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Designed high-performance lithium-ion battery electrodes using a novel hybrid model-data driven approach



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ABSTRACT

Lithium-ion batteries (LIBs) have been widely recognized as the most promising energy storage technology due to their favorable power and energy densities for applications in electric vehicles (EVs) and other related functions. However, further improvements are needed which are underpinned by advances in conventional electrode designs. This paper reviews conventional and emerging electrode designs, including conventional LIB electrode modification techniques and electrode design for next-generation energy devices. Thick electrode designs with low tortuosity are the most conventional approach for energy density improvement. Chemistries such as lithiumsulfur, lithium-air and solid-state batteries show great potential, yet many challenges remain. Microscale structural modelling and macroscale functional modelling methods underpin much of the electrode design work and these efforts are summarized here. More importantly, this paper presents a novel framework for next-generation electrode design termed: Cyber Hierarchy And Interactional Network based Multiscale Electrode Design (CHAIN-MED), a hybrid solution combining model-based and data-driven techniques for optimal electrode design, which significantly shortens the development cycle. This review, therefore, provides novel insights into combining existing design approaches with multiscale models and machine learning techniques for next-generation LIB electrodes.

1. Introduction

Lithium-ion batteries (LIBs) have been widely applied in many industrial fields such as consumer electronics, electric vehicles(EVs) and grid scale energy storage, providing tremendous benefits for modern society [1-7]. However, despite almost 30 years of research and development, the fundamental design of LIBs has remained largely unchanged, and is gradually approaching a plateau [8]. These conventional designs mostly consist of transition metal oxide cathodes and graphite anodes. Therefore, industrial and academic fields are now seeking high-performance batteries with higher energy density, better rate performance and longer lifespan, leading to various emerging battery technologies such as lithium-sulfur (Li-S), lithium-air (Li-Air) and solidstate batteries. However, crucial challenges still exist before their large scale commercialization is possible [9-13].

Electrodes play a crucial role in defining the performance of the battery. An ideal electrode requires several criteria including high areal capacity, rate capability and cycle life. However, enhancing one criterion often comes at the cost of another; improvements in energy density and rate performance, for instance, often contradict each other as highlighted in Fig. 1. Electrode energy density can be increased by adopting thick electrode designs, emerging materials and proper cell-to-pack design, so that more energy can be stored per unit volume to enable lightweight design for vehicles. These thick electrode designs offer a promising route to increase the energy density as they remove electrochemically inactive components in the cell. However, at the same time, fast charging techniques are severely restricted by the increased diffusion distances in these thick electrodes, leading to increased resistance and uneven lithiation. Unfortunately, the cycle life generally decreases at higher charging rate, due to the decreased overpotential of side reactions and the increased likelihood of lithium plating, and whilst high surface area anodes can be used, this generally increases the rate of parasitic side reactions such as the growth of the solid-electrolyte interphase (SEI) laver.

Fig. 1 also illustrates how the energy density increases with increased thickness before decreasing after a certain point. The rate performance,

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Fig. 1. Schematic illustration of the trade-off between energy density and rate performance of electrodes. Emerging materials. Reproduced with permission from Wiley [112]. Novel battery chemistries. Reproduced with permission from Wiley [140].

however, continually decreases as the electrode thickness increases. This relationship between thickness and rate-capability, therefore, forms an optimal region (marked in blue) in the trade-off between energy density and rate performance. However, this is still an over simplified representation of the optimization of electrode thickness, which requires many simulations and experiments which factor in a wider set of parameters, such as lifetime for example. This traditional research process, therefore, suffers from long and in turn costly development times.

In order to achieve high energy density electrodes, there are two fundamental approaches. As discussed, one is increasing the proportion of active material by increasing electrode thickening, while the other is to apply emerging battery chemistries such as Li-Air, Li-S and Li-metal anodes. Recent researches have illustrated that the energy density of current LIBs can be increased up to 300-350 Wh/kg in the short term by using high-nickel cathodes and thicker electrode [14–19]. Thick electrode design approaches, including 3D conductive percolation networks, low tortuosity designs and other promising methods, can substantially decrease the inactive material ratio to increase active material loading, resulting in higher energy density [20]. However, the energy density boost that these conventional approaches bring reach a limit with the increasing thickness of electrode, where increasing thickness proportionally increases the charge transport distance and resistance to the motion of ions and electrons, leading to poor electrode rate performance [21]. Therefore, considerable effort has been committed to improving the alternative strategies for low-tortuosity electrode fabrication, including the use of additive manufacturing and templating approaches, which possess a low tortuosity to promote the ion and electron transport. However, these complicated electrodes come with a higher manufacturing cost, making it much more difficult to be widely applied in industry [20].

The safety of the battery system is also a major concern. As the practical demands on LIBs grow, so does the need for fast charging. However, this comes with associated risks of dendrite formation and internal short circuiting (ISC) of the battery. At present, there are various standards for fast charging depending on the regions and applications [22]. The Ministry of Industry and Information Technology of the People's Republic of China, for instance, define fast charging as a 30 minute charge from 0 to at least 60% of the whole battery capacity [23]. Compared with conventional slow charging, which is normally defined as 5h to 8h charging (0-100%SOC), the charge rate of fast charging needs to be at least 2C to meet the demands for EVs (30-80%SOC within half an hour). Aspirational work aims to shorten this to less than 5 min in the future, which means at least a charging rate of 12 C is required. Here, many researches have reviewed the fast charging literature, including detailed discussions around Li plating, aging mechanisms and charging protocols [24–27]. For conventional electrodes, the thickness is usually under 100 μ m in order to access most of the capacity, avoid Li plating and retain a sufficiently acceptable lifetime. Generally, a 20 mg cm^{-2} mass loading, resulting in a 7 mAh cm⁻² areal loading, is recommended, with lower loadings suggested for fast charging cells. In addition, a current density limit of 4 mA cm^{-2} is recommended to avoid these issues [28].

Research interest in high energy density electrode architectures is high, but requires consideration of many different factors which has led to slow development. This paper, therefore, reviews previous research on the optimization of electrochemical performance for electrodes using conventional electrode design strategies and emerging techniques for next-generation energy devices. A review of the corresponding modelling activities then gives a perspective on how digital tools can assist in this effort. After that, a review of efforts towards combining modelbased and data-driven techniques is presented, and a framework named Cyber Hierarchy And Interactional Network based Multiscale Electrode Design (CHAIN-MED) is presented. This offers a hybrid framework, combining model and data driven solutions, with cloud-based optimization for next-generation electrode design. Finally, a summary and future perspectives for novel electrode designs are highlighted.

2. Electrode Designs for Conventional and Emerging Battery Chemistries

2.1. Electrode Design for Conventional Battery Chemistries

LIBs have been widely used for over 20 years because of their high specific energy, acceptable cycle lifespan and acceptable cost; however current designs are approaching their theoretical limits. Many researchers have since investigated novel design methods in order to improve the performance, which can generally be categorized into two: conventional electrode design strategies and emerging techniques for next-generation energy devices. This subsection reviews electrode design strategies that have been applied to conventional battery chemistries.

2.1.1. Low-tortuosity Porous Electrodes

Despite the higher energy density thick electrode designs may bring, the electrochemical performance is inevitably restricted by the poor transport of ions and electrons through the thick electrode. Here, ions transfer through the narrow channels in the pores, while electrons travel through the network formed by conductive additives. In both cases the percolation and tortuosity of the pore and conductive network phase can significantly influence the electron and ion transport in electrode [29-33]. (Fig. 2(a,b)) In the case of a thick electrode, diffusion distances are by definition larger than thin electrodes, and therefore it is critical to have a low tortuosity design to ensure sufficiently high active material utilization at practical current densities. This is highlighted in Fig. 2(a,b)). In order to quantify the impact of tortuosity on the diffusion properties, the effective ionic conductivity can be defined as: $D_{eff} = D\varepsilon/\tau$, where ε is the porosity, τ is the tortuosity and D is the intrinsic ionic conductivity. Reducing the tortuosity of electrode can promote ion transportation and electrolyte permeability, which has become an effective method in thick electrode design. At the same time, the directional tortuosity also is related with the porosity as shown in Fig. 2(c,d) and needs to be considered, with the through plane transport often the dominant figure of merit [34].

Generally, there are two major methods for low-tortuosity electrode which are centered around additive and subtractive approaches. Additive methods can be defined as selective deposition of slurry to build electrodes layer by layer, which includes methods such as 3D printing [35-40]. The most challenging part of 3D printing is the creation of printable electrode slurries, which should exhibit sufficient stability and viscosity to build a stable, well-functioning thick electrode. However, if this can be achieved structured electrodes which allow for facile ingress of the electrolyte can enable high rate performance. Hu et al. designed a LiMn_{0.21}Fe_{0.79}PO₄@C nanocrystal cathode with 3D printing which exhibited ultra-high rate and high capacity performance with a capacity of 108.45 mAh g^{-1} at 100C and a reversible capacity of 150.21 mAh g⁻¹ at 10C after 1000 cycles (Fig. 3(a)) [35]. Through 3D printing, the thickness and width of electrode pillars, can be controlled by adjusting the nozzle speed and dispenser pressure. Moreover, their research also found that for their materials, solid-phase diffusion is not the limiting factor but rather, it is the effective electrolyte diffusion properties which relate to factors like porosity and thickness.

In contrast, subtractive methods can be defined by the removal of some component of the electrode to create a porous structure in a certain shape. Compared to additive approaches, subtractive techniques are generally easier to apply. In conventional electrode manufacturing processes, active material, binder and conductive additives are mixed together and directly coated on the surface of a current collector, such as copper and aluminum foils, followed by air or vacuum drying at elevated temperature. During this drying process, due to the lack of a templating media, the tortuosity of the resulting electrode is relatively high. This has led many researchers towards approaches which create oriented structures to decrease the tortuosity [41–48].

Among all the design strategies which introduce an aligned porous structure, the most commonly used method for creating relatively straight channels in the electrode utilize different forms of external force, including magnetic and viscous shearing forces. Sander et al., for instance, reported a magnetic control method to create directional pore arrays in lithium-ion battery electrodes, which can reduce ionic resistance [46]. The electrode exhibits extraordinary electrochemical performance, with threefold higher areal capacity up to 9.5 mAh cm⁻² than conventional electrodes at practical charge/discharge rates like 1C. Apart from the magnetic approach, Xia et al. designed a mechanical shearing method for a discotic lamellar liquid crystal (LC) phase of 2D titanium carbide $\mathrm{Ti}_3\mathrm{C}_2\mathrm{T}_X$, leading to vertically aligned $\mathrm{Ti}_3\mathrm{C}_2\mathrm{T}_X$ MXene films [47]. The electrode design substantially enabled faster ion transport and electrochemical tests show excellent thickness-independent rate performance with specific capacitance of nearly 250 F g⁻¹ and homogeneous thickness performance up to 200 μ m. More recently, natural structures such as: wood, coral and ice, have inspired aligned structural template methods, which have shown promising potential in lowtortuosity thick electrode designs [[44],[48]]. Zhao et al. reported a supercapacitor with wood-like aligned morphology (Fig. 3(b,c)), showing high electrode mass loading and excellent areal capacitance of 831 mF cm⁻² at 6.2 mA cm⁻². The aligned electrode matrix exhibits remarkable properties such as broad morphological tunability and mechanical flexibility.

Despite the improved energy density of thick electrode designs, increasing thickness can also result in many other consequences such as undermining the potential stability and lithium plating under fast charging conditions. When increasing the thickness, it is more difficult to ensure a homogeneous Li-ion concentration distribution throughout the electrode thickness, which recently has been shown to have crucial influence on Li deposition behavior [49]. In addition, the Li-ions may become depleted near the current collector under high C-rate conditions, resulting in inactive parts of the electrode and increased local current density near the separator [50]. These enhanced current densities near the electrode-separator interface can also result in amplified mechanical stresses in these locations, leading to particle fragmentation and accelerated degradation of the battery [[47], [51]]. Therefore, the balance between electrode thickness and charge rate should be balanced, with suggestions by Gallagher et al. that the current density should remain below 4 mA cm⁻² [28].

2.1.2. Binder-free/3D Current Collector Design

Conventional electrode configurations normally contain three main components - active material, binder and conductive additives. Increasing the proportion of active material is necessary to improve the electrode energy density on the cell level, which means reduction of the ratio of binder and conductive additive is necessary. However, the conventional conductive network is usually formed by conductive additives like carbon black which lack mechanical strength. Therefore, polymeric binders such as PVDF are needed for the active material and conductive additives to form a mechanically stable electrode and adhesion with the current collector. Unfortunately, in most cases, the binder itself is usually electrically insulated, resulting in restricted electron transport. The consequence of this is that the utilization of binder may cause a reduction in the electrochemical performance. Modification of binders and conductive additives have been thoroughly investigated over the past few years, and the most commonly used strategies is to build binder-free



Fig. 2. Ion pathway for electrode with different thickness and tortuosity design. (a) Conventional thin electrode for fast charging with high tortuosity (single layer). (b) Thick electrode with low and high tortuosity. The blue dashed line with arrow indicates the ion pathways during delithiation. Tortuosity-porosity relations for (c) NMC, (d) LCO. (c),(d) Reproduced with permission from Wiley [34].

conductive percolation networks through the use of advanced conductive additives like carbon nanotubes (CNTs) [52–56].

Among all the conductive materials, 1D and 2D conductive additives like CNTs, graphene and MXene are commonly used to fabricate binderfree percolation networks in electrodes [[57],[58]]. CNTs are capable of functioning as a binder and conductive additive at the same time due to their 1D structure, enabling fast electron transport as well as mechanical support due to the entanglement of the CNTs. Park et al., for instance, designed a 800 μ m thick electrode with extraordinary performance, displaying conductivity up to 10⁴ S m⁻¹ [57]. The segregated composite network is produced directly on Cu substrates with hierarchical structures by a scalable slurry-casting method, showing excellent mechanical and electrical performance. The resulting electrode features state-of-the-art areal capacity of 30 mAh cm⁻² and energy density of 480 Wh/kg.

Apart from 1D materials, 2D materials such as graphene and MXene, offer excellent conductivity and mechanical strength for percolation networks. Since the exciting breakthrough of graphene by Andre Geim and Konstantin Novoselov in 2004, the material has frequently been used in LIBs as a 2D conductive additive [59–62]. Yet, despite the long standing application of the material, there still exists a gap between laboratory studies and practical application of graphene, owing to the high cost of pure graphene. Therefore, recent researches have mainly focused on producing graphene-based electrodes by reduction of graphene oxide (GO) to reduced RO (RGO), which can be easily synthesized out of commercial graphite. Zhao et al. reported a co-assembly method to construct thick cathodes based on 3D graphene frameworks (IGF) with low tortuosity and mechanical robustness [63]. The LiFe_{0.7}Mn_{0.3}PO₄ nanoplates (LFMP NPs) and graphene were aligned vertically along the growth channels of ice, enhancing ion transfer across the electrode by 2.5-fold compared with random structured electrodes. Fig. 4(a) shows the schematic preparation of a vertical-channel sandwich electrode (LFMP-IGF) aerogel. Here, the framework with uniformly graphene wrapped LFMP NPs was formed by dispersing LFMP and ascorbic acid. The resulting electrode with vertical channels exhibits a high mass loading (21.2 mg cm⁻²), excellent rate performance (0.2 - 20C) and cycle performance of 1000 cycles.

However, due to the specific structure formed by layers of the RGO flakes, defects exist which reduce the electrochemical performance and cycle life, especially at high mass loadings. Recently, research has been carried out to address these defects and ultimately use them to enhance the performance of the cell [[64],[65]]. Sun et al. designed a three-dimensional holey-graphene/niobia (Nb₂O₅) composite electrode with ultra-high charge rate up to 100C at practical levels of mass loading(>10 mg cm⁻²) [64]. In the electrode, Nb₂O₅ was supported by a conductive scaffold formed by the holey porous graphene. The composite was synthesized by a two-step synthesis process using GO and Nb₂O₅ as precursors, which displayed a hierarchical porous structure, facilitating ion transport. In addition, a high areal capacity and high-rate capability at high mass loading were achieved by systematically optimizing the porosity in the graphene backbone.

As a relatively new class of 2D materials, MXenes have shown great potential in the fabrication of free-standing electrodes in recent years [66–71]. Compared with other conducting materials such as CNTs and graphene, MXenes offer excellent strength and remarkable electrical conductivity, and relatively simple fabrication processes for electrodes.



Fig. 3. (a) LMFP nanocrystal 3D-printed electrode. a) Reproduced with permission from Wiley [35]. (b) Wood-like natural aligned structure (c) Schematic of the fabrication of all-solid-state hydrogel supercapacitors via integration of APA as the matrix and PPy as the electrode material. (b),(c) Reproduced with permission from Wiley [44].

Zhang and coworkers developed a scalable method for the fabrication of pure MXene films containing highly aligned large MXene flakes with a tensile strength of 570 MPa and high conductivity of 15,100 S cm⁻¹ [66]. Chen et al. reported 2D MoS₂-on-MXene heterostructures through *in-situ* sulfidation of Mo₂TiC₂T_x MXene, with a remarkable capacity of $\approx 600 \text{ mAh g}^{-1}$ and good cycling stability, maintaining 500 mAh g⁻¹ after 100 cycles, demonstrating the possibility of large-scale manufacturing of MXene-based energy storage devices. Nowadays, most work focuses on the particular application of MXenes. Despite all the exceptional properties that MXenes possess, there is still a lack of specific explanations about what makes them so multi-functional, leading to further research on how to control and modify them.

In a conventional cell configuration, one complete layout contains four components - current collectors, electrodes, electrolyte, and separator, showing a sandwich-like structure layer by layer. In addition to the electrode, the current collector is also critical of the battery, providing high electronic conductivity in the in-plane direction. Normally, metallic foils are used as a current collector in a planar electrode, on which the electrode layer is laminated. Stable contact between the electrode and current collector are required in order to meet the demands of rapid electron transport under fast charging. However, one of the most significant concerns in conventional current collectors is the delamination of the active material, caused by the drying process of the slurry. During the drying process, the evaporation of solvents results in stresses which can cause peeling, and when compounded with the migration of binder towards the surface of the electrode leads to a weakening of the electrode-current collector interface. During operation, the volume expansion of the active material particles can further exacerbate this delamination problem, leading to the deterioration of electrochemical performance. In order to address this issue, many researchers have investigated current collectors.

Unlike the percolating conducting phase network in electrodes discussed above, this section mainly focuses on the 3D construction of the current collector, though they both share similar requirements, namely



Fig. 4. (a) The fabrication of 3D vertically aligned and interconnected porous LFMP-IGF cathodes by ice templating. (a) Reproduced with permission from Wiley [63]. Surface pictures of (b) lithium metal, (c) copper mesh, and (d) the Li-metal/3D copper mesh composite. Scanning electron microscopy images of (e) lithium metal, (f) copper mesh, and (g) the Limetal/3D copper mesh composite. (b)-(g) Reproduced with permission from Wiley [75].

the need to build conductive pathways to ensure rapid electron transport. In recent years, 3D current collectors based on various conductive materials have shown great potential for improving the electrochemical performance of a battery. There are now a range of materials for conductive current collectors, such as graphite, metals and alloys, and polymers [[38],[72–75]].

One representative design approach is the substitution of conventional planar metallic foils with 3D metal current collectors with a porous structure. This not only provides the pores for containing active material particles but also functions as the host framework for the electrode. Li and coworkers reported a 3D Cu current collector/Li composite anode with a porous structure, which provides a scaffold for Li metal deposition/stripping and accommodates volume change at the same time [75]. (Fig. 4(b -g)) In addition, due to the confinement effect, the specific design of this kind of porous structure can uniformly provide shortened ion transport channels with high homogeneity, resulting in a good rate performance at relatively high mass loading.

2.1.3. Electrode/Electrolyte Interfacial Design

During the working process of LIBs, the solvents and salts dominate the diffusion kinetics of Li⁺ in the electrolyte and SEI layer, both of which are the key factors of interfacial design. The SEI plays a necessary role in the stable operation of a battery, and is formed by the reactions between electrolyte and anode during the first cycle, forming a protective layer between these two components to prevent the constant decomposition of the solvents [[76],[77]]. Therefore, an ideal SEI film should have high ionic conductivity and excellent mechanical performance to ensure the rapid migration of ions and stability towards the volume expansion experienced during cycling.

Towards understanding this, there are various research efforts focusing on developing low-resistance and artificial SEI films [78-82]. Shi et al. reported a novel electrolyte additive - fluorosulfonyl isocyanate (FI), which enabled a low-resistance SEI film to be formed on graphite anodes [82]. The conductive SEI was formed by the reduction reactions of FI due to its high reduction potential (>2.8V vs. Li/Li +), and contains a highly protective inorganic inner layer to prevent the outer layer from constantly growing. Compared with the reference liquid electrolyte (LP30), the resulting battery with 2 wt% FI exhibits good rate capability and low temperature performance due to the low interfacial resistance. Zheng and his coworkers reported that an optimal dual-salt/carbonate-solvent-based electrolyte, containing 0.05M LiPF₆ in LiTFSI-LiBOB can substantially improve the rate performance and cycling safety stability of the battery cell [83]. Moreover, a robust and conductive SEI film is formed due to the polycarbonate containing constituents. With a limited increase in electrode overpotential, the resulting battery exhibits excellent electrochemical performance with current density of 1.75 mA cm⁻² and a capacity retention of 97.1% after 500 cycles. (Fig. 5(a,b)) However, despite all the progress in developing highly conductive and dendrite-free electrolytes, a complete understanding of the specific mechanisms occurring in the SEI is still lacking, which provides a promising yet challenging problem. Through years of exploration, solid-state electrolytes seem to be recognized as the emerging direction with one of the greatest potentials, which will be discussed in the next subsection.

As discussed before, the electrolyte is a fundamental component in the battery and dictates the ease at which mass transfer and ion migration can occur, as well as the lifetime and safety. Hence the electrolyte is also an important consideration in the design of a cell. Electrolytes



Fig. 5. (a) SEI thickness of organic and inorganic layer formed with different electrolyte. (b) Rate capacity of graphite/Li coin cells with LP30 and LF-2 at -20°C. (a), (b) Reproduced with permission from Elsevier [82]. (c) Schematic illustration of the effect of CTAC addition on the Li plating process. (c) Reproduced with permission from American Chemical Society [99].

can be divided into several types depending on composition, such as aqueous electrolytes, organic electrolytes and ionic liquid electrolytes [84–87]. Nowadays, organic electrolytes have taken up the majority of the commercial market, due to their high voltage window (2.6-2.9 V vs Li/Li⁺) [88]. These organic electrolytes normally contain organic solvents and lithium conducting salts which are dissolved in the solvents.

Fast charging is now an increasingly important requirement for nextgeneration of energy storage devices, which requires highly conductive electrolytes and separators. However, under fast charging conditions, batteries tend to encounter several drawbacks, such as high operating temperature, Li plating, and increased risk of thermal runaway (TR) [89–92]. Recent review by Logan have well summarized the current electrolyte designs especially for fast charging batteries, in which approaches like using low-viscosity cosolvents, highly concentrated electrolytes, and large polyanions were thoroughly reviewed [94]. In the past few years, a variety of electrolyte formulations have been developed to improve rate performance and safety [95–98]. Compared with organic and ionic electrolytes, aqueous electrolytes usually exhibit higher ionic conductivity, leading to better rate capability and lower specific internal resistance [97]. Han and coworkers reported (trimethylsilyl)isothiocyanate (TMSNCS) based on aminosilane as an additive in LiPF₆-based electrolytes [95]. It was found that TMSNCS effectively deactivates the reactive species owing to a high donating ability to scavenge HF and PF5 and maintains long-term stability of interfacial films formed on electrodes. The cycling experiments showed that NCM622/graphite full cells with 0.1% TMSNCS can exhibit superior discharging capacity up to 144 mAh g⁻¹ and capacity retention of 91.8%. Qian et al. designed a highly concentrated electrolyte composed of ether based solvents and lithium bis (fluorosulfonyl) imide salt, which enabled a lithium metal anode to maintain a high Coulombic efficiency (about 99.1%) even under high-rate cycling conditions without dendrite growth [98]. Excellent electrochemical performance was achieved with the lithium bis (fluorosulfonyl) imide as the electrolyte, exhibiting high current density of 10 mA cm^{-2} for more than 6,000 cycles in a lithium/lithium cell. In order to address the potential problem of severe dendrite formation, Dai et al. used hexadecyl trimethylammonium chloride (CTAC) as an electrolyte additive to form a uniform Li deposition by lithiophobic repulsion mechanisms, as shown in Fig. 5(c) [99]. The resulting battery cell with CTAC additives exhibited a significantly enhanced cycle life of 300 h at 1.0 mA cm⁻² and rate performance up to 4.0

mA cm⁻². Despite of the achievements in developing electrolyte for high C-rate charge application, one of the major issues in ionic transport still remains in measuring transport properties other than conductivity, future researches on fusion of electrochemical characterization techniques are desirable.

2.2. Electrode Design for Next-Generation Energy Devices

Despite all the achievements in the modification and optimization of conventional LIBs, current battery technologies are gradually approaching the theoretical energy density limits. As the demands for higher energy density, higher charge/discharge rates and safety concerns grow, researchers are gradually focusing more attention on emerging technologies for novel energy storage devices, leading to development of next-generation battery chemistries. There are now many promising candidates for next-generation LIBs, which possess not only better electrochemical performance but also safety characteristics. Among all the novel technologies for LIBs, development of a stable Li metal anode has recently been recognized as the 'Holy Grail' anode due to its low electrochemical potential and very high specific capacity. Recently, anode-free design also provides a novel pathway for high energy density Li metal batteries. Solid electrolytes and chemistries such as Li-S/Li-Air are also promising technologies. However, despite all the innovative designs for such next-generation batteries, there still exist many issues that have to be addressed. In this subsection, the electrode design for these emerging batteries will be discussed, including their progress, advantages and challenges.

2.2.1. Li-Metal Anode Design

Lithium metal exhibits the lowest electrochemical potential (-3.04V vs. standard hydrogen electrode) and the highest theoretical capacity (3860 mAh g^{-1}) among all anodes under investigation [[1],[100]]. In addition, the lithium metal electrode plays an irreplaceable role in emerging battery chemistries such as Li-S and Li-Air batteries. However, despite all the electrochemical benefits that Li metal anodes can bring, there still are several fundamental challenges in this field including volume change, Li dendrite growth and unstable SEI formation, all of which severely hinder the practical application of the Li metal electrode [101–104]. In fast charging conditions, the conventional planar Li metal electrode will go through dramatic volume changes and SEI fracture, resulting in hazardous consequence like Li filament formation and TR. In order to address these issues, research has been conducted to investigate methods of modifying the performance of Li metal electrode. Recent research found that applying various 3D conductive hosts to lithium metal anodes can be beneficial for preventing dendrite growth, confining dramatic volume change and decreasing the local current density, resulting in improvements in the electrochemical and safety performance [105-108]. In addition, Li alloys shows great potential in developing long-life batteries by uniformizing Li deposition to avoid the catastrophic consequence of dendrite growth [109–111].

In the past few years, many researchers have achieved significant progress in Li metal anode design [[60],[112–118]], Inspired by the natural structure of the eggplant, Cui's group have developed a channel-like structure from a carbonized eggplant (EP) as a stable "host" for Li metal melt infusion [112]. (Fig. 6(a)) With further interphase modification, with the use of lithium fluoride (LiF), the resulting EP–LiF composite anode exhibits approximately 90 % of the theoretical capacity of Li metal and excellent lifetime performance by suppressing dendrite growth and volume fluctuation during cycling. This kind of nature-inspired design will likely motivate further artificial electrode framework designs for both Li anode and sulfur cathodes, leading to a novel design guidebook for future high-energy density batteries.

The infusion strategy, however, is often difficult for a Li metal composite anode. Chi et al. reported a thermal infusion strategy that can infiltrate lithium into a nickel foam to construct a composite Li metal anode [119]. The thickness of the nickel foam was 800 μ m, where the struts of the nickel framework were approximately 100 μ m. Compared with conventional pure Li metal electrodes, the resulting anode exhibits an excellent electrochemical performance with high current density of 5 mA cm⁻² for more than 100 cycles, showing lower interfacial resistance and smaller polarization. The presence of a stable scaffold provided repeatable nucleation sites for lithium deposition/stripping, however it makes the application of pressure to close voids more difficult.

Generally, during the electroplating process of metal anodes, Li dendrites can be easily formed because of the intrinsic lattice and surface energy properties of lithium metal, severely hindering the practical application of Li metal anodes. The occurrence of Li dendrites exacerbates the reactions between electrolyte and Li metal, forming more SEI layers. Besides, plated Li metal are easily detached from current collectors due to the dissolution process, namely the 'dead lithium', resulting in inversible loss of lithium inventory. In order to address the concern of Li dendrites, Zhang's group investigated the underlying mechanisms behind dendrite growth behavior as well as the critical conditions and rate limiting processes in the plating and stripping process, offering guidance for the rational design of 3D structured lithium metal anodes [120]. Fig. 6(b) shows the simulated process of structured lithium metal anodes, where the consumption of lithium ions aggravates dendrite growth and result in further SEI formation. After that, Zhang and coworker developed dendrite-free Li metal anodes by constructing an optimized lithium host with balanced lithiophilicity and conductivity, resulting in excellent electrochemical performance with high Coulombic efficiency of 98% stable for over 300 cycles at 1.0 mA cm⁻² [118]. Li hosts, with synergistically coupled lithiophilicity and conductivity, can effectively form uniform Li deposition and inhibit Li dendrite growth. Liu et al. also reported a scalable high-performance Li metal anode with uniform Li plating/stripping process through lithium-cobalt alloying on hierarchical scaffolds [114]. Fig. 6(c) shows the growth of CNTs on electrospun fibers and morphology changes during alloying and Li plating, the CNTs on the fibers go through an in-situ growth, towards these hierarchical structures which highly facilitate facile plating/stripping process, forming uniform Li deposition on the surface.

Recently, some researchers brought up a concept of anode-free lithium metal full cells, which basically contain pairs of fully lithiated cathodes with a bare metal current collector. The specific working mechanisms of anode-free designs require the cell to be assembled in the discharged state, with only cathodes and metal foils. During charge, Li⁺ in the lithiated cathodes are gradually reduced and plated as Li metal on the Cu foil, forming a relatively thin film of deposited lithium. This anode-free configuration is capable of storing 60% more energy per volume than conventional cells [121]. However, the anode-free designs suffer from severe capacity loss and short lifespan, mainly due to the poor efficiencies of lithium plating and stripping during charge/discharge processes. Recent review by Nanda et al. have summarized the various strategies to sustain the lithium inventory for anode-free cells, such as modification of electrolyte, current collector and cycling optimization [122]. Despite their great potential, anode-free designs still need significant improvement before they can approach commercial application.

2.2.2. Solid-State Electrolyte Based Electrode Design

Despite significant improvements in the electrochemical performance with Li metal composite electrodes, conventional battery configurations with liquid electrolyte still face severe safety concern such as ISC and TR, especially under fast charging condition with high current density and internal temperatures. In contrast, solid electrolytes are renowned for their safety performance, which contributes to their potential as a replacement for conventional liquid electrolytes. The practical application of solid electrolyte requires several comprehensive properties such as high ionic conductivity, low interfacial resistance, excellent thermal stability, good mechanical strength and relatively low cost. During the development of solid electrolytes, various material possibilities have emerged such as inorganic ceramic electrolytes, solid polymer electrolyte materials and mechanically compliant materials like iono-



Fig. 6. (a) Channel-like structure of a carbonized eggplant (EP) as a stable "host" for Li metal melt infusion. (a) Reproduced with permission from Wiley [112]. (b) Schematic illustration of the simulated structured lithium metal anode. (b) Reproduced with permission from Elsevier [120]. (c) Proposed preparation mechanism for the growth of CNTs on electrospun fiber for scalable high-performance lithium-metal anodes. (c) Reproduced with permission from Elsevier [114].

gels [123–131]. Previous review by Manthiram has thoroughly categorized the current solid electrolytes and discussed the merits as well as limitations for these electrolytes [132]. Despite of the efforts in improving the performance for solid-state electrolyte materials, crucial challenges still remain in achieving acceptable ionic conductivity, maintaining electrochemical stability and addressing the interface issue between electrolytes and electrodes.

Nowadays, the most representative type is the inorganic solid electrolyte. Due to their intrinsic mechanical properties, the inorganic solid electrolyte holds a rigid form which can prevent Li dendrite and other materials from penetrating into the electrolyte, resulting in good chemical and physical stability. In addition, inorganic electrolytes also possess excellent ionic conductivity, which can approach or even surmount that of liquid electrolytes [133–136].

Recently, research has been conducted to seek the next-generation solid electrolytes with even better properties. 3D garnet solid electrolytes have been recognized as a promising design option for solid-state batteries, which usually contain a dense layer as separator and a porous layer as conductive 3D host. The special configuration of 3D garnet electrolytes, not only enables rapid ion transport but also high mechanical performance to mitigate volume change in the porous layer, while the dense layer acts as a separator to inhibit Li dendrite penetration [126–130]. Xu and coworkers reported a three-dimensional mixed electron/ion conducting framework (3D-MCF) based on a porous-dense-porous trilayer garnet electrolyte structure which was formed by a con-

formal coating of CNTs on the porous garnet structure [126]. The Li metal anode was formed by slow electrochemical deposition into the 3D-MCF, resulting in sufficient contact between the metal anode and garnet electrolyte (Fig. 7(a-c)). In addition, the porous garnet structure exhibits 40 times higher contact area compared to a conventional dense pellet, enabling 40 times higher overall areal current density of 3 mA cm⁻² for stable Li deposition with the same local current density.

Although ceramic electrolytes particularly the garnet type have shown great potentials in developing Li metal batteries with excellent safety performance, mostly because of its nonflammability and outstanding chemical stability to Li metal, poor interfacial contact and brittle mechanical property still hinder the practical application for creating flexible film out of ceramic solid electrolytes. In order to address these issue, Jiang et al. reported a garnet-based composite electrolyte using flexible 3D Li_{6.75}La₃Zr_{1.75}Ta_{0.25}O₁₂ (LLZTO) framework with a succinonitrile solid electrolyte [127] Figure 7d shows the preparation of the composite electrolyte, resulting in wide electrochemical window (4.8V versus Li/Li⁺) and relatively high lithium transference number (0.53). The Li-ions tend to transfer through the continuous channels between interconnected LLZTO particles and succinonitrile. These channels and the soft electrolyte/electrode interface both contribute to achieving a high ionic conductivity of 1.2×10^{-4} S cm⁻¹ and good stability of the Li symmetric battery. In addition, the solid-state batteries based on the composite electrolyte possess high cycle stability and excellent specific capacity of 153 mAh g^{-1} .



Fig. 7. (a) Schematic illustration of the porous–dense–porous trilayer garnet framework with a mixed electron/ion conductor incorporated on the anode side. (b), (c) Working mechanism of anode during discharge and charge. (a)-(c) Reproduced with permission from American Chemical Society [126]. (d) The preparation schematic of 5PTFE-100LLZTO-16[SN₂₀–LiTFSI] solid-state electrolyte. (d) Reproduced with permission from Wiley [127].

2.2.3. Electrode Designs for Novel Battery Chemistries

Compared with other commercial battery chemistries, the Li-Air battery exhibits ultrahigh specific energy density of nearly 1700 Wh kg⁻¹, almost 6 times higher than normal LFP-based batteries. However, several significant issues continue to hinder the practical application of the Li-Air battery, including the instability of electrolytes based on organic carbonates, competing mass transport issues and blockage of pores in the porous air cathode, to name but a few. The electrolyte is regarded as an essential component in the Li-Air battery, which requires excellent stability of both the O₂ and LiO_x compounds, sufficient ionic conductivity, O₂ solubility and mobility [[137],[138]]. However, unlike conventional battery chemistries, the reactions of the Li-Air cell involves ambient O2 that comes from the external environment on a porous cathode, highlighting the essential role of the cathode structural design. Since the anode of the Li-Air cell is normally Li metal, which has been discussed in Section 2.2.1, this part will mainly focus on the structure design of the cathode in Li-Air batteries.

The fundamental mechanisms of Li-Air batteries require oxygen, and their products, to undergo the reduction and evolution reactions, necessitating a porous cathode to introduce the ambient oxygen into the electrode. This is similar to the design challenges for fuel cells. Therefore, a similar low-tortuosity design requirement for Li-Air cathodes is needed to not only enable good ion/electron transport kinetics. In addition, Li-Air batteries usually suffer from the accumulation of reduction product which are typically Li₂O₂ and can lead to pore blockage, further restricting mass transport of reactants. In order to address this issue, an increased density of active sites is required for the electrode design to prolong the lifespan and improve areal capacity.

Recently, many researchers have explored the structural design optimization for Li-Air cathodes, with conductive 3D porous thick electrode designs as promising routes [139–142]. The design principle shares many similarities with the binder-free design that was discussed in Section 2.1.1, both of which usually apply 1D and 2D conductive materials like CNTs and graphene. However, the optimization of this reaction requiring due consideration of the triple phase boundaries for facile gas, ionic and electronic transport.

Unlike the common porous carbon-air cathode where the O_2 gas and electrolyte usually compete for transport pathways, Xu and coworkers reported a novel textile-based air cathode with decoupled pathways for O_2 and electrolyte, which can effectively improve the overall battery performance [140]. The textile is formed by wetting the fiber with a CNT coating to enable fast electron transport along the



Fig. 8. (a) Schematic illustration of the design and decoupled pathways (O_2 gas, electrolyte) within the wet textile-based air cathode. (a) Reproduced with permission from Wiley [140]. (b) Schematic illustration of the synthesis process of the oval-like carbon microstructures (OLCMs). (b) Reproduced with permission from, Wiley [153].

CNT-coated surface as well as Li⁺ transport along the cotton fibers. As Fig. 8(a) shows, the 3D hierarchical structure exhibits high porosity to introduce oxygen through the pathways between the CNT-coated fiber, enabling rapid oxygen diffusion with open macropores. The resulting textile-based cathode features stable and excellent electrochemical performance with a high capacity of 8.6 mAh cm⁻² over 50 cycles.

Despite these novel modification methods to achieve better performance, Li-Air batteries still face several crucial problems which makes them far from industrial application. Continual effort still needs to be put into balancing the volumetric energy density owing to the highly porous electrode and the transport of ion, electron and gas. Pore engineering, improvement of electrolytes and catalyst are all promising to overcome the obstacles before the commercialization of Li-Air batteries.

Apart from the Li-Air chemistry, Li-S also has been widely recognized as a promising candidate for next-generation batteries due to its high specific capacity of 1672 mAh g^{-1} and specific energy of 2600 Whkg⁻¹. However, to achieve these theoretical values thick electrodes are required. Yet, due to the insulating properties of sulfur and polysulfides, electrode design for Li-S batteries can be quite challenging, with more of a focus on optimizing the electronic conductivity [143–147]. However, during operation, a Li-S battery suffers from many problems. Firstly, insulation layers can be formed by discharged S-products on the electrode surface, severely undermining the electrochemical utilization. Secondly, the polysulfides may be reduced on the surface if the electrode is too thin. Most importantly, due to the fact that the polysulfides are highly soluble in organic electrolyte, the dissolved polysulfides are likely to diffuse through the porous separator and form short-chain polysulfides the anode surface. These polysulfides sometimes can diffuse back to the cathode if concentrated, resulting in repeated migration of polysulfides, which is the shuttle effect [[9],[148]]. For now, shuttle effect still remains a crucial challenge that Li-S batteries is facing for the undesirable consequences such as continuous loss of active materials, anode degradation and the resulting degraded electrochemical performance.

Some researchers have investigated routes for higher sulfur loading [149–152]. Ye et al. reported an electrode consisting of oval-like carbon microstructures formed through double "Fischer esterification" to fine-tune the surface characteristics, and function as the sulfur host for the thick sulfur electrode [153]. (Fig. 8(b)) The areal loading of sulfur is normally controlled to be under 2 mg cm⁻² to achieve good reversible capacity. In this research, the resulting electrode displays excellent electrochemical performance with a high sulfur loading of 8.9 mg cm⁻² and specific discharge capacity of 8.417 mAh cm⁻² due to the tuning of the surface performance.

Hollow porous carbon materials (HPCM) have also been widely applied in the electrode design for Li-S batteries, greatly enhancing the electrochemical performance of both sulfur cathodes and lithium anodes [148]. The HPCM-based electrodes can be divided into several categories, including spheres, 1D structures, 2D structures, hierarchical structures, and composite structures [[148],[154–156]]. However, low cycling stability, capacity and safety concern, still hinder Li-S electrode design.

3. Multiscale modelling for electrode design

Modelling methods are now commonly recognized as an essential step in electrode optimization, which is just as important as the synthesis and characterization methods. Multiscale modelling normally contains several physical scales, ranging from the atomic and particle scale, to the electrode and even vehicle scale, with different modelling methods required according to the length scale. In the case of the electrode length scale, different modelling methods including microscale structural modelling and macroscale functional modelling are investigated in this chapter.

3.1. Microscale Structural Modelling

Microscale structural modelling of battery electrodes is now an integral part of electrode optimization, in terms of understanding how coupled features such as local tortuosity and electronic phase percolation, affects the overall electrochemical performance. In this section, we mainly focus on the structural modelling methods based on image characterization as these are able to capture the complexities of real microstructures.

As discussed in Chapter 2, microstructure design objectives such as low-tortuosity structures play a crucial role in electrode design for LIBs, affecting the macroscale electrochemical performance and the safety capability. In the conventional design process, microscopic structural models usually act as auxiliary methods to provide theoretical calculations, to understand the processes occurring in a cell. In this approach, the majority of design optimization is still experimentally conducted by trial and error, which is time-consuming and costly. Recently, researchers have realized the necessity of microscopic models for electrode design, with increasing numbers of studies using modelling to inform design decisions proactively rather than retrospectively [157–160].

Unlike continuum models such as the pseudo 2D (P2D) model or the single particle model (SPM), both of which usually are used to investigate internal reaction mechanisms and external electrochemical performance through homogenization of bulk electrode properties, microscopic models mainly focus on the local physical or electrochemical properties of an electrode, such as local porosity and tortuosity.

Image characterization methods like focused-ion beam scanning electron microscopy (FIB-SEM), neutron computed tomography (NCT), and X-ray computed tomography (XCT) have been recognized as effective methods of quantifying battery microstructures, with researchers taking these images and using them in electrochemical models [161– 167]. Fig. 9(a) illustrates the 3D characterization methods that commonly used in microstructure reconstruction of electrode materials, among which XCT has shown outstanding potentials in non-destructive 4D studies.

However, whilst these techniques are powerful most battery electrodes are composites of active materials, conductive additives and binders, with many features at the nano-scale. Of the various techniques, FIB-SEM currently offers the best microstructural resolution, however it requires the destruction of the electrode through ion beam milling. Given the porous nature of these electrodes, a resin is often infiltrated to provide mechanical support however this can make segmentation of carbon containing phases more difficult to discern. To address this, Biton et al. reported a novel method of electrode sample preparation with a brominated epoxy impregnation to improve image contrast, which enables higher FIB-SEM resolution (3D imaging) [161]. Fig. 9(a) shows the FIB-SEM 3D images of their LFP electrode, and the size distribution of each phase was analyzed accurately from the complex 3D electrode microstructure using advanced quantification algorithms. Through the use of the brominated epoxy the carbon phase could also be differentiated from the pore phase, which is often challenging in FIB-SEM reconstructions. In electrode systems using low electronic conductivity materials, therefore, omitting details of the electronic conducting phase can lead to errors between the simulations and experimental results.

As a representative 3D characterization method, XCT has been widely applied in three-dimensional characterization with excellent resolution capability for multiscale imaging from the system to particlelevel. XCT can operate in multiple modes: absorption as described by the attenuation coefficient, diffraction as described by the Bragg equation, fluorescence as described by Moseley's law, with each technique offering unique information and bringing individual merit [168]. With the help of high-throughput XCT, the inner process of cell degradation and lithium intercalation can be identified. Recently, Ziesche et al. reported the success of the temporally and spatially resolved tracking of lithium intercalation and electrode degradation processes using correlative neutron and X-ray tomography with a virtual unrolling technique [163]. This technique was used to precisely detect the minor fluctuations inside the electrode layers which are hard to observe by conventional 3D rendering tools. In addition, the direct lithium diffusion process and electrode wetting by the electrolyte were directly captured by complementary neutron computed tomography.

Combined with modelling methods, 3D microstructure reconstruction techniques have shown great potential in developing multiscale modelling platforms. Kashkooli et al. developed a multiscale modelling platform based on the actual microstructure morphology characterized by nanoscale XCT [169]. The microscopic models that reveal the intrinsic mechanisms were established and connected with the macroscale models, where external electrochemical performance metrics like capacity can be simulated. In the micro level, 3D reconstructed microstructures were established on real-time XCT images (Fig. 9(b)), while at the macro-level the electrochemical performance models were modified simultaneously by solving the equations with real physical parameters such as porosity and tortuosity. The resulting simulations of charge/discharge show excellent consistency with experimental data, verifying the possibility of creating a virtual model based on the results of 3D reconstruction techniques. 3D reconstruction techniques provide a solution for identifying the microscopic parameters for electrodes, which certainly affect the electrochemical performance of batteries. Therefore, microscale structural modelling methods have become possible choices for model-based approaches due to their direct descriptions of electrode microstructures.

In addition to 3D reconstruction, X-ray diffraction computed tomography (XRD-CT) has also been proven to provide 3D diffraction information, which is crucial to analyze the internal stress/strain conditions as well as heterogeneous phase information [[170],[171]]. In addition, owing to its special non-destructive characteristics, XCT is also capable of connecting microscopic structure dynamics with lifetime performance by conducting 4D or *in-situ* characterization, although the requirements for samples are relatively strict compared with conventional analysis.

3.2. Macroscale Functional Modelling

Despite all the achievements in microscale modelling, many researchers focus on macroscale performance and key metrics such as areal capacity, rate performance and safety [172–178]. As the energy and power demands for fast charging grows, a more accurate and quantitative description of macroscale models for electrochemical performance is required, including quantifying maximum C-rates, rate capacity and areal capacity. It is widely known that different properties are correlated with each other to some extent, sometimes even contrary. For example, areal capacity and rate performance are two conflicting properties, the areal capacity can be enhanced by increasing electrode thickness, resulting in reduced rate performance. Therefore, a trade-off must



Fig. 9. (a) Illustration for 3D characterization methods that commonly used in microstructure reconstruction. FIB-SEM represents focused-ion beam scanning electron microscopy, XCT represents X-ray computed tomography and NCT represents neutron computed tomography. (b) High resolution D image of LFP particle size distribution and Carbon particle size distribution. (b) Reproduced with permission ECS [161]. (c) Distribution of lithium concentration inside the electrode microstructure during discharge at 1C for 0.5 SOC. (c) Reproduced with permission from Elsevier [169].

be made and quantified, which requires massive experimental datasets and theoretical analysis to build an accurate model.

Since the electrochemical performance can be easily measured, many models are combined with data that is based on semi-empirical equations [172–175]. Park et al. reported a semi-empirical model to analyze capacity versus rate performance for various electrodes of different thicknesses [172]. The areal capacity and rate performance were quantitatively described with the low-rate areal capacity (Q_A) and the characteristic time of charge/discharge (τ) respectively. (Fig. 10(a)) A constant τ/Q_A is defined to represent the correlation between τ and Q_A by data fitting. They use the model to reveal the correlation between capacity and rate performance, providing a data-driven approach for the optimization of the capacity-rate trade-off, which can be obtained by increasing the volumetric capacity, electronic conductivity and electrode porosity. In addition, the model also implies that solid-state diffusion and reaction kinetics only make a significant difference to the performance of low areal capacity electrodes.

With regard to the rate performance in battery electrodes, Tian and coworkers fully quantified the limiting factors with three parameters from capacity versus rate data [173]. Considering the contribution of diffusion and reaction kinetics, the quantitative models specifically describe the rate performance by deriving the expression of capacity as a function of rate. (Fig. 10(b)) The core parameter that mainly limits the rate performance of the electrode is the characteristic time τ associated with charge/ discharge, which can be linked to several physical parameter

ters by different rate-limiting processes in the form of a second equation. The model shows good consistency with prior research and provides very helpful guidance for electrode design by identifying which rate-limiting processes are dominant under certain circumstances. Furthermore, Tian et al. modified the equation of the rate and capacity performance of battery electrode based on the chronoamperometry method, giving capacity as a function of charge/discharge rate, *R* [177]. Under high rates, they found a new decay feature in the curves, which can be associated with the early part of the current transient, indicating the relationship between high-rate performance and electrode conductivity.

Among all the parameters in the quantitative models for rate performance, the electronic conductivity plays a key role in electron transport process which severely affects the charge/discharge process. Therefore, the relationship between rate performance and electronic conductivity should be theoretically investigated. Tian et al. thoroughly researched the conductivity in electrode and derived a corresponding model for various electrodes, using different conductive additives such as carbon black, CNTs and graphene [174]. They found an extreme anisotropy in the electrode conductivity, with the out-of-plane (OOP) conductivity considerably smaller than the in-plane (IP) conductivity, perhaps due to network alignment effects. However, their research generally does not discuss which conductivity is more important, presumably this depends on the conducting path lengths in a given direction of the cell design, though the significant differences between OOP and IP conductivity can be attributed to the anisotropy of the films containing conductive net-



Fig. 10. (a) Areal capacity (Q/A) plotted as a function of rate for NMC/CNT electrode with a range of M/A-values. a) Reproduced with permission from Wiley [172]. (b) Specific capacity plotted as a function of rate using Equation given above panel. (b) Reproduced with permission from Springer Nature [173]. (c) Schematic presentation of the electrochemical reactions occurring in the anode during cell charging. (d) Comparison of degradation model results considering SEI growth and Li plating with experiment data in terms of variation of cell discharge capacity at different rates. (c), (d) Reproduced with Permission from Elsevier [189].

works. Through massive data analysis of the characteristic time versus conductivity, a nearly perfect fitting equation was established, implying that the rate capability can be substantially improved by increasing the electrode conductivity.

Apart from macroscale models describing the electrochemical behavior, developing safety models is also critical. Recently, the frequency of lethal EV accidents has gradually lowered the expectation of the reliability of EVs [91]. During their whole lifespan, LIBs will always go through some moderate degradation like capacity loss, mainly caused by loss of active material (LAM) and loss of lithium inventory (LLI) [[26],[179-181]]. Under some extreme conditions like nail-penetration tests and overheating, TR is likely to happen, resulting in catastrophic failure [91]. For now, there have been many researches about modelling for battery safety on a cell or pack level. With regard to the safety models for electrodes, more detailed mechanisms need to be accounted for. In Feng's review, TR mechanisms of the commercial lithium ion batteries are comprehensively summarized, containing three abuse conditions – mechanical abuse, electrical abuse and thermal abuse [182]. In recent years, Feng and coworkers have undertaken significant research to investigate the TR of batteries [[89],[183-187]]. In most cases, the catastrophic outcomes of TR originates from a cell level where ISC happens inside a single cell due to the penetration of the separator. The broken separator leads to the chain reactions inside the cell, finally resulting in an irreversible process of rising temperature and forming a Heat-Temperature-Reaction loop. However, Liu et al. reported an interesting cause of TR without ISC, named chemical crossover [184]. Their research shows that during TR, the oxygen released by the cathode is consumed by the lithiated anode, resulting in tremendous heat generation without a severe ISC.

Since this review mainly focuses on the electrodes, we discuss some representative safety models for electrodes. As mentioned before, Li plating is likely to happen on the anode surface during fast charging and low operating temperature, and the plated Li can react with the electrolyte to trigger TR . Li et al. reported an earlier TR for cells after fast charging with plated Li on the anode surface, which causes vigorous exothermic reactions [89]. As an important concern for electrode stability, especially under fast charging, many models describing Li plating have been created in recent years [188–190]. Wang et al. investi-

gated the transition from linear to nonlinear aging and reported a corresponding aging prediction model considering the SEI growth and Li plating,which can successfully match the aging curve [189]. (Fig. 10(d)) Fig. 10(c) illustrates the Li plating mechanisms occurring on the anode surface. Their research also found that the SEI growth is mainly responsible for the linear period of the capacity loss, while the nonlinear period, is mostly attributed to the increasing Li plating rate. However, while this model combined both SEI and Li plating mechanisms, a true coupling between the two was not presented, whereby freshly formed lithium is likely to form a fresh SEI. Some researchers also discussed the fact that Li plating is accompanied with a stripping process. Recently, Jossen and coworkers reported a P2D model considering both Li plating and stripping process [190]. The simulation shows excellent consistency with the experimental data about the characteristic discharging voltage plateau caused by Li stripping.

4. Multiscale Electrode Design Based on the Cyber Hierarchy and Interactional Network Methodology

As discussed above, significant progress in experimentally exploring electrode modification methods have been made in recent years, with the corresponding models of the electrodes pushing current electrode designs to their theoretical limits of energy density. However, despite this, some underlying mechanisms and the accuracy of electrochemical models still need to be improved. With the vast improvements in artificial intelligence (AI) technology, some researchers have investigated some novel methods to optimize battery electrodes. This requires high computational power to run complicated models to simulate the electrode, however fewer, or perhaps even no, experiments are then needed. In this section, a novel hybrid electrode design framework named cyber hierarchy and interactional network based multiscale electrode design (CHAIN-MED) is presented, along with its platform infrastructure and application instances.

4.1. Digitalization Trends in the Battery Industry

At present, the digitalization trends in the battery industry have gradually attracted more attention in both academic and industrial



Fig. 11. CHAIN: Multiscale Processing from Battery Materials to System Control. Reproduced with permission from CellPress [207].

fields. Apart from physics-based models, with increasing developments in the field of big data and AI, data-driven approaches have been reported to provide a promising pathway to next-generation battery design, which leverages off cloud computing capabilities, including emerging material developing, fast charging strategy, state estimation, smart manufacturing [191-200]. Attia et al., for instance, reported a significant breakthrough in the optimization of fast charging strategies with machine learning (ML) based on past experimental data [191]. An earlyprediction model was established to reduce the time per experiment, which was combined with a Bayesian optimization algorithm to obtain a balanced charging protocol. The automatic closed-looped optimization of fast charge protocols showed excellent improvement in the electrochemical performance and extraordinary reduction of the experimental time. Wanner et al. thoroughly reviewed the quality modelling in battery manufacturing using soft sensoring and sensor fusion, both of which can help to overcome thoroughly restrictions imposed by a limited number of hardware sensors [201].

ML based approaches also have shown great potentials in material science for property prediction and new materials discovery due to the excellent improvement in both time efficiency and prediction accuracy [202-206]. In Butler's review [195], the recent progress of ML in the chemical sciences as well as the potential suitable fields were summarized. Here they suggested that the future of chemical and material discovery can be substantially enhanced with AI techniques. Liu et al. thoroughly reviewed ML assisted materials design and discovery for rechargeable batteries, focusing on the feature engineering in material science and applications of ML in rechargeable battery materials science [204]. They also brought up the multiscale computation methods in lithium-ion battery research and development, ranging from electron to pack level, providing a comprehensive summary about the multiscale modelling and simulation for LIBs. [205] Furthermore, in order to address the tedious hyperparameter tuning that current feature selection algorithms usually require, they proposed a data-driven multi-layer feature selection method incorporating domain expert knowledge without manual tuning of the hyperparameters, showing highly improved efficiency while ensuring equal or better prediction accuracy [206].

Our group recently presented a novel concept named cyber hierarchy and interactional network (CHAIN), which is a general management framework for multiscale design and manufacturing process from the material level to battery system level [207]. (Fig. 11) The CHAIN framework provides novel insights through cloud-based models, enabling the simultaneous design and optimization process for research and development users. Further, perspectives about battery digital twins, which basically focuses on the fusion of models, data and AI for smart battery management systems were later presented, focusing on specific diagnostic data types that can be used in such a cyber-physical framework [208]. Despite the fact that CHAIN and battery digital twins share a similar concept which is mapping relationships between physical and virtual world, CHAIN actually offers a more comprehensive framework for different application scenarios of multiple scales, such as material synthesis, battery manufacturing and battery management systems. Therefore, as an important and irreplaceable methodology of CHAIN, battery digital twins certainly provide some useful cyber-physical elements for the hierarchical structure of the CHAIN framework.

Inspired by this methodology, the process of electrode design and manufacturing can also be optimized simultaneously with this modelbased and data-driven approach. Therefore, in order to meet the growing demand for integrated and intelligent design, we present a new generation of electrode design framework: CHAIN-MED, which consists of model-based and data-driven methods run on cloud servers to optimize the design process for battery electrode and share information between length scales. The concept of CHAIN-MED consists of two major components - multiscale digital models and a research database, all of which can be allocated on the cloud servers.

4.2. Framework for CHAIN-MED

Fig. 12 shows the hierarchical framework of CHAIN-MED, including a hardware layer, function layer and decision layer.

4.2.1. Hardware Layer

The cloud servers provide fundamental support as the hardware layer integrated with the function layer, which includes the multiscale digital models and research database. Hardware layer functions as physical storage devices as well as cloud computing platform with massive computing capability. The ideal configuration of hardware layer is distributed cloud servers, which can handle the task for data security and cloud computing. During the operation of CHAIN-MED, massive highdimensional data can be generated in real-time, which requires highperformance computing platform to simultaneously perform data processing (including data pre-processing and post-processing) and storage devices to categorize and store these operation data. At the same time, the model parameters are uploaded to the cloud servers to update the digital models, resulting in simulation outcomes as outputs for the research database. Combined with the model outcomes and guidance provided by the research database, a hybrid optimization for electrode design can be obtained and then put into another round of model iteration as inputs, eventually achieving a closed-loop optimization of electrode design.

4.2.2. Function Layer

As can be seen in Fig. 12, the function layer embedded in the cloud servers dominates the electrode design process, containing two func-



Fig. 12. Hierarchical processing framework of cyber hierarchy and interactional network based multiscale electrode design.

Nano-micro-macro scale: Reproduced with permission from Springer Nature [224]. Reproduced with permission from Wiley [172]. Model parameters: Reproduced with permission from Elsevier [93]. Simulation outcomes: Reproduced with permission from Elsevier [169]. Material synthesis: Reproduced with permission from Wiley [141]. Multiscale model: Reproduced with permission from The Electrochemical Society [225].

tional components: multiscale models and multiple database. Multiscale digital models are the key components of the novel framework, which represent the whole working process of the electrode design system. The embedded multiscale models can be narrowed down to several lengths, ranging from nano-micro-macro scale to manufacturing scale. Generally, the working process of multiscale models are sequential from nano-micro-macro model simulations to the final manufacturing process, outputting a virtually optimized electrode for the researchers.

Nanoscale models usually involves molecular dynamics and some other processes that happen within nanometers. Microscale structural models which correspond to the literature mentioned in section 3.1 are enabled by different 3D reconstruction characterization methods like XCT and FIB-SEM. These structural models help to capture the complexities of real microstructures as well as provide microscopic parameters like porosity and tortuosity. These microscopic parameters can be utilized in the macroscale functional models mentioned in section 3.2 like the semi-empirical model of capacity versus rate performance for various electrodes of different thicknesses, forming the connections between microscale models between macroscale models. In addition, the provided microscopic parameters are also fundamental inputs of manufacturing models for electrode manufacturing enabled by battery digital twins. This can be another necessary requirement for original equipment manufacturer to further improve and optimize the electrode manufacturing. Macroscale functional models corresponding to section 3.2 generally can be described as quantitative descriptions of external performance including electrochemical performance like areal capacity, maximum C-rate and safety performance like the likelihood of thermal runaway. For example, the models for Li plating highlighted in section 3.2 are of great importance for early prediction and detection of Li plating on the anode surface, which is critical for the macroscale safety performance for electrode.

As discussed in Chapter 3, conventional electrode modelling methods are still a crucial step in the electrode design process, building a theoretical foundation and providing guidance for actual experiments. Despite all the achievements in conventional electrode modelling, CHAIN-MED still requires highly predictable and precise models that reflect the manufacturing process. At the stage of manufacturing, the dynamic manufacturing execution process requires to be monitored and managed by collecting and uploading the attributes, performance, parameters, and process conditions of production factors. With origins from the designs for spacecrafts, a digital twin can be well suited to fill this gap in electrode manufacturing [209-211]. As for the concept of smart manufacturing, it comprises the real-time understanding, reasoning, planning and management of all aspects of manufacturing processes, facilitated by the pervasive use of advanced sensor-based data analytics, modeling, and simulation [212]. Digital twins consist of physical and digital representations of a system which are linked with real-time data, which means the whole manufacturing process can be abstracted into digital models and these two kinds of models evolve and affect each other at the same time, outputting optimized outcomes for electrode designs and manufacturing. Every processing step can be abstracted to a set of models which reflect the external performance and internal electrochemical mechanisms of the battery cell.

The multiple database also involves various functions like reclaiming literature, storing literature and simulation datasets in the cloud servers. Chapter 2 has thoroughly reviewed the electrode designs for conventional and emerging battery chemistries. A research database can be seen as the storage container for existing and previous data, including the design parameters, experimental/simulated outcomes, manufacturing parameters and other digital information. All of the previous literature for electrode designs can be uniformly added to the current research database for further researches. For example, additive and subtractive designs for fabricating low-tortuosity electrode mentioned in section 2.1 are abstracted into high-dimensional datasets. In the case of 3D printing nozzles used in fabricating nanocrystal cathode, parameters like nozzle speed and dispenser pressure as well as the electrochemical performance like capacity can be packed into specific data format, which can be further dimensionally compressed and stored in the research database. If later researchers apply similar fabrication methods, the multiple database can retrieve those data and offer some guidance based on former experiments. However, the information reclaimed from literature sometimes is not unified, which requires the support from big data techniques to perform data representation like feature extraction. In order to fulfill different tasks for unique demands, raw data of various

forms may require pre-processing and converted to hierarchical structures first, where the amounts can be narrowed down with the spurious data identified and eliminated. After unifying these datasets into a general format and extracting some characteristic features, machine learning approaches like DNN can be performed combined with model-based approaches, offering an optimized electrode design for the researches. More importantly, as shown in Fig. 12, the multiscale models and multiple database are constantly iterating and upgrading by interactional optimization between them when more knowledge are contributed to the database.

In addition, the review process of previous literature usually takes significant time depending on the technology. This can be easily accessible to researchers if the information is unified in a single database. Currently, there are several databases like the Inorganic Crystal Structure Database (ICSD), containing numerous material data for online application. Table 1 summarized the public structure and property databases for material science, all of which can offer specific knowledge depending on certain categories. However, there inevitably exists some human and other errors in databases. Therefore, in order to achieve accuracy and efficient ML models, training data may require labeling with predictions of input data to enable supervised learning with acceptable fidelity. In the case of unsupervised learning, only input training data is required to predict the trend. As long as the pre-processing is completed, accessible data can be unified into a general data format to perform the ML, which is often referred as data representation. The quality of data highly impacts the effect of ML model and the usability of a database. The selection of a suitable ML algorithm depends on many different factors. For example, the data may be discrete or continuous, requiring classification and regression models respectively.

There are now many advanced ML algorithms which can be applied in the battery field including: naïve Bayes classifiers, decision trees and neural networks, each of which is appropriate for different application scenarios. Table 2 shows the typical machine learning algorithms that can be applied in material science or other related fields, including some brief descriptions about advantages and disadvantages [195]. Neutral networks, especially deep neural networks (DNN), represent one kind of method that simulate the operation of brain, with neurons, input data, hidden layers and output data. Nowadays, DNN has been proven to be extremely effective in state estimation like SOC [194]. The ML model of DNN usually contain several neurons which are connected layer by layer, and each of these connections have weights, which represent the hidden information of this network. The process of model training is referred to the adjustment of these weights and other super-parameters by massive training data.

4.2.3. Decision Layer

Decision layer is the specific object in CHAIN depending on the field. Inherited from CHAIN, decision layer of CHAIN-MED represents the final optimized electrode design. Decision layer are fulfilled by the function layer. Based on the current knowledge, the hybrid model-data driven approach was utilized to achieve the optimized results (highperformance electrode designs), including the electrochemical performance prediction, battery chemistries decision, new material discovery and other aspects related to electrode design.

4.3. Optimization of Electrode Design Using CHAIN-MED

With the guidance of the CHAIN-MED framework, many applications are becoming realistic and possible. For example, material synthesis can be effectively guided by the cloud platforms with material databases and digital models, and emerging battery chemistry techniques can be presimulated with theoretical outcomes. This section focuses on the main aspect of the CHAIN-MED, which is the optimization of the electrode design process using the hybrid model-data driven approach.

Fig. 13 shows the dynamic relationships between external performance, multiscale models and electrode design. Customers are more fo-

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Database		Category	Storage content	Data sources	URL	
ChemSpide	L	All	Royal Society of Chemistry	Experiment	https://chemspider.com	
ChEMBL		Bioactive molecules	Drug-like properties		https://www.ebi.ac.uk/chembl	
COD	Crystallography Open Database	All	Organic, inorganic, metal-organic compounds and minerals		http://crystallography.net	
ICSD	inorganic crystal structure database	Inorganic crystal	Structure		cds.dl.ac.uk/cds/datasets/crys/icsd/llicsd.html	
CSD	Cambridge Structure Database	Organic crystal	properties		www.ccdc.cam.ac.uk/pages/Home.aspx	
PFD	Pauling file database	Inorganic crystal	Constitution, structure, powder diffraction data, and physical		paulingfile.com	
MP	Materials Project	All	Structure and energy properties	Computation	www.materialsproject.org	
OQMD	Open Quantum Materials Database		Structural, thermodynamic properties, etc.		https://oqmd.org	
CMR	Computational Materials Repository		Physical, chemical properties, etc.		https://cmr.fysik.dtu.dk/	
NOMAD	Novel Materials Discovery		Structural, electronic properties, etc		https://nomad-coe.eu/	
NIST			Chemical, physical properties, etc.		http://webbook.nist.gov	
AFLOWLIB		Alloy	Electronic structure, electromagnetic properties, etc		https://aflowlib.org	
HCEP	Harvard Clean Energy Project	Organic solar cell material	Electronic structure, Molecular information		cepdb.molecularspace.org	
ESP	Electronic Structure Project	Inorganic compound	Electronic structure properties		http://gurka.fysik.uu.se/ESP/	
GDB		Organic	Hypothetical small		http://gdb.unibe.ch/downloads	
ZINC		Molecules	2D and 3D formats		https://zinc15.docking.org	
NREL Mater	rials Database	Renewable-energy applications	Computed properties of materials		https://materials.nrel.gov	
TEDesignLa	р	All	thermoelectric materials		http://tedesignlab.org	

Table 2

List for typical machine learning algorithms.

Type of machine learning	Algorithm	Category	Description
Supervised	Support vector machines	Classification	Suitable for high-dimensional problems; Hard to search for proper kernel functions:
	Naïve-Bayes	Classification	Suitable for multiclass classification; Sensitive to input data;
	K-nearest neighbors	Classification	Convenient; Unsuitable for large sample size;
	Logistic regression	Classification	Easy to interpret; Underfitting;
	Linear regression	Regression	Intuitive; Poor robustness;
	Decision trees	Regression& Classification	Low computational burden; Overfitting;
	Random forests	Regression& Classification	Suitable for high-dimensional problems; Sometimes overfitting;
	Neural networks	Regression& Classification	Excellent performance in different aspects; Sometimes overfitting;
Unsupervised	 K-means	 Clustering	 Widely used clustering method; Hard to choose the k value;
	Principal component analysis	Dimensionality reduction	Low computational burden; Limit for the matrix;
	Apriori	Association analysis	Suitable for sparse datasets; Too many scanning for database;
Semi- supervised	Semi-supervised support vector machine	 Classification	 Improved learning performance by training unlabeled data; High time and space complexity
Reinforcement	Q-learning	Off-policy	 Wide range of datasets; High time and space complexity when O-table is large;
	Deep-Q-Network	Off-policy	Combination of Q-learning and neural networks;
	State-Action-Reward-State- Action	On-policy	Safer than Q-learning;

cused on the performance of battery systems, such as rate performance, allowable temperature, and cycle lifespan. For researchers, the design process seems to be more necessary, including the choice of anode, cathode, and modifications that can improve the performance of battery. Multiscale models, which are enhanced by multiple database, provides a hybrid approach for iterative optimization of electrode design.

The underlying mechanisms between external performance and cell design are a set of semi-empirical equations or models. In the case of the electrode, it can be the P2D model with physical parameters like thickness and porosity, and electrochemical parameters like ionic conductivity. Previous studies have shown great potential in model-based optimization of electrode design [[213],[214]]. De et al. reported a model-based procedure for simultaneous optimization of porous electrode parameters [213]. The thickness and porosity of both anode and cathode were selected to be the dependent variables, and the optimization of these four parameters were conducted in serial, each of which requires corresponding model simulation and simultaneous optimization. The whole optimization process requires multiple model simulation and optimization loops. Based on control vector parameterization (CVP) of optimization theory and a reformulated P2D electrochemical model, the optimization procedure achieved good outcomes with reasonable improvement by nearly 2% in energy density. In addition, the study can be extended towards optimization of cell design, such as minimizing the temperature gradient and cell configuration for higher energy density. Different from the typical models discussed above, designs of experiments (DoE) also provides a collection of statistical tools that can derive robust and trustworthy empirical models from limited experiments. Based on DoE, Rynne et al. found strong descriptive correlations between electrode formulation and different limitations, and successfully optimized the electrode formulation which topped typical formulations reported in the literature [215].

In addition to the mechanistic models, ML models also have proven to be effective in predicting physical parameters as well as electrochemical characteristics [216-223]. Based on experiments from the literature, Sendek et al. developed a data-driven model using logistic regression to classify ionic conductivity and identify the candidate structure that exhibits fast lithium conduction [217]. Their study also showed that a multi-descriptor model can offer a useful degree of predictive power while simple atomistic descriptor functions cannot. Wang and coworkers proposed a new solid polymer electrolyte design that integrates coarse-grained molecular dynamics (CGMD) with machine learning, obtaining comprehensive relationships between the lithium conductivity and intrinsic properties at the molecular level [216]. For ionic liquids (ILs), Gharagheizi et al. applied a least square support vector machine (LSSVM) model, which was trained by a database comprising 977 experimental data for 54 ILs, to represent and successfully predict the electrical conductivity of ILs [222]. In order to investigate the cathode delithiation process, Eremin and coworkers combined topological analysis, density functional theory (DFT) modeling, operando neutron diffraction, and machine learning algorithms to perform a comparative analysis of Li(Ni, Co, Al)O₂ cathode materials [221]. By applying the machine learning approach, they found that topology of Li layers and



Fig. 13. Dynamic relations among external performance, multiscale models and electrode design applying CHAIN-MED.

relative disposition of Li and Al in $LiNi_{0.8}Co_{0.15}A_{10.05}O_2$ (NCA) structure play the most significant descriptors during the energy balance estimations. As for the anode, Parthiban et al. developed an artificial neural networks (ANN) to predict the electrochemical performance of capacity and verified it with experimental data [220].

On the macroscale level, battery charging and rate performance as functions of both applied current and cell temperature, can be correlated towards 'dynamic mapping'. The relation between external performance and internal mechanisms can be predicted by the hybrid model-data driven approach offered by CHAIN-MED. The mapping relationships are simulated by multiscale models as well as trained by AI approaches, both of which can be generated and uploaded simultaneously. In addition, this kind of dynamic mapping actually could offer mutual guidance for improving the accuracy of hybrid model-data driven approach as well.

The conventional process of electrode design is quite timeconsuming, which requires countless experiments for synthesizing the material and testing the performance repeatedly. However, based on CHAIN-MED, the whole design process can be simulated and accelerated by adjusting the model parameters and applying the hybrid approach automatically before carrying out actual experiment. With the assistance of large databases where the existing researches are collected, combined with AI technology, eventually the electrode design can be optimized by iterative calculation, providing firm guidance for the electrode design process. For example, the rate performance of a battery cell is strongly correlated to the parameters of electrode models, mostly the thickness. A set of model parameters like porosity and thickness can be classified as input parameters, and the rate performance like the maximum charge/discharge rate can be chosen as output data. Through several layers of neural network, which can be well trained by abundant data collected in the database of cloud servers, the maximum charge/discharge rate can be predicted and compared with the experimental data. Therefore, rounds of iterative calculation deliver the optimized outcomes to inform the optimal electrode design. Finally, CHAIN-MED offers optimized optimal electrode design including the battery chemistries, design parameters, as well as the performance prediction of this design as guidance for researches.

Unfortunately, before fully understanding the intrinsic mechanisms and integrating all of these resources required to construct the whole platform, the methodology of CHAIN-MED is still quite conceptional for now. One day in the future, CHAIN-MED can provide a promising prototype for next-generation high-performance electrode design, and hopefully, this cloud-based optimization methodology can be extended to other energy storage fields.

5. Summary and Perspectives

Over the last 20 years, during the commercialization of LIBs technology, batteries have gone through several huge breakthroughs for higher specific energy, longer cycle lifespan and better safety, however conventional designs are now approaching their theoretical limits of energy density. Electrodes are the main components in a battery, enabling the intercalation of Li into the active material. Proper design of the electrode can significantly affect the electrochemical performance of the battery cell, even the whole battery system.

For now, significant amounts of research have been conducted to optimize and explore next-generation electrode designs for higher energy density, rate capability and better safety. The goals of high energy density and power density have become two major directions, with different methods like thick electrode design and aligned structures to improve the electrode rate performance. It may be difficult to determine which is the ideal solution for now. Therefore, understanding and quantifying the trade-off between thicker electrodes and rate performance has become a challenge for researchers. Herein, this paper presents a novel multiscale electrode design framework named CHAIN-MED, which mostly relies on the cloud servers to optimize the design process for next-generation electrode.

Many researchers have investigated novel design methods to improve the performance of LIB electrodes, which generally can be categorized as the conventional design strategies and emerging techniques for next-generation energy devices. Thick electrodes with low-tortuosity design have proven to be an effective method to improve intercalation kinetics for higher areal capacity and better rate performance. Binderfree and 3D current collector designs provide a novel insight of constructing a conductive 3D scaffold for electrode using 2D materials or other conductive additives, enabling fast electron transfer and higher specific energy density. In addition, numerous formulas for electrolyte additive and interfacial design have been investigated for better performance of mass transfer and ions migration.

Emerging techniques for next-generation energy devices recently have shown great potential in replacing the conventional electrode design. Li metal anodes exhibits the lowest electrochemical potential and the highest theoretical capacity among all the anode selections, resulting in a promising candidate for future anode choices. As safety concerns continue to be important, solid-state electrolyte based batteries offer a promising direction in preventing ISC and TR, especially under fast charging conditions with high current density and internal temperature. For other battery chemistries like Li-Air/Li-S batteries, some corresponding electrode design have been conducted, showing excellent improvement in electrochemical performance. Under the framework of CHAIN-MED, all of these existing and previous experiment cases and design methods can be stored in the cloud servers for future researchers.

Digital electrode models play an irreplaceable role in the CHAIN-MED framework, which generally can be categorized as the microscale modelling and macroscale modelling. Unlike conventional electrochemical models such as P2D model, microscopic models mainly use XCT and other 3D reconstruction techniques to investigate the local physical or electrochemical properties of electrode, such as porosity and tortuosity, all of which have been recognized as essential factors for macro electrochemical performance. Based on previous experimental data, macroscale modelling methods enable quantitative analysis of external electrochemical performance by semiempirical equations, such as rate performance and areal capacity for the electrode.

The CHAIN-MED framework offers researchers a hybrid solution with iterative design optimization based on model simulations as well as dynamic mapping between external performance and multiscale model. Combined with model-based and data-driven techniques, the research and development process of next-generation electrode can be substantially accelerated under the theoretical guidance of CHAIN-MED, freeing researchers from time-consuming trial and error.

Nevertheless, there still exists several technical challenges for CHAIN-MED from practical application.

- (1) There has not been sufficient research around mechanistic models especially for electrodes. In order to meet the need of high accuracy for model simulation, more sophisticated and implementable multiscale models are required to understand the underlying mechanisms for further application in the cloud platform.
- (2) The working process of CHAIN-MED involves sophisticated calculation and data analysis. As the size of the system increases, the computations required grow dramatically in complexity. During the whole data mining process, huge amounts of real-time data are generated and stored in the cloud servers, requiring highly effective computational resources and huge storage hardware to ensure the stability and sustainability.
- (3) The analysis of microstructures and their linkages to their manufacturing conditions involve nondestructive detection methods and tools. Therefore, highly accurate and sensitive wireless sensors capable of detecting microstructural changes are urgently required for real-time sampling and modelling for electrode design and online diagnosis of flaws that may arise from manufacturing process.

Declaration of Competing Interest

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

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