.

Incorporating degradation processes provides a comprehensive framework for analyzing the battery aging. This combines data-driven approaches and machine learning to analyze large scale real-world battery usage data under various conditions.

Advanced methodologies have been included to simulate long-term aging across different times, temperature, and scales, from atomic to macroscale [63,64]. Hence, understanding the thermal, mechanical and chemical interactions is crucial for optimizing the battery longevity and reliability.

3. Heat generation in Li-ion batteries: sources, effects, and scale interactions

Various internal processes, including electrochemical reactions, internal resistance, and thermal runaway mechanisms, promote heat generation within the battery, as summarized in Fig. 10, which highlights its complex multifaceted nature [10,65].

Li-ion batteries generate heat through irreversible processes such as Joule heating, also known as ohmic heating, and reversible heat. Irreversible heat occurs due to the internal resistance and polarization losses at the interfaces. Whereas, reversible heat is produced by the electrochemical reactions at the electrode-electrolyte interfaces, in other words, the intercalation and deintercalation reactions [66].

3.1. Temperature-dependent factors and their impact on Li-ion battery performance

Heat generation manifests and interacts differently across various scale, from particle to pack level, as illustrated in Fig. 11, which shows the key heat generation factors at each scale.

High temperature boosts the electrochemical reactions due to increased consumption in the electrolytes and available Lithium [68,69]. Leng et al. [70] proved an increase in the maximum charge storage capacity using a Sony prismatic cell under an operating temperature between 25 °C and 55 °C. Additionally, the latter shows a boost in the degradation capacity rate over cycling induced by higher temperatures, as represented in Fig. 12. These affect phase changes, create surface modifications, and cause electrode deterioration.

Further, low temperatures, generally below 0 °C, significantly degrade the overall performance. This occurs through increased electrolyte viscosity, reduced ionic conductivity, and higher internal resistance [71]. Aris et al. [72] found that reducing temperatures from

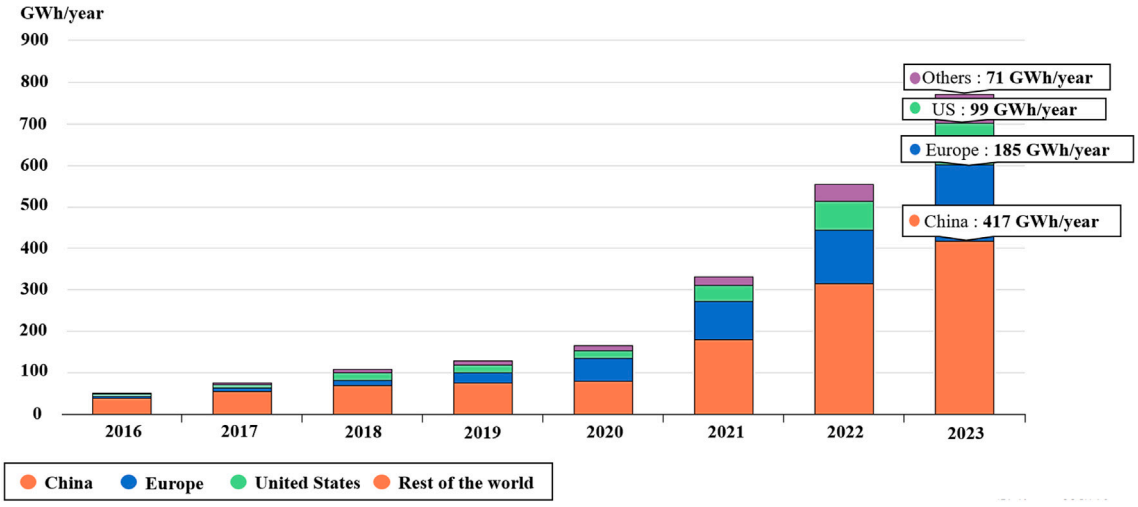
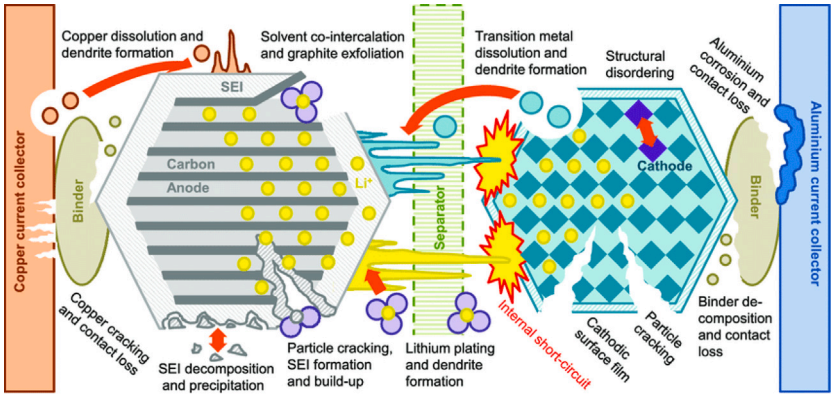
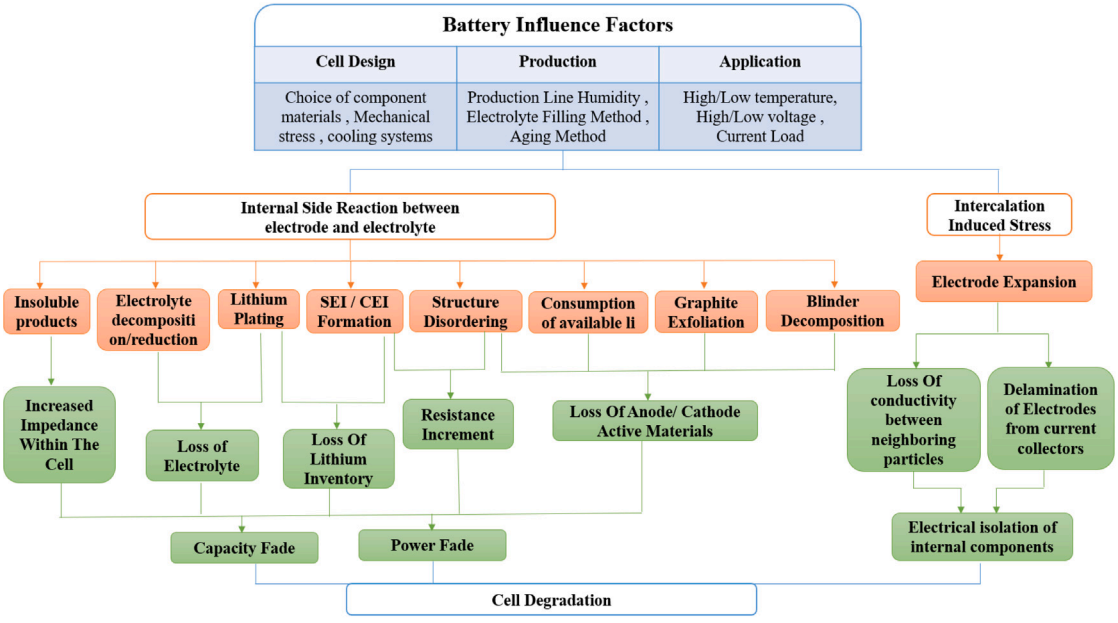


Fig. 7. The annual electric vehicle battery capacity demand by region from 2016 to 2023 in GWh/year [49].



(a)



(b)

Fig. 8. (a) Overview of different degradation mechanisms in Lithium-ion batteries [53], (b) Pathways and consequences of cell degradation in Lithium-ion batteries.

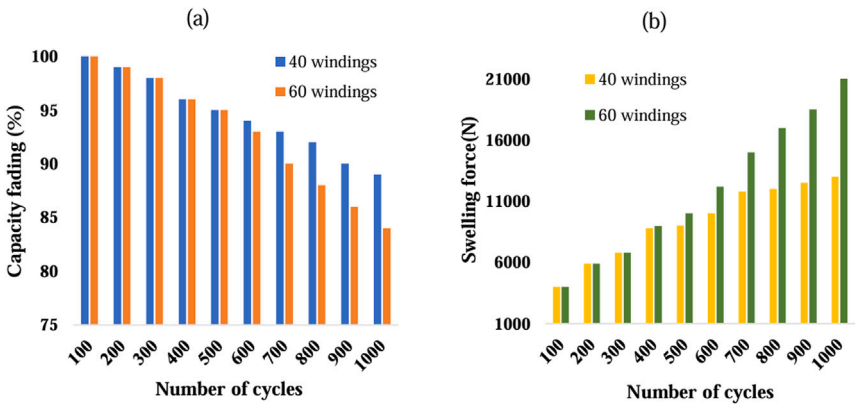


Fig. 9. Variations of battery swelling force and capacity fading with respect to battery life cycles [61].

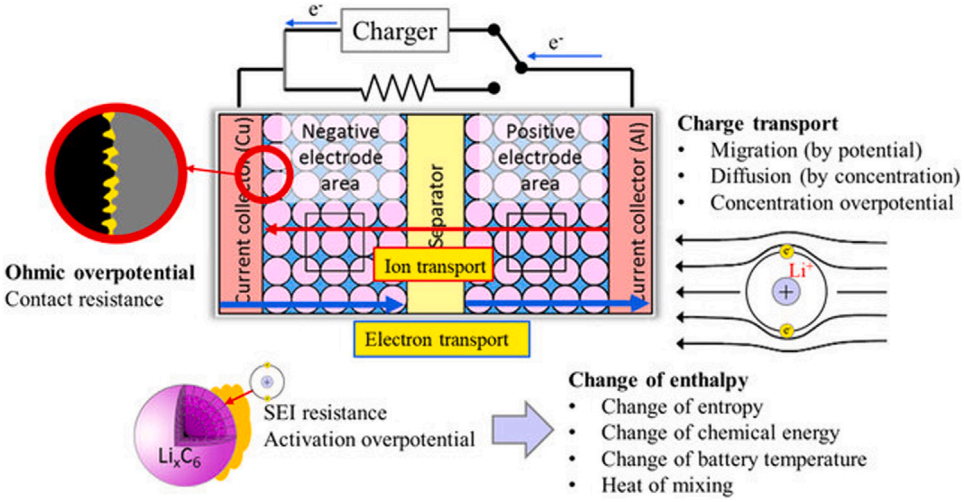


Fig. 10. Mechanisms of overpotential and energy transport in Li-ion batteries [67].

25 °C to −15 °C decreased the SOC by 7%–23% during charging, due to increased impedance effects.

Uncontrolled temperatures, whether high or low, trigger hazardous scenarios such as thermal runaway (TR) and impact battery safety. Fig. 13 highlights the importance of maintaining optimal temperature range to prevent thermal runaway.

3.2. Thermal runaway

Thermal runaway poses pivotal safety risks given its potential to induce explosions, gas generation or fires.

For a battery pack, thermal runaway in a single cell trigger catastrophic failure through chain reactions and component breaking down [79,80].

Thermal runaway process usually starts at 135 °C with a sharp temperature increase and voltage drop, indicating separator melting and battery swelling [81]. As shown in Table 3, a series of exothermic reactions can lead to complete TR above 450 °C, producing combustible and toxic gases through electrolyte decomposition [68,82].

Doughty et al. [84,85] examined the thermal runaway behaviors of 18650 type Li-ion batteries using accelerating rate calorimetry (ARC), outlining these exothermic reactions in three stages of the TR process, as illustrated in Fig. 14:

- Stage 1: The onset of overheating
- Stage 2: Heat accumulation and gas release process
- Stage 3: Combustion and explosion

3.2.1. Abuse mechanisms triggering thermal runaway

The root cause of TR can be mechanical [87], thermal [88], or electrical abuse [89].

a. Mechanical abuse

Mechanical abuse, such as nail penetration or structural degradation, forms a significant determinant of the internal short circuits (ISC) [90]. Fig. 15 simplifies the difference between ISC and external short circuit (ESC). Zhang et al. [91] studied thermal runaway in large-format Li-ion batteries using nail penetration technique, revealing how battery design affects TR propagation speed. Wang et al. [92] compared TR behavior across different nickel contents and NCM cathodes types, highlighting the structural impact of structural differences on thermal propagation.

b. Electrical abuse

Electrical abuse through overcharging causes excessive energy accumulation, side reaction heat, and joule heating, leading to thermal runaway [90,94]. Hamisi et al. [95] developed a 2D electrothermal model for an 18650 cylindrical battery under overcharged abuse and high hazardous thermal scenarios. They demonstrated an increase in the battery surface temperature with charging current. Ren et al. [14] found that while overcharge rates do not significantly alter TR characteristics, they do elevate voltage and temperature boost.

c. Thermal abuse

Thermal abuse often results from mechanical or electrical stress-induced overheating or improper connections, triggering exothermic reactions and ISC from separator shrinkage [90,96].

Understanding thermal characteristics of lithium-ion batteries under various operating conditions is critical for battery safety. The

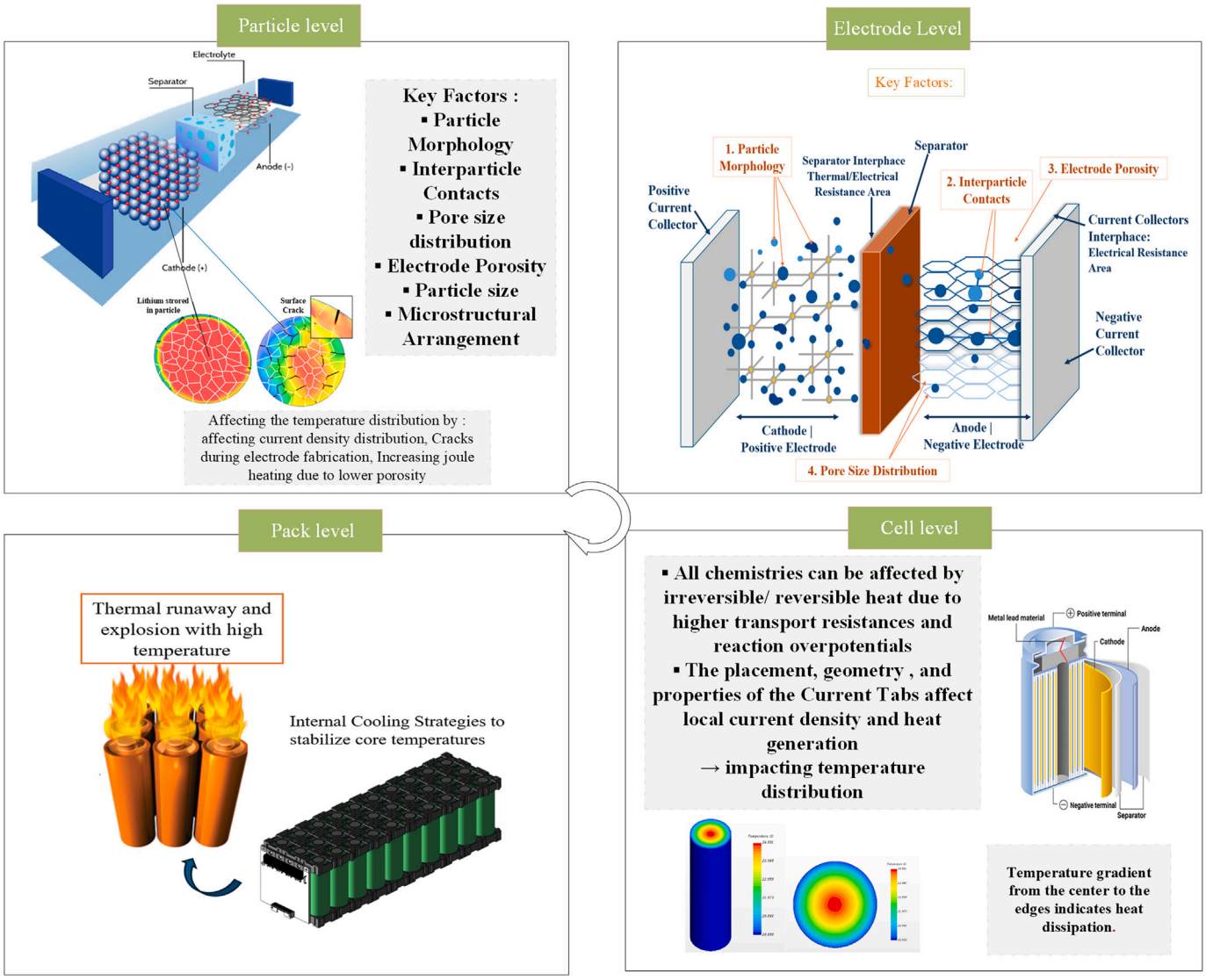


Fig. 11. Multi-scale heat generation and management in lithium-ion batteries.
Source: Adapted from [73–78].

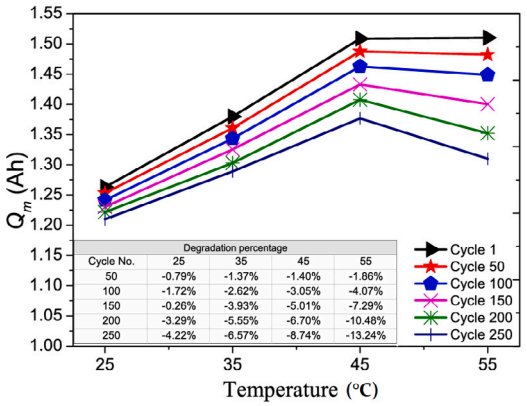


Fig. 12. Maximum charge storage capacity as a function of temperature: effect of temperature on battery capacity and degradation across multiple charge cycles, from [70].

widespread use of Li-ion batteries in real-world applications necessitates developing multi-dimensional thermal-electrochemical models to predict thermal behavior across different temperatures and hazardous scenarios.

3.2.2. *Safety features after thermal runaway*

Preventing thermal events and catastrophic failures requires multi-faced safety features, operated at various scales, from the cell to the battery pack level.

At the cell level, commercial cells, such as 18650 cells, incorporate directional safety vents and current interrupt devices (CID) that manage internal pressure relief and isolation. These enable controlling the temperature by an accurate mechanical design. Besides, applying multi-layer separators act as thermal fuse and provide thermal shutdown capabilities [97].

At the module or pack level, hot gas distribution is controlled by a strategic and sophisticated mechanical design of casing and vents. Advanced venting systems and thermal barriers are integrated to prevent cell-to-cell thermal propagation. They enhance thermal safety through spray sensors and mist cooling systems, providing rapid temperature regulation [98]. Active monitoring coordinated by the battery management system allows early detection and preventive shutdown: temperature sensors that track surface and internal temperatures, spray

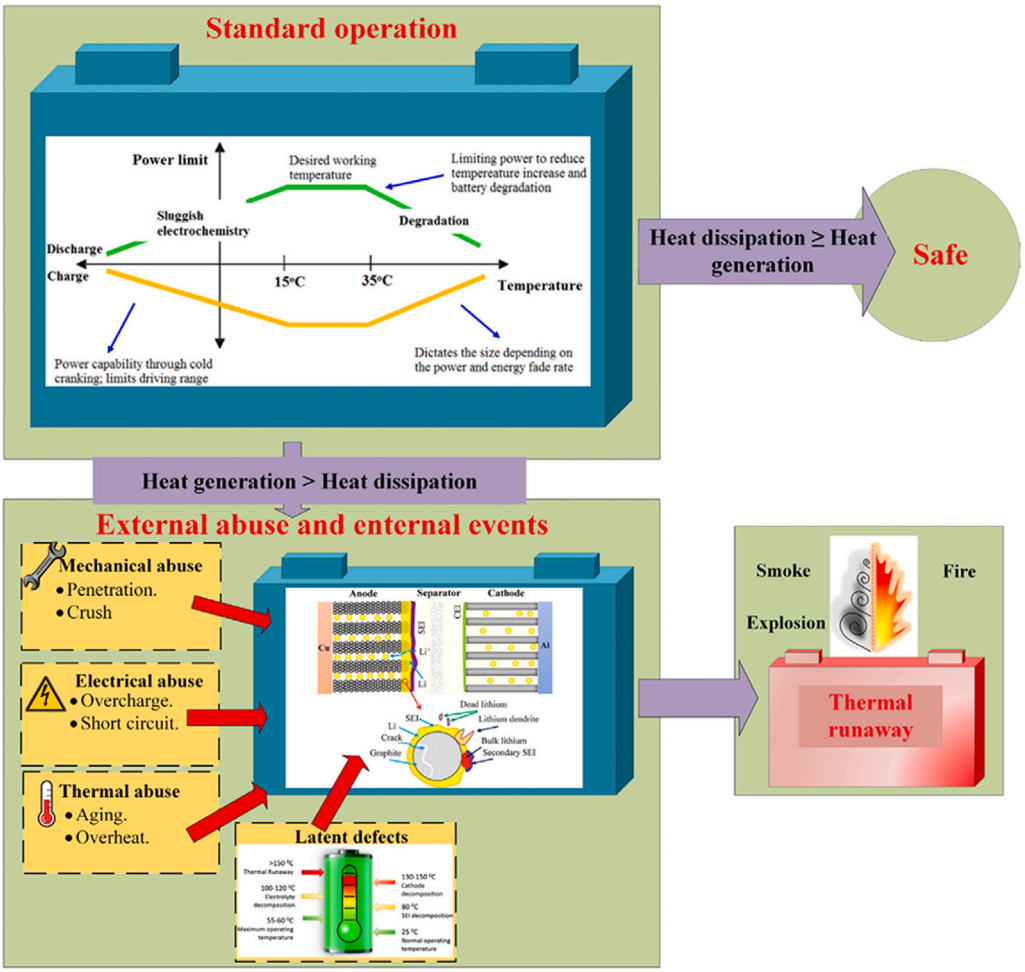


Fig. 13. Lithium-ion battery safety and thermal management: standard operation vs. thermal abuse scenarios, from [11].

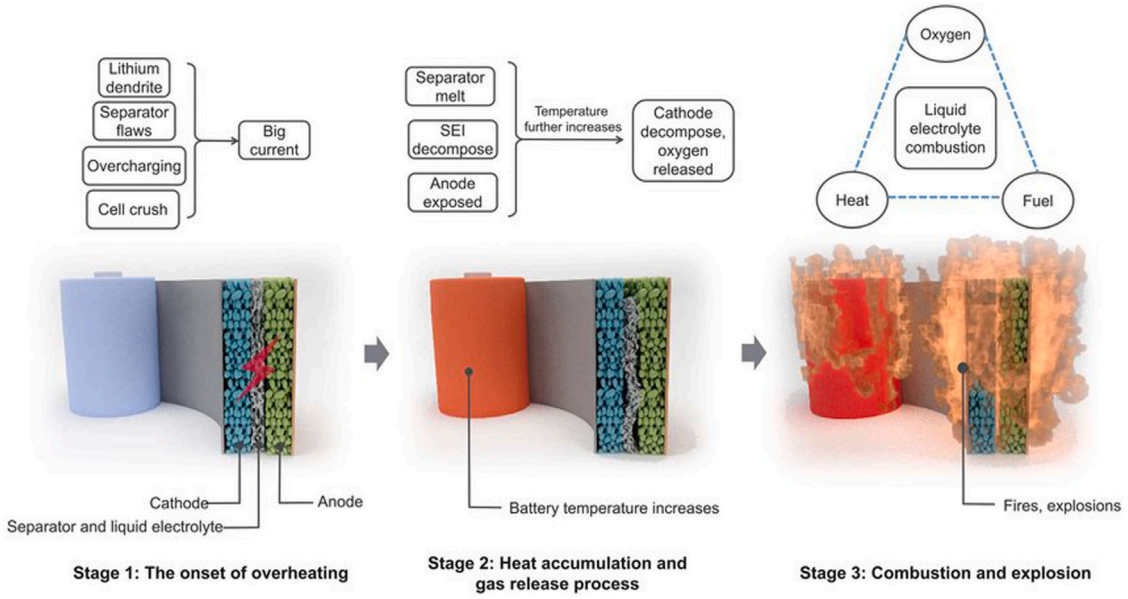


Fig. 14. Three stages of the thermal runaway process. Stage 1: the onset of overheating. The batteries change from a normal to an abnormal state and the internal temperature starts to increase. Stage 2: heat accumulation and gas release process. The internal temperature quickly rises, and the battery undergoes exothermal reactions. Stage 3: combustion and explosion [86].

Table 3
Thermal runaway exothermic chain reactions and consequences on lithium-ion batteries. *Source:* Adapted from [83].

Temperature (°C)	Reaction	Consequence
30 to 60	Reactions of Li and electrolytes	SEI Growth, dead lithium
60 to 130	Initial SEI decomposition	Heat, flammable gases release, oxygen
110 to 250	Balanced reaction of SEI decomposition and regeneration: Intercalated Li in the negative electrode and electrolyte reaction	Heat, flammable gases, lithium oxides
120 to 160	–	Separator shutdown (pore closure)
130 to 250	Positive electrode decomposition	Heat, oxygen release
200 and above	–	Hard short circuit
230 to 350	Electrolyte decomposition	Heat
230 and above	Li metal reactions with binder and air after venting	Heat, fire, hydrogen production
250 and above	Decomposition of graphite with electrolyte (Graphite structure collapses, balance of SEI decomposition and regeneration is broken)	high risk of heat spread towards adjacent cells leading to thermal runaway
450 and above	Combustion	Complete thermal runaway

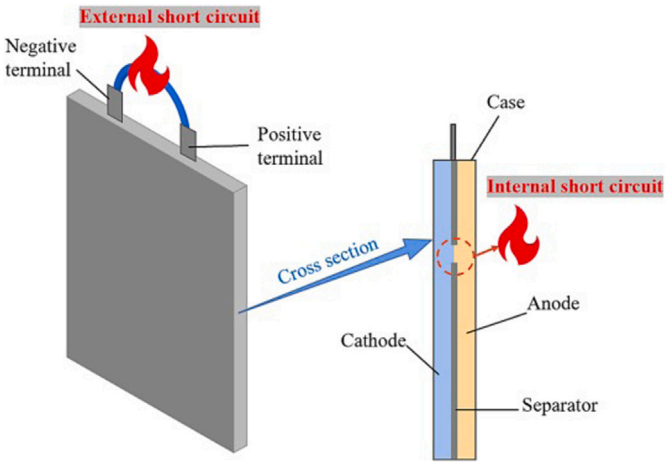


Fig. 15. Representation of the difference between ISC and ESC [93].

detection systems, and cell voltage surveillance that detect continuously abnormal voltage behaviors, short circuits, and degradation signals [97,99].

4. Battery modeling approach

Thermal runaway emerges from complex interactions between thermal, electrochemical, and mechanical processes, requiring a multifaceted approach to ensure material, cell, and system safety [100,101]. This strategy focuses on preventing hazardous scenarios, limiting the feasible damage propagation, immediately relieving dangerous situations, improving thermal resilience, and incorporating early detection systems. Which aims could be realized by taking into account the mechanical protection safeguards, chemical innovations, and thermal management techniques [102,103].

4.1. Overview of modeling techniques

The development of cost-effective safety measures for Li-ion batteries relies heavily on sophisticated modeling approaches [102,104]. These models cover a wide range of complexities and applications, ranging from electrochemical simulations as physics-based models which examine internal battery states to simpler electrical models [16, 105].

On the other hand, thermal analysis, can be conducted using a one-dimensional, two-dimensional, or three-dimensional spatial resolution, depending on the heat components considered [106]. Numerous studies use finite element analysis (FEA) and the finite volume method in computational fluid dynamics software (CFD) [107–109] to analyze heat propagation and temperature variations across various cell types.

By combining the strengths of electrochemical simulations and thermal models, internal reactions will be accurately represented and heat transfer dynamic will be captured throughout the cell or battery pack [110]. Researchers such as Chiew et al. [111], Mesbahi et al. [106], and He et al. [110] have adopted coupled electrochemical-thermal models. They demonstrated how a pseudo-3D coupled models enable comprehensive understanding analysis of full-scale battery systems, as illustrated in Fig. 16.

These intricate models are primarily implemented using powerful software platforms such as Python-based packages like PYBAMM [112, 113], ANSYS [114], COMSOL Multiphysics [115], MATLAB, and OpenFOAM [116], carrying their unique capabilities to solve partial differential equations (PDEs) and describe battery dynamics.

The combination of the electrochemical simulations with the thermal models, under different operations, offers a deeper understanding and knowledge of the battery performance.

Hence, accurate and robust prediction necessitates incorporating and testing new model-based methods, such as data-driven approaches. These approaches involve a large amount of data and it is receiving great attention recently [117]. A hybrid approach that combines these physics-based knowledge with data-driven methods could provide a robust framework in predicting battery behavior across various temperatures, aging states and hazardous scenarios [118]. The following sections examine in depth the distinct electrochemical and thermal approaches across various scales. It captures internal processes and thermal management strategies.

The authors focus primarily on physics-based and thermal models. The potential for further investigations and future integration with data-driven methods is addressed in our perspectives section for future research to address current limitations.

4.2. Multi-scale modeling approaches

Multiscale modeling bridges the gap between microscopic interactions and macroscopic behaviors in Li-ion batteries. The growing complexity of battery systems necessitates a detailed understanding of the underlying transport phenomena across various spatial and temporal scales, crucial for manufacturing insights.

This section examines the distinct modeling approaches, their governing equations, advantages, and limitations across various levels, as illustrated in Fig. 17. These models are based on fundamental principles governing electrolyte and electrode particles processes.

Nevertheless, before examining the multi-scale battery modeling, it remains important to recognize the fundamental molecular-scale modeling techniques. Recent research investigations have been extended at this level, including molecular dynamics (MD) simulations and density functional theory (DFT) calculations. They showcase significant insights into the battery materials, interface reactions and atomic and electronic dynamics. Molecular dynamics simulations reveal the Li-ion diffusion through the electrode active materials. They have been incorporated in simulation of the initial formation of the SEI layer at the solvent-graphite interface and the lithium-ion transport through the

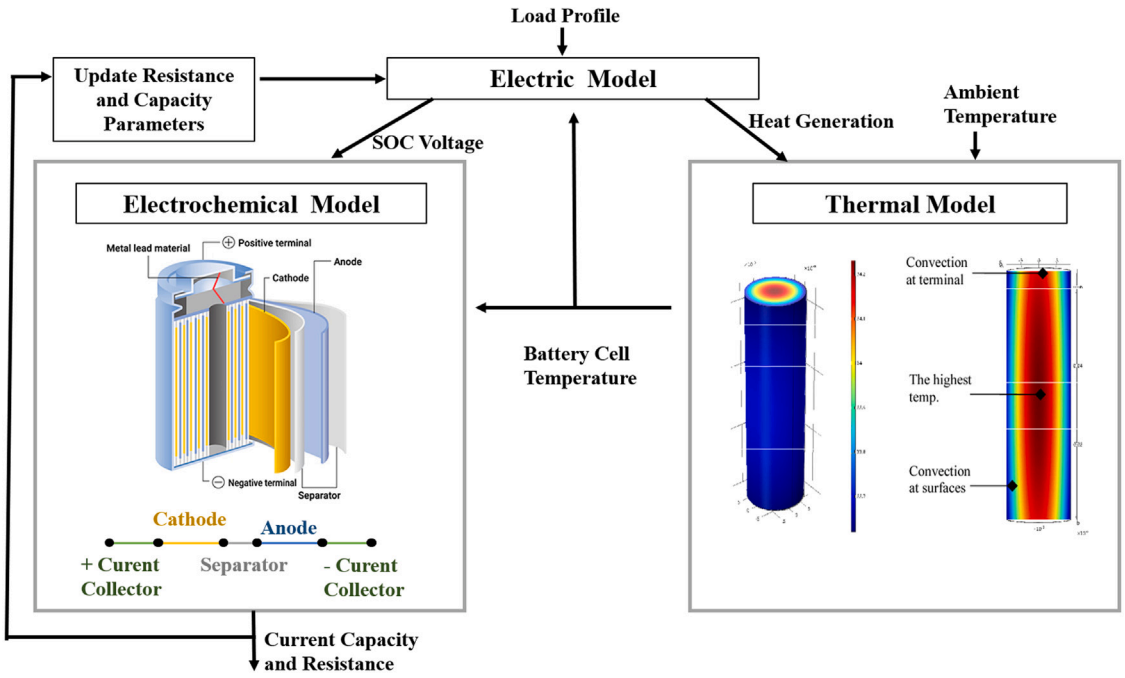


Fig. 16. Coupled electrochemical, electrical and thermal battery models layout.
Source: Adapted from [75,119].

polycrystaline cathode, in particular, the diffusion through the amorphous intergranular film (IGF). DFT calculations have proven effective prediction and explanation of the structural changes when electrode materials are subjected to multiple cycles. They have been employed in determining the materials electronic structure and examining the interaction effects on mechanical properties and unit cell expansion.

For instance in Ramadesigan et al. [120], the authors provide a comprehensive understanding framework that integrates molecular-scale insights with the microscale and macroscale models, combining physics-based and data-driven methods. Despite the accurate and fast MD and DFT simulations and their power in providing atomic-scale understanding, their detailed discussion lies beyond the scope of this review, which deals with models at a slightly larger scale.

Hence, this section analyzes thoroughly the multiscale electrochemical and thermal modeling approaches across various scales. It describes the microscale models, in Section 4.2.1, where Doyle–Fuller–Newman (DFN) and pseudo-two-dimensional (P2D) models offer highly accurate simulations of electrochemical transport operations. The particle scale models in Section 4.2.2 present the single particle model (SPM) and its extension, with electrolyte dynamics (SPMe), providing a compromise between computing efficiency and simplicity. Furthermore, at the battery-level, in Section 4.2.3, the equivalent circuit model (ECM) is presented and offered a comprehensive understanding on the battery electrical behavior. Stepping up in scale, Section 4.2.4 offers an explanation of the thermal approaches implementation. Finally, the application of the battery thermal management system techniques and the fluid modeling approach at the pack level is detailed in Section 4.2.5. After fully understanding the challenges and analyzing each model in depth at each scale, Table 9 summarizes the key features of these multiscale modeling approaches, the advantages, limitations, applications and softwares used in some literature for implementing these models.

4.2.1. Microscale model

The microscale approach evolved to be the fundamental basis of the battery modeling. It provides a detailed overview of the various electrochemical reactions occurring within the battery. Herein, the pseudo-2D model (P2D) and Doyle–Fuller–Newman (DFN) are presented since they are currently at the cutting edge of the microscale modeling [102,127].

a. Pseudo-two-dimensional (P2D) model

The P2D model is based on Doyle's work [128]. It considers the battery as a layered structure, in which the layer thickness consists of the two electrodes and the separator. It considers also the particle radial variations along the active battery particles. Fig. 18 shows a schematic representation of the numerical domain. Through using a series of coupled and non-linear partial differential equations (PDEs), it considers the complex battery aspects and it offers highly precise insights into physical and chemical phenomena [127,129].

In particularly, the P2D model captures Li-ions transport through diffusion and migration processes in the solid particles (Eq. (6)) and inside the electrolyte (Eq. (5)), using Fick's law and the modified Nernst–Planck equations, respectively. For the electrochemical reactions at the solid-electrolyte interfaces, the model employs the Bulter–Volmer kinetics (Eq. (7)), which relates the exchange current density (Eq. (8)) to the electrode overpential (Eq. (9)). Table 4 summarizes the set of equations, based on the information from the following Refs. [127,130,131].

The strength of this model resides in its ability to smoothly combine the charge transport mechanisms with mass conservation principle. Moreover, It describes both electron transport in the solid phase (Eq. (11)) and ionic transport in the electrolyte (Eq. (12)), which are based on the Ohm's law and the concentrated solution theory [127, 130]. While this comprehensive approach provides highly accurate battery simulations, it demands significant computational requirements, lot of memory and processing power due to its sophisticated PDE system [23,127].

b. Doyle-Fuller-Newman (DFN)

The Doyle-Fuller-Newman model, which is based on the P2D model, got further advancement in the Li-ion battery modeling with improved accuracy and detailed representation of internal processes [102,132].

While the DFN and the P2D model shared the fundamental concept of layered structure, the DFN model incorporates additional intricacies to better capture battery dynamics. It involves in-depth descriptions of electrochemical reactions, using the same equations as P2D model to describe the ions diffusion and migration, Eqs. (5),

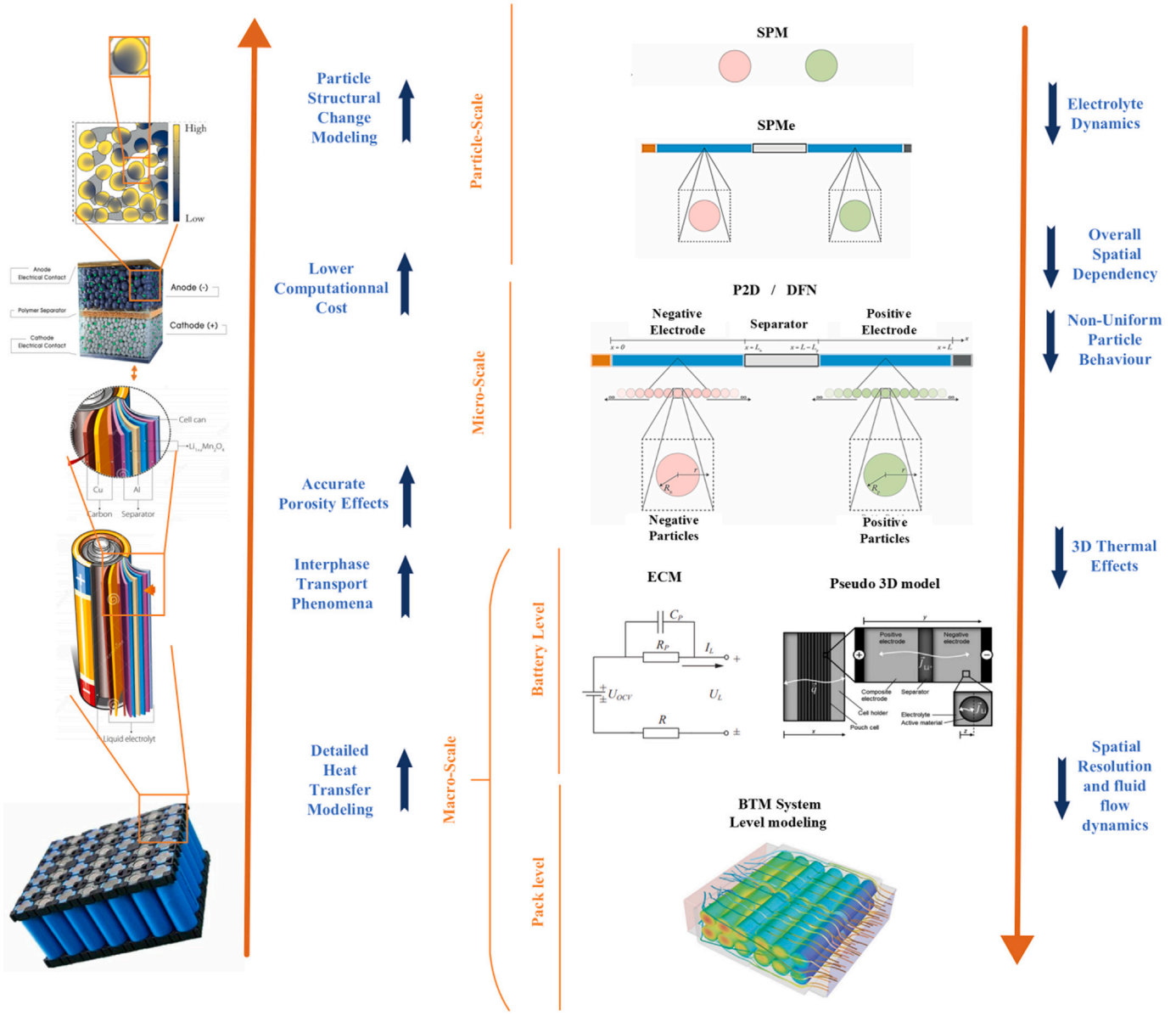


Fig. 17. Multi-scale battery modeling framework: from single particle to full cell dynamics.
Source: Adapted from [121–126].

(6) and (7). Moreover, further equations describe the potential distributions in the solid (Eq. (13)) and electrolyte (Eq. (14)) phases, the terminal voltage (Eq. (15)) and the maximum reversible capacity constrain (Eq. (16)). Table 5 gathers these equations, taken from the following references compilation [113,133,134]. The DFN framework provides a more accurate representation of the battery behavior by accounting for concentration-dependent variables like diffusivity and conductivity [135].

Although the complexity of the DFN model bridges gap between real world applications and theoretical knowledge, it requires even more computational resources for extensive analysis [134].

4.2.2. Particle scale model

While DFN and P2D models provide high accuracy through detailed parametrization, their complexity hide some physical understanding insights. Simpler model may provide better understanding of battery dynamics in scenarios when less detailed analysis is demanded. [136]. This trade-off between computational efficiency and model complexity leads to the development of the single particle model (SPM) [113].

a. Single Particle model (SPM) The key principle of the SPM is based on the idea of the high similarity behavior of the particles within each electrode, describing them by a single representative particle, as illustrated in Fig. 19. This model neglects the Li-ions concentration in the liquid phase. Its process offers high accuracy at a low current rate and improves the model calculation speed. Thus, it eliminates the need for a discretization of the numerical domain [113,121,137].

Moreover, in the SPM framework, the current distribution is assumed to be uniform along the thickness of the electrode and the electrolyte, resulting in high conductivity [138]. This assumption emphasizes the focus on: reaction kinetics at the electrode–electrolyte interface, transport of lithium-ions within the solid electrode particles, and solid-state phenomena while ignoring electrolyte properties [137, 139].

Table 4
Governing equations of the P2D model.

Electrochemical reaction	Governing equations	
Electrolyte phase Li-ion diffusion	$\varepsilon_e \frac{\partial c_e(x,t)}{\partial t} = \varepsilon_e \frac{\partial}{\partial x}\left(D_e^{eff} \frac{\partial c_e(x,t)}{\partial x}\right) + a\left(1-t_0^+\right) j_r(x,t)$	(5)
Boundary conditions	$\frac{\partial c_e}{\partial x}\Big\|_{x=0}$, $\frac{\partial c_e}{\partial x}\Big\|_{x=L}$	
Solid phase Li-ion diffusion	$\frac{\partial c_s(r,x,t)}{\partial t} = D_s \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial c_s(r,x,t)}{\partial r}\right)$	(6)
Boundary conditions	$\frac{\partial c_s}{\partial r}\Big\|_{r=0}$ $\frac{\partial c_s}{\partial r}\Big\|_{r=R_s}$	
The Butler-Volmer equation	$j_r(x,t) = \frac{i_0(x,t)}{F}\left(\exp\left(\frac{\alpha_a F}{\bar{R}T}\eta(x,t)\right) - \exp\left(\frac{\alpha_c F}{\bar{R}T}\eta(x,t)\right)\right)$	(7)
Exchange current density	$i_0(x,t) = F k_c^{\alpha_c} k_a^{\alpha_c} \left(c_{s,\max} - c_{s,surf}(x,t)\right)^{\alpha_a} \left(c_{s,surf}^{\alpha_c}(x,t)\right)\left(\frac{c_e(x,t)}{c_{e,ref}}\right)^{\alpha_a}$	(8)
Overpotential	$\eta(x,t) = \phi_s(x,t) - \phi_e(x,t) - E_{OCV}$	(9)
Charge conservation	$\nabla i_s + \nabla i_e = 0$	(10)
	$\nabla i_s = -a j_r$, $\nabla i_e = a j_r$	
Electron transport in solid phase	$i_s = -\sigma_s^{eff} \nabla \phi_s$	
	In other word, $\nabla(\sigma_s^{eff} \nabla \phi_s) - j_r = 0$	
	or $\frac{\partial}{\partial x}(\sigma_s^{eff} \frac{\partial \phi_s}{\partial x}) = j_r$	(11)
Boundary conditions	$\frac{\partial \phi_s}{\partial x}\Big\|_{x=l_n}$, $\frac{\partial \phi_s}{\partial x}\Big\|_{x=l_n+l_s}$	
Ionic transport of Li-ion in electrolyte phase	$i_e = -\sigma_e^{eff} \nabla \phi_e + \frac{2\bar{R}T\sigma_e^{eff}}{F}(1 + \frac{\partial \ln f_{\pm}}{\partial \ln c_1})(1-t^+)\nabla(\ln c_1)$	(12)
Boundary conditions	$\frac{\partial \phi_e}{\partial x}\Big\|_{x=0}$, $\frac{\partial \phi_e}{\partial x}\Big\|_{x=L}$	

Table 5
DFN additional equations with respect to P2D model.

Electrochemical reaction	Governing equations	
Solid-phase potential	$\frac{\partial}{\partial x}(\sigma \varepsilon_s \frac{\partial \phi_s}{\partial x}) = \frac{3\varepsilon_s F}{R_s} j_r$	(13)
Boundary conditions	$\frac{\partial \phi_s}{\partial x}\Big\|_{x=0}$, $\frac{\partial \phi_s}{\partial x}\Big\|_{x=L}$, $\frac{\partial \phi_s}{\partial x}\Big\|_{x=\delta_n}$, $\frac{\partial \phi_s}{\partial x}\Big\|_{x=L-\delta_p}$	
Electrolyte phase potential	$\frac{\partial}{\partial x}(\kappa \varepsilon_e \frac{\partial \phi_e}{\partial x} + K \varepsilon_e (t_+^0 - 1)\frac{2RT}{F}\frac{\partial \ln c_e}{x}) = -\frac{3\varepsilon_s F}{R_s} j_r$	(14)
Boundary conditions	$\frac{\partial \phi_e}{\partial x}\Big\|_{x=0}$, $\phi_e\Big\|_{x=L}$	
The terminal voltage	$V(t) = \phi_s(L,t) - \phi_s(0,t) - \frac{R_{cc}}{A} i_{app}(t)$	(15)
The maximum reversible capacity constraint	$Q = AF(\delta_i)\epsilon_i c_i^{max}(s_{i,100\%} - s_{i,0\%}), i \in \{n,p\}$	(16)

Therefore, at the particle level, SPM solves two main processes: electrochemical reactions and diffusion. The mass balance, in other words, the diffusion of Li-ions in the solid electrode particles (Eq. (17)), is represented using Fick's second law in a spherical coordinate system. The electrochemical reaction kinetics occur at the solid/electrolyte interface and are described by the Butler-Volmer equation (Eq. (19)). They relate the overpotential (Eq. (20)) with: the exchange current (Eq. (21)), the surface state of charge, and the temperature dependent reaction rate (Eq. (25)). In addition to these PDEs, the model represents an algebraic equation describing the potential difference between the cell terminals [139,140].

b. Single Particle model with electrolyte dynamics (SPMe)

Despite its simplicity, SPM has limitations in capturing the electrolyte dynamics effects on the battery performance. This restrictions brought researchers to address this issue and develop an extended version of the SPM, known as SPMe: the single particle model with electrolyte dynamics. SPMe offers better understanding of the liquid and solid phases interactions within the battery. It incorporates the diffusion of the electrolyte phase while maintaining the reduced computational complexity [141].

In the SPMe framework, the negative and positive electrodes are modeled with spherical particles surrounded by the electrolyte [141], as illustrated in Fig. 20.

Due to its simple representation, the SPMe shares the same equations described by the SPM (Eqs. (17), (19), (24)). Extending this concept a step further, this model captures the electrolyte and solid state phase transport phenomena. It incorporates an additional equation that describes the linear diffusion of the electrolyte (Eq. (18)), including constant current source terms [142,143]. The block diagram of the SPMe, see Fig. 21, shows the nonlinear function of the terminal voltage output, relating current, electrolyte state, and solid state concentrations boundary values [141].

SPMe provides a higher-advanced version of SPM despite being a simplified version of DFN. As they share the same simplified geometry, Brosa Planella et al. [144] developed a modeling approach, using PYBAMM, to compare these three models, DFN, SPM, and SPMe. This approach simulated a commercial battery with graphite-SiOx negative electrode and NMC 811 positive electrode at 2C discharge. This code compare the concentrations, potentials, and global states of the battery (current, discharge capacity, and voltage) using these three models, DFN, SPM, and SPMe. The results are represented in Fig. 22, demonstrating that the SPMe model closely captures DFN spatially distributed effects, while the lack of electrolyte dynamics in the SPM limits its voltage prediction, especially at high charge rates. This comparison highlights the importance of choosing appropriate model complexity based on specific accuracy requirements and computational constraints.

Table 6
Governing equations of SPM/SPMe models.

Electrochemical reaction	Governing equations	
Li-ion diffusion in solid electrode particles	$\frac{\partial c_s(r,t)}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}(D_s r^2 \frac{\partial c_s(r,t)}{\partial r})$	(17)
Boundary conditions	$\frac{\partial c_s}{\partial t}\Big\|_{x=0,t}$, $\frac{\partial c_s}{\partial t}\Big\|_{x=R_s,t}$	
Diffusion in the electrolyte	$\epsilon_k \frac{\partial c_e}{\partial t} = b_k D_e \frac{\partial^2 c_e}{\partial x^2} + \begin{cases} \frac{(1-t^+)I}{FL_n}, & k=n, \\ 0, & k=sep, \\ \frac{-(1-t^+)I}{FL_p}, & k=p, \end{cases}$	(18)
Boundary conditions	$\frac{\partial c_e^{\pm}}{\partial t}\Big\|_{x=0^+,t}$, $D_e^- \frac{\partial c_e^-}{\partial x}\Big\|_{x=L^-,t} = D_e^{\text{sep}} \frac{\partial c_e^{\text{sep}}}{\partial x}\Big\|_{x=0^{\text{sep}},t}$, $D_e^{\text{sep}} \frac{\partial c_e^{\text{sep}}}{\partial x}\Big\|_{x=L^{\text{sep}},t} = D_e^+ \frac{\partial c_e^+}{\partial x}\Big\|_{x=L^+,t}$	
Butler-Volmer equation	$j_r = k_i c_e^{-\alpha}(c_{s,\max} - c_{s,surf})^{1-\alpha}(c_{s,surf})^{\alpha}\left(\exp\left(\frac{(1-\alpha)F\eta_j}{\bar{R}T}\right) - \exp\left(\frac{-\alpha F\eta_j}{\bar{R}T}\right)\right)$	(19)
Overpotential at each electrode	$\eta_p = \frac{2\bar{R}T}{F}\sinh^{-1}\left(\frac{jr,p}{2i_{0,p}}\right)$, $\eta_n = \frac{2\bar{R}T}{F}\sinh^{-1}\left(\frac{j_r}{2i_{0,n}}\right)$	(20)
The exchange current	$i_0 = k(T)c_e^{-\alpha}(c_{s,\max} - c_{s,surf})^{1-\alpha} c_{s,surf}^{\alpha}$	(21)
Positive electrode potentials	$\theta_{s0,p}(t) = \frac{2\bar{R}T}{F}\sinh^{-1}(\frac{-I(t)}{2a_p L_p K_{eff}\sqrt{c_e^0 c_{ss,p}(t)(c_{s,max,p} - c_{s,surf,p}(t))}})$ $+E_{OCV}{}_{s_{p,100\%}} - \frac{R_{SEI,p}I(t)}{2a_p L_p}$	(22)
Negative electrode potentials	$\theta_{s0,n}(t) = \frac{2\bar{R}T}{F}\sinh^{-1}(\frac{-I(t)}{2a_n L_n K_{eff}\sqrt{c_e^0 c_{ss,n}(t)(c_{s,max,n} - c_{s,surf,n}(t))}})$ $+E_{OCV}{}_{s_{n,100\%}} - \frac{R_{SEI,n}I(t)}{2a_n L_n}$	(23)
Potential difference between cell terminals	$V(t) = \phi_s(L,t) - \phi_s(0,t) - \frac{R_{cc}}{A} i_{app}(t)$	(24)
Temperature dependent reaction rate	$k_i(T) = k_{0,i}\exp\left(\frac{E_{a,i}}{\bar{R}}\left(\frac{1}{T_{\text{ref}}} - \frac{1}{T}\right)\right)$	(25)

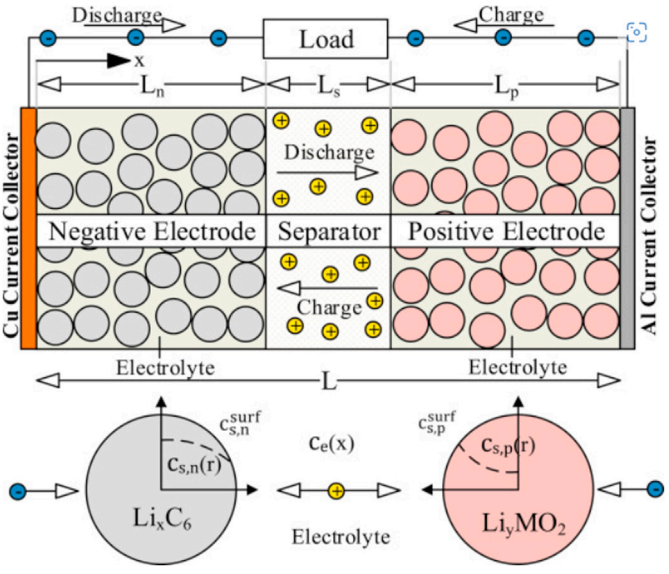


Fig. 18. Schematic representation of the P2D model domain, from [23].

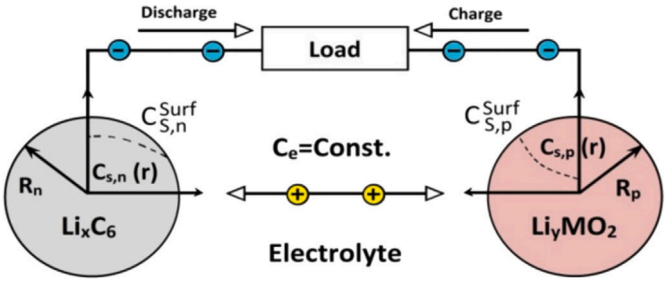


Fig. 19. Schematic representation of the Single Particle Model (SPM) for a Li-ion cell, from [140].

4.2.3. Battery scale model

Battery scale modeling provides integral insights into the overall dynamic behavior of complete battery systems. At this level, the Equivalent Circuit Model (ECM) is widely used, representing the electrochemical processes through electrical components such as voltage sources, capacitors, resistance-capacitance (RC) networks, and resistors. This approach forms a circuit network and models the Li-ion battery as an electrical energy storage device [127].

Model parameters are primary identified by Hybrid Pulse Power Characterization (HPPC) testing, an industry-standard characterization technique fundamental to ECM development. During HPPC test, controlled direct current (DC) pulses are applied, i.e., discharge pulse followed by a relaxation then a charge pulse followed by another relaxation. This testing evaluates the battery systems and cells by determine key parameters including internal resistance, polarization effects, and capacitance values. It creates a characteristic voltage response patterns, revealing battery performance and behavior throughout its lifecycle. Through analysis of the test responses, ECM enables to effectively captures the input–output battery dynamic behavior and charge discharge characterization without complex electrochemical modeling examination [146].

Various ECM models have been developed: the Rint model, the resistance capacitance model, the Thevenin model, and the PNGV (Partnership for New Generation Vehicle) model, each of them has unique features and representations, describing the battery current and voltage [123,147,148].

The fundamental ECM form is the Rint model (Fig. 23(a)), representing the internal resistance. It offers a simple series of resistors and treat the battery an ideal voltage source.

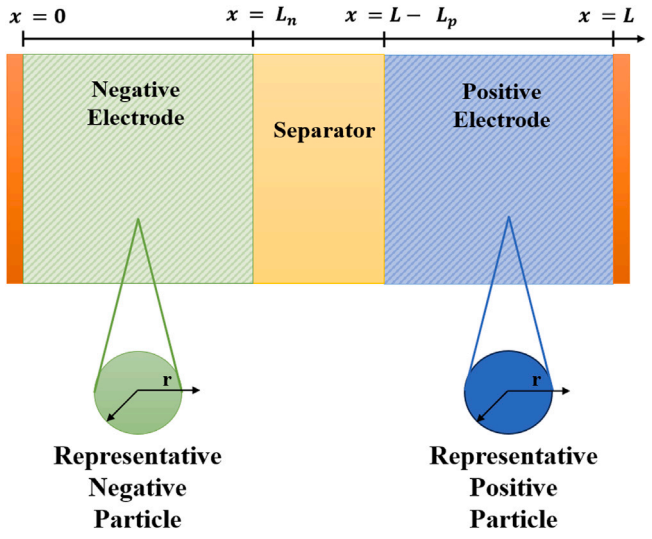


Fig. 20. Geometry of the SPMe model with the representation of only one particle at each electrode.

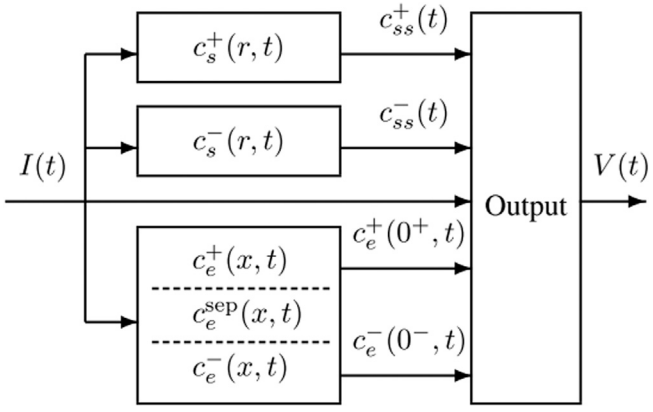


Fig. 21. Block diagram of the SPMe [141].

The resistance capacitance model, also known as the RC model (Fig. 23(b)), developed by the SAFT battery company, better approximates real battery behavior by using capacitors within circuit branches. Its second-order RC, designed with two RC pairs enables detailed simulation under diverse load conditions.

Moreover, the Thevenin model (Fig. 23(c)) is another form of ECM and the most popular time-domain model among the others. It is built on the Rint model by adding a parallel RC network in series and taking into account the polarization effects. In addition, a modified version of the Thevenin model has been developed and it is known as the PNGV (Fig. 23(d)). This model incorporates a slight increase in circuit elements adding in series a capacitor $1/U'_{oc}$ to improve the model fidelity. The basic governing equations for those models are summarized in Table 7.

In order to accurately estimate the battery states such as state of charge (SOC) and state of health (SOH), the ECM is required due to its accurate simulations of battery power output.

4.2.4. Integration of thermal model

The electrochemical and electrical models, such as DFN and ECM, while effectively predict battery performance through external and internal characteristics, they are not yet capable of fully answering the multi-scale challenges. In particular, they fail to capture the heat transfer phenomena and thermal characteristics that affect electrochemical reaction and Li-ion transport [19]. Tanaka et al. [149] established a link between the temperature, T, and the battery ionic transport and kinetics parameters. With this aim, Tanaka et al. introduced a function Ψ in form of an Arrhenius relation:

$$\Psi = \Psi_{\text{ref}} \exp\left(\frac{E_a(\Psi)}{\bar{R}}\left(\frac{1}{T_{\text{ref}}} - \frac{1}{T}\right)\right) \tag{34}$$

Katrašnik et al. [150] provided valuable insights to this relation into the Li-ion battery thermal dynamics by building a multi-scale modeling framework, summarized in Fig. 24. This model links the cathode crystallographic structure, heat generation, electrode topology, and thermal runaway.

These limitations have driven the development of advanced numerical simulations for an overall comprehensive understanding of thermal behavior. Wang et al. [151] and Xu et al. [152] proposed a 3D electrochemical thermal model to predict cell temperature profiles. Jaguemont et al. [153] utilized a fully coupled electrochemical-thermal model for extreme temperature conditions using a single pair of large LiFPO4 pouch cell layer. Goutam et al. [154] took a different approach, representing a three-dimensional electro-thermal model. It combines a multilayer 2D potential distribution model with 3D temperature analysis to study the effects of design factors on cell temperature distribution.

Therefore, to appropriately identify the different thermal and electrochemical coupling approaches, three main models are presented to describe the battery internal heat generation: the Bernardi model [155,156], the electro-thermal coupled model (ETC) [157], and the electrochemical-thermal coupled (ECTC) model [19,157]. The following sections will offer an in-depth examination of the aforementioned models.

a. Bernardi model

The Bernardi model, effectively estimates battery pack heat generation without detailed electrochemical analysis, making it valuable in optimizing battery thermal management systems (BTMS) [156]. To reduce the inherent complex processes in heat generation, Yang et al. [158] computed heat by Bernardi model and reveal the significant high accuracy of this model. Jindal et al. [159] and Xu et al. [160] employed the Bernardi model based on various thermal and electrical parameters to evaluate and predict the heat generation rate, Q. The latter is described in the following equation:

$$\mathrm{Q} = -\left(\mathrm{Q}_r + \mathrm{Q}_i\right) = -IT\frac{\mathrm{d}E_{OCV}}{\mathrm{d}T} + I\left(E_{OCV} - U_{app}\right) \tag{35}$$

including the irreversible heat Q_i, generated from ohmic loss and activation and polarization losses, and the reversible heat Q_r, produced during intercalation and deintercalation processes and associated with the battery material entropy coefficient by dE_{OCV}/dT. Furthermore, replacing when needed $(E_{OCV} - U_{app})$ by the product of the squared current intensity I^2 with the ohmic internal resistance R.

These studies validated thermal models by measuring temperature, current, overpotential, and entropic coefficients under realistic driving conditions, such as continuous and pulse discharge protocols [159]. Zhai et al. [161] conducted extensive testing of 18650 LFP cells, including discharge temperature rise tests, discharge rate, open-circuit voltage (OCV) tests. Using the Bernardi model, they analyzed the heat generation rate (HGR), under a drive cycle with different heat-transfer coefficients and ambient temperatures, and accurately simulated the thermal runaway.

b. Electro-thermal models

As well, electro-thermal models predict heat generation by incorporating heat radiation, heat propagation delay, and various thermal–electrical parameters. Barcellona et al. [162] developed a highly accurate electro-thermal model that predict heat dissipation-generation based on the battery current exchanged, achieving temperature prediction within 2 °C of experimental measurements. Mesbahi et al. [163] effectively simulate the interaction between the thermal and electrical battery behavior by proposing a coupled electro-thermal model for

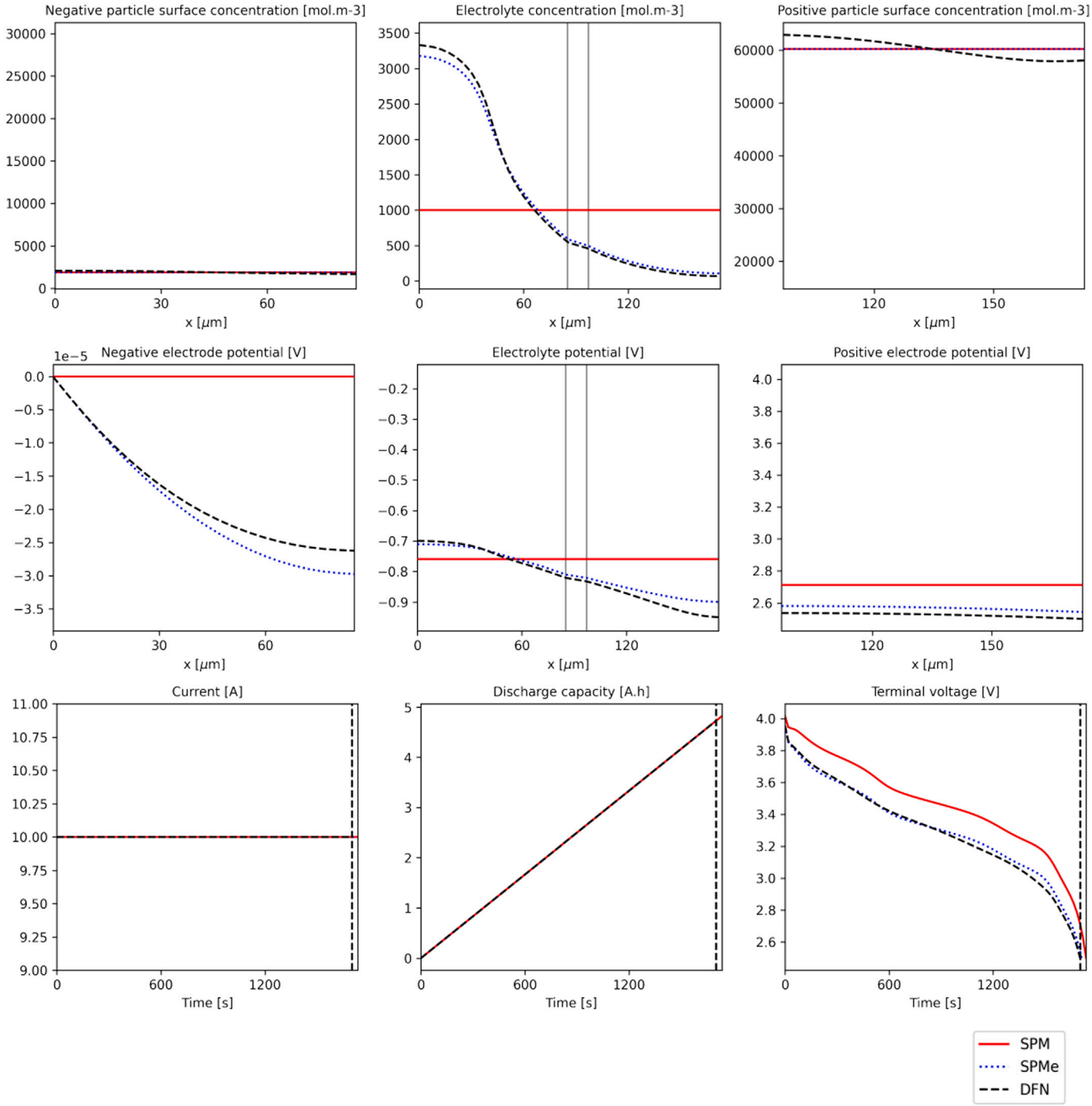


Fig. 22. Comparison between the Doyle Fuller-Newman model (DFN), the Single Particle Model (SPM) and the Single Particle Model with electrolyte (SPMe) [144], for a 2C discharge simulation using the data from [145].

a 40 Ah LIB cell, capturing both high-power dynamics and internal physicochemical phenomena.

Besides, Li et al. [164] combined a second-order RC equivalent circuit model with a thermal model to optimize charging strategy. Their five-stage CC charging method reduced peak temperature by 0.83% and charging time of 13.87%, compared to the traditional 0.5C CC-CV (Constant Current-Constant Voltage) charging methods. Similarly, Naguib et al. [165] coupled an ECM with a lumped model to effectively predict pouch cell behavior, under a temperature range between −20 °C and 25 °C and real-world drive cycles,

However, these previous models, while providing valuable insights, ignore, at some point, various complex chemical interactions occurring inside the battery. Recognizing the operations of these batteries as multidomains, multiphysics, and multiscales issues, many studies have been developed to integrate both thermal and electrochemical aspects.

c. Electrochemical thermal coupled (ECTC) P3D model

Xu et al. [166], Maheshwari et al. [167], Carelli et al. [122] and Lin et al. [168] developed an electrochemical thermal coupled (ECTC) model. These models combined a pseudo-2D electrochemical (P2D) model with thermal analysis and create a pseudo-three-dimensional (P3D) framework that simulates charging discharging profiles, lithium plating, and SEI degradation mechanism, under various conditions.

Table 7
Governing equations for the different ECM models.

ECM models	Governing equations	
Rint Model	$U_L = U_{OCV} - I_L R_o$	(26)
RC Networks	$\begin{bmatrix}\dot{U}_b\\ \dot{U}_c\end{bmatrix} = \begin{bmatrix}\frac{-1}{C_b(R_E+R_c)} & \frac{1}{C_b(R_E+R_c)}\\ \frac{1}{C_c(R_E+R_c)} & \frac{-1}{C_c(R_E+R_c)}\end{bmatrix}\begin{bmatrix}U_b\\ U_c\end{bmatrix} + \begin{bmatrix}\frac{-R_c}{C_b(R_E+R_c)}\\ \frac{-R_E}{C_c(R_E+R_c)}\end{bmatrix}[I_L]$	(27)
	$[U_L] = \begin{bmatrix}\frac{R_c}{R_E+R_c} & \frac{R_E}{R_E+R_c}\end{bmatrix}\begin{bmatrix}U_b\\ U_c\end{bmatrix} - \left[R_t + \frac{R_E R_c}{R_E+R_c}\right][I_L]$	(28)
Thevenin Model	$\dot{U}_{Th} = -\frac{U_{Th}}{R_{Th}+C_{Th}} + \frac{I_L}{C_{Th}}$	(29)
	$U_L = U_{OC} - U_{Th} - I_L R_0$	(30)
PNGV model	$\dot{U}_d = U'_{OCV} I_L$	(31)
	$\dot{U}_{PN} = -\frac{U_{PN}}{R_{PN}+C_{PN}} + \frac{I_L}{C_{PN}}$	(32)
	$U_L = U_{OCV} - U_d - U_{PN} - I_L R_0$	(33)

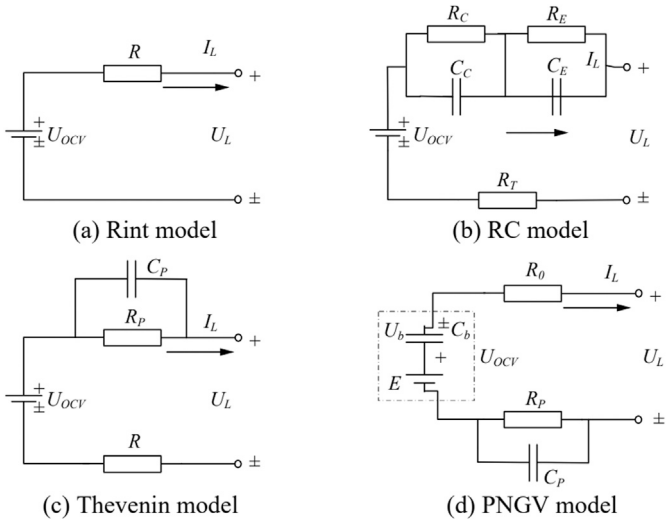


Fig. 23. Various equivalent circuit model (ECM) layouts [123].

P3D accurately predicts temperature and voltage by capturing complex electrochemical reactions and multiscale mass and heat transport phenomena, as validated against both literature and experimental data [169,170].

This consist of 3D (conceived as 1D + 1D + 1D) domain, illustrated in Fig. 25, integrating and focusing on various phenomena occurring at different scales. P3D model capturs 3D phenomena at different scales. At the *x-scale*, diffusion kinetics in the active material particles. At the *y-scale*, both electrochemical reactions at the electrode/electrolyte interface and Li-ion transport in electrolyte and solid particles. These two scales are described in detailed in the P2D model, Table 4. In addition, at the *z-scale*, P3D model captures heat transport phenomena through the cell thickness, including temperature distribution across the cell and thermal management system effects. Three heat generation source terms are involved in the energy balance of this thermal model [131, 169], reversible reaction heat, irreversible polarization heat, and ohmic heat generation, as summarized in Table 8. The reversible reaction heat (Eq. (36)), estimates heat generated from electrochemical reactions and proportional to entropy change. The irreversible polarization heat generation (Eq. (37)), handles the heat produced due to the internal resistances, overpotentials, or voltage losses. The ohmic heat generation (Eq. (38)), addresses the heat produced from the electrical resistance in the current collectors, electrodes and conductive materials. The total heat combines all these terms. Additional components may be included based on study requirements, such as environment heat dissipation, conjugate heat transfer, and the temperature-dependent reaction rates.

4.2.5. Pack level model

The risks associated with temperature peaks and the heat generation inside a cell (thermal runaway, see Fig. 13, highlights the importance of introducing a Battery Thermal Management Systems (BTMS). These systems are employed to maintain a uniform temperature both within individual cells and across the battery pack by controlling heat dissipation and operating temperature [175].

a. Types of battery thermal management systems

The current state-of-the-arts in BTMSs presents a broad spectrum of solutions [176]: air cooling [177], indirect and direct liquid cooling [178,179], PCM cooling [180], heat pipe cooling [181], spray cooling [182], and hybrid cooling [183,184]. Cooling systems differ primarily in their coolant medium, cost, weight, efficiency, and complexity. Thus, the selection depends on applications requirements and operating conditions, considering factors such as heat transfer rate, thermal conductivity, construction simplicity, space utilization, and temperature uniformity [185].

Although various cooling methods are widely used, each has specific limitations.

Air cooling, while simple, light, and cost-effective, offers low and insufficient thermal conductivity characteristics and non-uniform temperature distribution, especially during TR events [175,186].

Indirect liquid through cooling plates and heat-pipe cooling presents additional thermal resistance from multiple interface layers with high thermal conductivity, high heat capacity, and efficient heat transfer. However, their complex additional system equipment requires more weight, cost and power beside the small contact area and the risks of the leakage problems at connection points.

Likewise, PCM-based cooling systems, despite their low cost and large latent heat, have low thermal conductivity, thermal stability, and phase change temperature limitations.

These limitations drive ongoing research to optimize thermal management, particularly for preventing TR [187].

Immersion cooling addressed these challenges by demonstrating superior thermal capabilities, ensuring uniform cooling through the cell-liquid interface with superior heat transfer efficiency and thermal conductivity, beside the large contact area compared to other cooling systems. This latter system category should be designed carefully, by featuring a homogeneous flow of coolant in the pack to ensure a proper temperature uniformity, as demonstrated in [188]. While the immersion cooling fluid requires careful consideration and system complexity, its superior thermal performance proved to be promising for next-generation high-power battery applications [189–191].

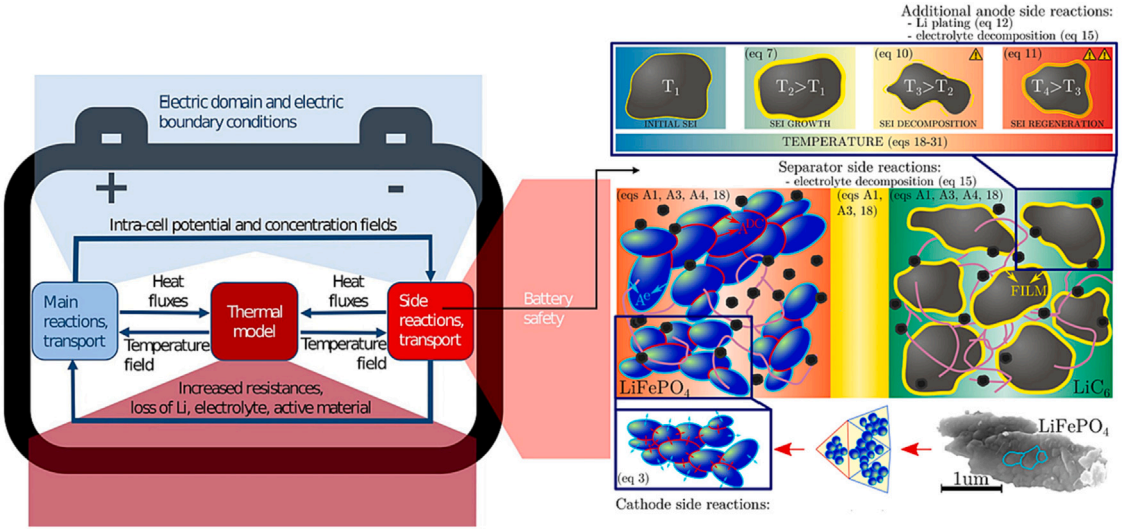


Fig. 24. Representation of the model integrating electrochemical, side reactions and thermal interactions in the multi-scale modeling.
Source: Adapted from [19,150].

Table 8
Governing equations for heat generation and energy balance in electrochemical thermal P3D model.

Heat reaction	Governing equations	
Reversible reaction heat generation	$Q_{\text{Rea}} = a j_r T \frac{\partial E_{OCV}}{\partial T}$	(36)
Irreversible polarization heat generation	$Q_{\text{Pol}} = a j_r \eta$	(37)
Ohmic heat generation	$Q_{\text{ohm}} = -i_e \cdot \nabla \phi_e - i_s \cdot \nabla \phi_s$	(38)
Total heat generation	$Q_{\text{Tot}} = Q_{\text{Rea}} + Q_{\text{Pol}} + Q_{\text{ohm}}$	(39)
Energy balance	$\rho C_p \frac{\partial T}{\partial t} - \lambda \nabla^2 T = Q_{\text{Rea}} + Q_{\text{Pol}} + Q_{\text{ohm}}$	(40)

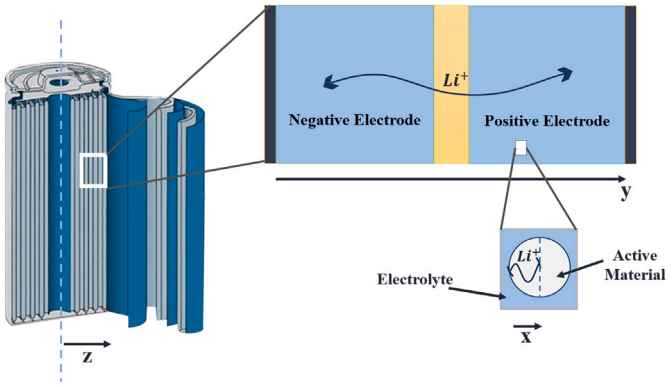


Fig. 25. 3D domain illustrating the different scales (x, y and z) of an electrochemical-thermal P3D model.
Source: Adapted from [171].

Furthermore, immersion cooling considers submerging directly the battery cells in a non-conductive dielectric fluid, providing full contact with the cell, and creating a dynamic heat transfer system [192,193], see Fig. 26. As the fluid circulates, it absorbs heat and the latter is transferred it either to a secondary cooling system such as chiller or radiator [194]. Immersion cooling fluids, such as silicone oil [195], volatile solvents [196], hydrofluoroethers [175], and hydrocarbons (mineral oil) [197], are preferred to be electrically insulating and non-flammable [175]. In order to guarantee thermal stability and cooling effectiveness, large quantities of fluids with low viscosity and low weight are required [192,194].

Given these advantages and widespread implementation in high-performance applications, this review focus on further numerical modeling analyses of immersion cooling systems. In particular, authors provide detailed examination of the fundamental thermal-fluid dynamics and heat transfer mechanism at the solid–fluid interface.

b. Numerical modeling of fluid flow and heat transfer in BTMS

The following references present some numerical models applied to immersion cooling BTMS. Wu et al. [199] developed a numerical model of a BTMS using silicone oil as a coolant medium that safely prevented the TR propagation. Similarly, Hussain et al. [200] analyzed a channeled dielectric fluid immersion by limiting the non-uniformity in the battery pack to 1 K and its maximum temperature to 302 K through evaluating different parameters like battery aging, gradeability, and aggressive drive cycle. Solai et al. [201] and Ahmad et al. [192] proposed numerical models which underlie the fundamentals of the transient conjugate heat transfer mechanisms occurring in immersion cooling method. These analyses take into account the heat transfer as follows:

- In the Li-ion solid battery components, such as electrode separator current collectors, the heat source term q_g represents ohmic heat and entropic heat. An expression involving internal resistance and electrical current is usually used to represent the electrical heating processes. The internal resistance model can be temperature and SOC dependent and it is often fitted from experimental data. The electrical current can be considered as function of time or SOC, depending on the charge/discharge profiles under study. The source term q_g at battery scale, is included in the heat equation as follows to compute the temperature evolution within the battery:

$$\frac{\partial T}{\partial t} = \nabla \cdot (\alpha_s \nabla T) + \frac{q_g}{\rho_s C_{p,s}} \tag{41}$$

Table 9
Summarized overview of the multiscale battery models used in literature.

Models	Scale of modeling	Key features	Advantages	Limitations	Applications	Literature	Software used in literature
Pseudo 2D (P2D) model	Micro scale	Detailed Electrochemical reactions, Spatial resolution/variations in electrode-separator-electrolyte domain as well as active particles	High accuracy, Deep comprehensive understanding of the internal process	Computationally intensive	Detailed battery design, Academic and real world research simulations	[129,145,172]	ANSYS Fluent, COMSOL Multiphysics, PYBAMM
Doyle Fuller Newman (DFN) model	Micro scale	Enhanced and more detailed P2D model, Improved accuracy in with Solid-Electrolyte potentials and electrolyte dynamics	Highly precise insights of the internal representation, Higher resolution in dynamic simulations	Complex for real time scenarios, Higher computational requirements	Detailed performance estimations and analysis, Advanced Research	[17,112,133]	MATLAB, PYBAMM
Single Particle Model (SPM)	Particle Scale	Focus on single representative particle	Computationally efficient	Neglects electrolyte dynamics, Less accurate with high currents	Simple control systems, Basic requirements	[17,112,138,139]	MATLAB, PYBAMM
SPMe	Particle Scale	SPM with electrolyte dynamics, More accurate than SPM and Less than DFN/P2D	Balancing simplicity with accuracy, Captures electrolyte effects	Can miss some multidimensional effects	Efficient performance estimation	[17,112,142,143]	MATLAB, PYBAMM
Equivalent Circuit Model (ECM)	Battery level	Represents the battery as electrical circuit, Many Complexity levels are available	Simple implementation techniques, Enhanced fidelity and power prediction	Limited chemical and physical insights, Possible lacks with dynamic conditions	State prediction in BTM	[173]	MATLAB
Pseudo 3D (P3D) model	Battery level	Combining Thermal and electrochemical models, Account for temperature effects, and capture heat transfer phenomena	Whole cell performance predictions, Captures electrochemical and thermal interactions	High computational cost, Increased complexity	Battery thermal management systems, Safety analysis	[174]	COMSOL Multiphysics

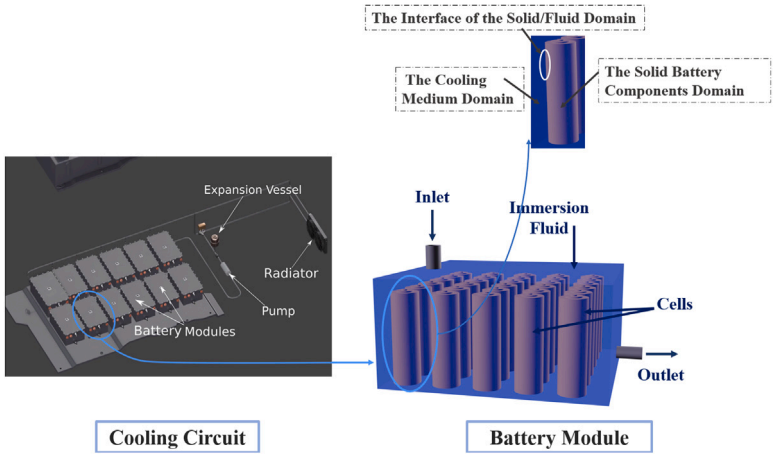


Fig. 26. Schematic diagram of direct immersion liquid cooling system mechanism integrating the overall cooling circuit (adapted from [198]), with a detailed view of the battery module and numerical cooling domains.

where T, $\alpha_s = \frac{\lambda_s}{\rho_s C_{p,s}}$, q_g, ρ_s, and $C_{p,s}$ are the temperature field, thermal diffusivity, heat source term, pressure field, and specific heat, respectively, in the solid domain.

- In the fluid medium domain, the Navier–Stokes equations capture and solve the interaction between the heat transfer and the fluid dynamics. These equations, including continuity, momentum, and energy equations, can be summarized as follows:

$$\begin{cases} \nabla \cdot \mathbf{u} = 0 \\ \dfrac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = \nabla \cdot (\nu_f \nabla \mathbf{u}) - \dfrac{1}{\rho_f} \nabla p \\ \dfrac{\partial T}{\partial t} + \mathbf{u} \nabla T = \nabla \cdot (\alpha_f \nabla T) \end{cases} \tag{42}$$

where $\mathbf{u}$, t, ν_f, ρ_f, p, T, and $\alpha_f = \frac{\lambda_f}{\rho_f C_{p,f}}$ are the velocity field, time, kinematic viscosity, density, pressure field, temperature field, and thermal diffusivity, respectively, in the fluid domain.

Basically, the heat generated from the internal battery domain is conducted to the battery surface through its materials. Then at the surface, by convection, this heat is dissipated through the fluid domain.

At the interface of the solid/fluid domain lies the thermally coupled boundary conditions of the heat flux and temperature continuity in most of the cases. Thermal resistance can be added at solid/liquid interface to simulate realistic battery casing and component layers. However, these two domains in the CFD simulations are meshed separately.

While BTMSs have already achieved significant improvement in ensuring optimal performance, challenges still remain. Therefore, it is necessary to adopt approaches that combine thermal, electrochemical, and electrical models with advanced BTMS. This multifaceted strategy allows a deeper understanding of Li-ion batteries behavior.

5. Perspectives and future directions

Beyond the physics-based and thermal approaches, recent innovations in artificial intelligence (AI) and machine learning have unlocked new possibilities and expanded capabilities in battery modeling to complement the existing methodologies. While the physics-based and thermal models provide a detailed understanding of the internal electrochemical and thermal battery behavior, data-driven approaches offer promising capacities in capturing the intricate non-linear real-world phenomena. These approaches accurately improve the model efficiency by combining experimental measurements with operational data [127, 201]. However, data-driven and machine learning approaches require a high quality of large dataset with careful consideration regarding the model architecture and hyperparameters [202].

Authors strongly believe that AI and data-driven models can be used to couple the macroscale experimental data obtained from in-situ battery measurements and to enhance the model predictivity at smaller scales.

Hard-to-measure parameters like state of charge, state of health, or internal resistance, could be assessed through AI, accounting for uncertainties to improve model predictions. The coupling of experimental data at macroscale and robust numerical predictions could close the scale gap between electro-chemical and BTMS scales.

Furthermore, via integrating fundamental theoretical science in practical engineering analysis, advanced optimization techniques are needed to develop more sophisticated adaptive models. Further works should focus on developing models that can effectively handle extreme conditions and abuse scenarios while managing computational demands for high-fidelity simulations.

Several other concerns affect new generations of batteries. Solid Li-ion batteries receive considerable attention in the EV industry sector [203]. These systems require significant efforts to develop accurate models at electro-chemical scales. Solid batteries tends to be also sensitive to several types of swelling, and new models should be developed to encompass this mechanical phenomenon to predict the potential impact of the thermal performances at pack scale.

6. Conclusions

This comprehensive review examines the sophisticated framework of Li-ion battery modeling. It deals with various topics from particle-level to pack-level analysis, providing in-depth insights into the battery performance and degradation. Overall, this article presents models bridging the gap between microscale phenomena and macroscale behavior.

Over cycles, Lithium-ion batteries experience various multifaced degradation mechanisms, which pose major safety concerns. These range from SEI growth and material loss, to mechanical stress and structural deterioration. Furthermore, temperature variations significantly affect battery behavior by impacting ion transport and reaction rates. These thermal effects lead to non-uniform aging and safety issues, especially in promoting thermal runaway. The review highlights the possible advantages of integrating the degradation processes and temperature effects into comprehensive multi-physics models.

The presented models integrate one, two and three-dimensional approaches across multiple scales. They combine electrochemical, thermal, and mechanical interactions to estimate the battery behavior. At the microscale the pseudo-2D (P2D) and the Doyle–Fuller–Newman (DFN) models provide detailed insights into the electrochemical reactions and internal battery states. These models capture the spatial variations along the thickness of the electrode, separator, and active particles. Although the DFN model requires higher computational cost than the P2D model, it offers a more accurate analysis of the internal dynamics.

The particle scale, on the other hand, includes the single particle model (SPM) with and without electrolyte dynamics. The SPM offers a balance between simplicity and accuracy. Its extended version, SPMe, incorporates electrolyte diffusion, without the full complexity of the microscale models, while maintaining simplicity. This makes it suitable for rapid control applications, without the complexity of the microscale models.

At the battery scale, the equivalent circuit model (ECM) captures the electrical behavior using various configurations. These include the Rint model, the resistance capacitance model, the Thevenin model, and the PNGV model.

Safety concern about uncontrolled temperature and thermal runaway underscores the need for improved numerical analyses. Various thermal models have been developed to address these issues, including the Bernardi model, electro-chemical models, and electrochemical-thermal approaches such as the pseudo-3D model. They successfully illustrate the complex relationship between temperature gradients, heat generation, mechanical stress and current distribution.

Moreover, at the pack level, this review highlights the importance of battery thermal management systems. Among all cooling techniques, the operation of direct immersion cooling is explained in more depth, as this method has proven valuable in practical real-world applications. Special attention is given to models as immersion cooling, which effectively simulates the interaction of heat transfer and fluid dynamics between solid and fluid domains.

Overall, this work provides a comprehensive framework for comparing various modeling approaches, by giving a clear view of physics solved at each scale. These findings highlight the key challenge of balancing predictive accuracy, computational efficiency and model complexity.

CRediT authorship contribution statement

Magui Mama: Writing – original draft, Methodology, Data curation, Conceptualization. **Elie Solai:** Writing – review & editing, Supervision, Conceptualization. **Tommaso Capurso:** Writing – review & editing, Formal analysis, Conceptualization. **Amelie Danlos:** Writing – review & editing. **Sofiane Khelladi:** Writing – review & editing.

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.

Data availability

Data will be made available on request.

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