

# Intelligence-assisted predesign for sustainable recycling of lithium-ion batteries and beyond

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The unprecedented consumption of lithium-ion batteries (LIBs) is underway to meet the needs of modern transportation electrification. Recycling-friendly designs intrinsically facilitate the long-term sustainable utilization of natural resources, reduces the detrimental impacts of spent LIBs on the environment, and improves the economic viability of recycling. For this, we propose the incorporation of an intelligence-assisted predesign strategy into the battery management system and battery chemistry, including ameliorating traceability of battery structure and operation status, promoting cost-effective material sorting and separation, and adoption of sustainable battery materials. Purposeful predesign of intelligent “Reuse-Refurbish-Recover” (3R) LIBs could be realized in an economically and environmentally sustainable (EES) manner. The intelligence-assisted predesign strategy will promote multi-disciplinary cooperation in a systematic and synergetic way, and empower government to establish and implement a series of regulations and standards to address the environmental pollution issues from spent batteries and avoid the ever-increasing consumption of natural resources.

## Introduction

In recent decades, lithium-ion batteries (LIBs) have played a major role in modern society and industries, from a power supply for electronic devices to large-scale energy storage management in smart grids. The production scale of LIBs is expected to surpass 1.3 TWh in 2030.<sup>1</sup> The massive production and consumption of LIBs demand a strong and continuous supply of various natural resources. By 2018, the consumption of lithium for batteries reached 270,000 metric tons of Lithium Carbonate Equivalent, while over half of Cobalt produced in the world has been used in battery production.<sup>2</sup> Under such circumstances, effective management of spent LIBs are urgently needed to help reclaim critical metal resources, alleviate the shortage of natural resources, and lenify environmental pollutions from the mining of raw materials, culminating in the achievement of economically and environmentally sustainable (EES) recycling.<sup>3</sup>

Currently, the recycling of spent LIBs faces rigorous challenges. Unlike the successful and cost-effective recycling of spent lead-acid batteries (LABs), the current recycling percentage, efficiency, and profitability of spent LIBs are far from satisfactory. This can be attributed to their poor full life cycle management and chaotic battery construction. Though certain economic returns can be achieved through metallurgy-based recycling (i.e. MBR, namely pyrometallurgy and hydrometallurgy) on account of the high market value of transition metals (e.g., Co),<sup>4</sup> the high energy and chemicals consumption inevitably leads to significant environmental footprints. More recently, human-assisted direct recycling

(HADR), consisting of a human-assisted pre-treatment and a chemical regeneration process, has been proposed in repairing the compositional and structural deficiencies of spent electroactive materials and regenerating them into new battery materials with minimum energy and chemicals inputs. At present, a financially viable commercial scale-up of the pre-treatment and material separation processes of HADR of LIBs has not been achieved. This is probably due to the main pitfalls of modern commercial lithium-based batteries: sophisticated chemical composition and poor traceability. To regenerate different electroactive materials, specific strategies are needed on account of their inconsistent phase transition, which entails significant human intervention and, therefore, high labour-cost in the sorting and disassembly.<sup>5, 6</sup> Moreover, obtaining pure electroactive materials for regeneration requires intricate separation. These challenges impede the commercial applications of HADR.

Fundamentally, the high cost and environmental footprint of current recycling processes largely stem from the fact that modern LIBs are not designed for post-use recycling.<sup>7</sup> A lack of consideration for life cycle management and recycling-unfriendly design means that LIBs cannot presently be recycled in an economically and environmentally sustainable manner. From the ground-breaking development of intelligent technologies such as Artificial Intelligence (AI) and Machine Learning (ML), the current battery management system is expected to gain substantial improvements in terms of battery health diagnosis and prediction and automatic battery cells sorting, and disassembly. There is room for improvement in the intrinsic recyclability and sustainability of batteries, especially in light of recently developed battery materials and novel battery structures.

From the aforementioned challenges of current LIBs recycling technology, it is time to systematically revisit the cell manufacturing protocol and engineering management to re-design the future batteries for EES recycling. Specifically, intelligence-assisted predesign of LIBs, including well-

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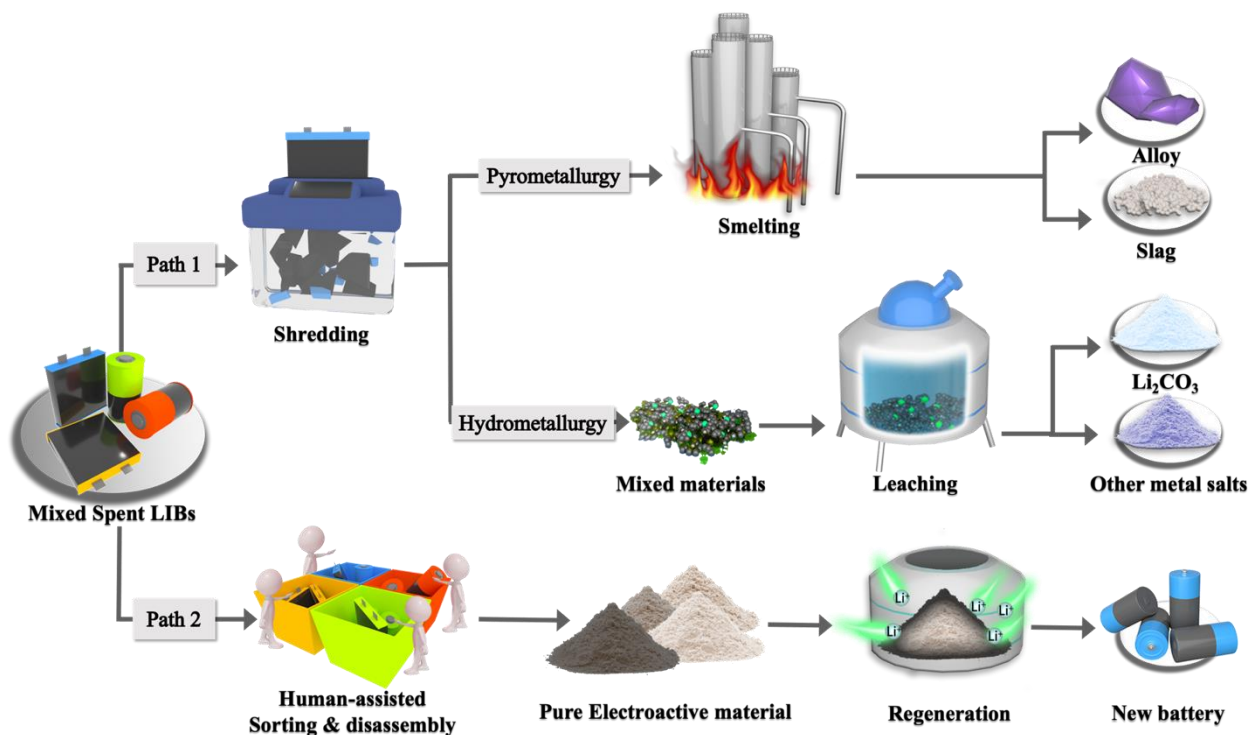
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**Fig. 1 State-of-the-art recycling processes for spent batteries:** Path 1: Metallurgy-based recycling (MBR), involving high energy consumption and high pollution production (but with low labor costs and selectivity). Path 2: Human-assisted direct recycling (HADR) - the environmentally friendly recycling process, including collection, manual sorting and disassembly, separation, regeneration, and new battery production (with high labor costs in the collection, sorting, and disassembly phases).

considered battery management system and battery chemical design, is proposed herein. Intelligence-guided “Reuse-Refurbish-Recover” strategies (hereinafter “3R” strategy) are, therefore, put forward in the EES recycling scenario. We will convey to readers the necessity of incorporating recycling into future battery design and provide a way of synergizing intelligent technologies and advanced materials for battery pre-design. We believe that our proposed strategies could significantly increase LIB recycling rates and realize a truly renewable and sustainable battery economy. Moreover, these strategies will empower government to establish robust regulations and standards to address the environmental issues and minimize the use of natural resources as we move to mass produce LIBs globally.

### State-of-the-art recycling technologies of LIBs

State-of-the-art recycling technologies for spent LIBs can be classified into two pathways, as illustrated in Fig. 1. Path 1 is the MBR process, while path 2 is the HADR process. Path 1 is commercially adopted in current LIBs recycling practices due to several reasons. Firstly, due to the complexity of LIBs, the adoption of existing metallurgical technology and relevant facilities demands less human intervention in sorting and separation; secondly, these technologies have less sensitivity to the impurity of the electrode materials; thirdly, they could be applied to a wide spectrum of electrode materials. By contrast, Path 2 demands a less intensive chemical process and, therefore, is more environmentally friendly. The widespread

adoption of Path 2 is highly challenging due to labor-intensive sorting requirements and identifying the LIB type, and the composition of the battery materials.

#### Metallurgy-based recycling (MBR)

To date, MBR has dominated the recycling market, which mainly aims at breaking cathode materials down into metal alloy or metal salts through pyrometallurgy or/and hydrometallurgy.<sup>8</sup> Pyrometallurgy (as depicted in Fig. 1), which handles the mixed batteries stream, involves a series of, sorting, disassembly, and melting processes, but at the expense of energy usage and associated greenhouse gases (GHG).<sup>9</sup> At high temperature, spent anode materials are burnt off, and Li species normally end up in slag along with the toxic organics released from the decomposition of electrolytes and binder.<sup>10</sup> Comparatively, hydrometallurgical recycling, as shown in Fig. 1, is also capable of processing mixed batteries stream but with ameliorated recycling efficiency and selectivity. For spent cathode materials, valuable metals are dissolved using strong acids and can be subsequently precipitated and recovered in the form of metal salts after intricate purification and separation processes.<sup>5</sup> Spent anode materials are separated from cathode materials and collected but further commercialized recycling processes remain unexplored. More information on MBR regarding the detailed technologies is out of the scope of this perspective and can be found in other works.<sup>10-12</sup> Still, hydrometallurgical recycling has certain environmental and economic limitations, including the large volume of wastewater produced, strong acid/alkali solution used, and the high number of chemicals required.<sup>5,11</sup>

Overall, MBR, including pyrometallurgy and hydrometallurgy in Path 1, has the main advantage of being able to cater to mixed batteries types at relatively low cost and with less human intervention. Nonetheless, its high energy consumption and associated GHG emissions, as well as difficult solid/liquid waste disposal requirements, detract from its environmental sustainability.

### **Human-assisted direct recycling (HADR)**

As depicted in Fig. 1 Path 2, HADR includes labor-intensive pre-treatment, material separation, and chemical regeneration processes for specific electroactive material. The first two steps concentrate high purity electroactive materials for regeneration.<sup>13</sup> Without breaking down the electroactive materials into their elemental state, the last step of HADR, namely regeneration, is a non-destructive process aiming at rejuvenating the Li deficiencies and structural damages in spent cathode materials and repair damage layer structure and remove impurities in spent anode materials with minimal energy and chemicals inputs.<sup>14</sup> Such processes are deemed more desirable as the decayed electrochemical performance caused by the impaired structure is replenished, and the regenerated electroactive materials can be directly used to prepare new batteries, thereby transferring waste into resources and reducing the environmental footprint of battery manufacturing. The regenerated cathode and anode material could deliver satisfactory electrochemical performance in battery setup.<sup>6, 15-18</sup> Compared with MBR, these processes require a lower reaction temperature and fewer chemicals. Short annealing processes are generally needed to improve crystallization further.

In an ideal situation, such a regeneration process will contribute to the carbon neutrality of battery manufacturing and recycling. However, academia mainly focuses on the optimization of chemical regeneration, while overlooking another imperative obstacle to its practical uses, that is, the high labour cost caused by ensuring purity during the labour-intensive pre-treatment and material separation. Therefore, the chemical regeneration process confronts considerable challenges before its commercial application. Firstly, regeneration targets a specific cathode and thus requiring specialized processes for different cathodes.<sup>15</sup> Facing the progressive variation of battery chemistry, regeneration places strict requirements on sorting. Currently, regeneration conducted on a lab scale normally adopts manual sorting, which is obviously labor-intensive and uneconomically viable for commercial upscaling.<sup>19</sup> Secondly, maintaining purity is the opportunity. The quality of the regenerated electroactive material is sensitive to trace doping elements and other impurities such as Cu and Al,<sup>20</sup> therefore manual disassembly and intricate material separation are necessary to help obtain pure electroactive materials. These two phenomena are actual reflections of material acquisition methods in lab-scale regeneration methods. We argue that this human-assisted manner is not economically viable in practice. Thirdly, the state-of-health (SOH) of electroactive materials is reported to have vital impacts on the efficacy. Unfortunately, at this stage, it is impossible to predict and obtain the SOH value of the

electroactive materials in today's LIBs. If not managed properly, recycling chaos will inexorably transpire. Lastly, the non-systematic regeneration mechanism will lead to the questionable quality of the regenerated materials in large-scale applications.<sup>21</sup> The effectiveness of the regeneration technology can be reinforced through in-depth research, but the cost-effective sorting and disassembly and the intractable SOH value will remain challenging.

### **Gaps and challenges in current battery recycling**

In the current battery recycling scenario, Path 1 in Fig. 1 is the most cost-effective choice, and therefore, it is preferentially selected to deal with the massive number of mixed spent batteries, but at the cost of adverse environmental impacts. The inherent drawbacks of high energy and chemicals consumption are difficult to overcome. Nonetheless, as previously discussed, the benefits of Path 2 in facilitating carbon neutrality and minimizing environmental footprints in manufacturing and recycling are clear. Regenerated electroactive materials have a maximized payoff to battery manufacturing and the environment.<sup>22</sup> Thus far, we argue, Path 2 would not grab the recycling market until its pitfalls mentioned above are handled. Optimizing regeneration technologies and diminishing human intervention are the two essential enablers of Path 2 commercialization. However, battery manufacturers prioritize the performance and profit without considering recycling, leading to poor management and recyclability of the battery itself. Therefore, the high cost for maintaining the purity for regeneration in sorting, disassembly, and material separation is hard to surmount based on today's battery design.

Additionally, the current recycling market adopting Path 1 is suspected of wasting resources. In the absence of a sound management system and unified recycling standards, batteries with remaining useful life (RUL) may be decomposed into substituent elements, which is a manifestation of the underutilization of batteries. A "Secondary Utilization" strategy has been proposed to reuse the retired LIBs from electric vehicles for energy storage grid and various other applications. For this reason, the relevant standards for measuring the service life of the battery need to be created, and the real-time traceability of the battery cells' and battery pack's condition over their full life cycle needs to be improved.

The current poor monitoring of the health status of battery materials and the limited traceability of related data, the unstandardized battery structure, the complex LIB composition, and various other challenges are preventing the establishment of an EES recycling system. Many of these challenges can be overcome by placing end-of-life (EOL) recycling as a core LIB product design objective.

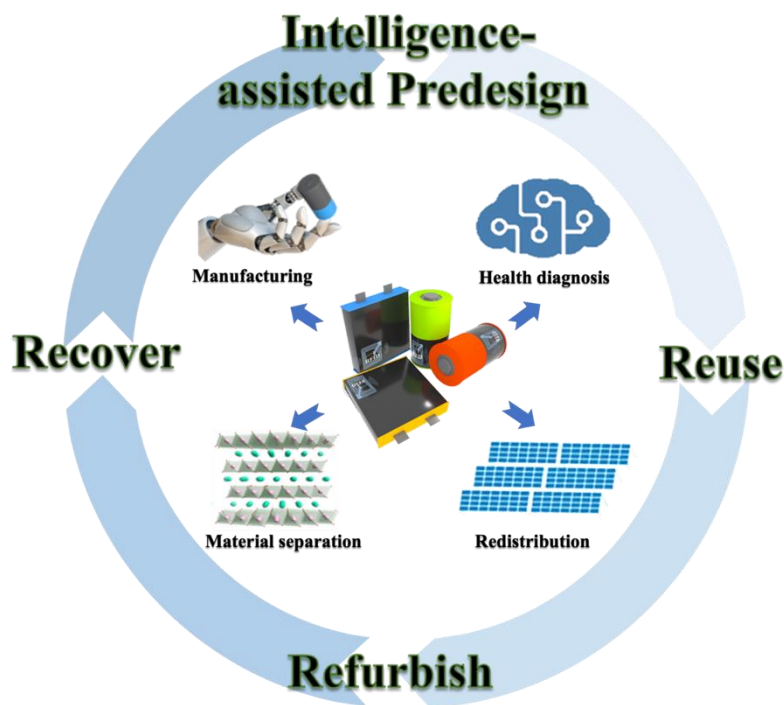
### **Strategies towards EES recycling**

The inherent recyclability and sustainability of batteries, including a full life cycle battery management system with high traceability and battery design with simplified structure and renewable materials, will be the cornerstone of the establishment of EES recycling. The former can help the orderly maintenance of batteries in use, refurbishment after use, and

regeneration at the EOL stage. The latter will promote simple material separation and enhance the sustainability of battery materials.

The complexity in the design of today's battery is a recognized challenge,<sup>12</sup> and the importance of design for

Scientific standards should be formulated to regulate the implementation of the "3R" strategy. Standardized health indicators, such as SOH and RUL, as well as the performance indicators for each step in the "3R" strategy, when evaluating the health condition, should be calibrated to help decision-



**Fig. 2 The intelligence-assisted predesign for "Reuse-Refurbish-Recover" in economically and environmentally sustainable (EES) recycling:** Intelligence-assisted predesign provides enablers for intelligence-guided health diagnosis, redistribution, sorting, and cost-effective material separation, which facilitate the "Reuse-Refurbish-Recover" to realize a closed-loop battery recycling and manufacturing scenario.

recycling has been argued.<sup>23</sup> In the past, some ideas such as applying the minimum number of battery components, standardizing the formats, and electroactive materials have been proposed to increase recycling.<sup>22, 23</sup> These ideas are rarely systematically embedded in the design and manufacture of batteries to guide industry and research. Consequently, these challenges remain unresolved and poor LIB recycling rates still prevail in society.

To this end, we envision a "3R" strategy based on the intelligence-assisted predesign of the battery management system and battery chemistry (see Fig. 2). Predesign will facilitate the intelligence-guided diagnosis, redistribution, sorting, and material separation to manage the battery in an orderly manner throughout its life cycle, thereby maintaining maximum economic value with the smallest environmental footprint in the circular battery economy. In the chain of the "3R" strategy, "Reuse" must ensure the safe operation of batteries. Malfunctioning constituents such as the damaged external protection circuits can be detected and maintained. "Refurbish" tackles the gradient utilization of retired batteries from high-demanding applications. "Recover" closes the loop by cost-effectively extracting waste electroactive materials, regenerating their structural defects, and providing regenerated electroactive materials for new battery manufacturing at a lower cost.

making. Normally, as a figure of merit of the condition of a battery, SOH is applied as the criteria for decision making not only because it implies the ability of the battery to provide a specified capacity and but also it affects the efficiency of regeneration. To illustrate a scenario, we envision the SOH as the criteria for decision-making (as illustrated in Fig. 2). For SOH within 80%, the battery is determined as before end-of-services and will be regulated through "Reuse" to prolong the safe operation. When the SOH value is lower than 80%, the battery reaches the end-of-services stage and will enter the "Refurbish" stage. Batteries with SOH values lower than 20% will enter the "Recover" stage for the chemical regeneration of electroactive materials.

To fulfil the goal of EES recycling, recycling-friendly designs need to be embedded into both the battery management system and the battery chemistry.

### Intelligence-assisted predesign

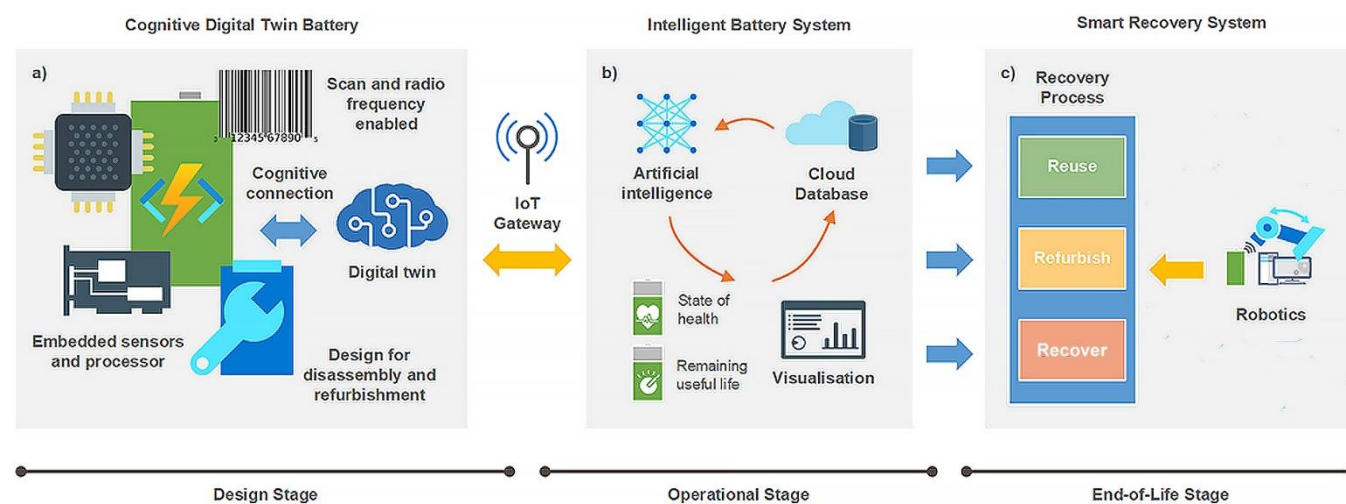
The intelligence-assisted predesign aims at resolving the challenges facing the commercialization of Path 2, consisting of the design of cognitive digital twin battery, intelligent battery system, material separation, and sustainable electrodes. These predesigns will be the essential enablers for the "3R" strategy proposed for EES recycling. The design of cognitive digital twin battery and the intelligent battery system integrates digital

twins, AI, Internet of Things (IoT), ML, and other high-tech into the design of the battery management system, which will improve the traceability of LIBs and establish a 'cradle-to-grave' online life-cycle database. Given the wide differences in utilization of LIB batteries and the associated variation in their health status, the creation of a predesigned battery management system is essential.<sup>12, 13</sup>

Predesign for material separation and sustainable electrodes will integrate advanced and renewable materials to improve the inherent recyclability and sustainability of the battery itself. Precisely, these designs will address how to open battery molds

battery features from the sensor and communicate with the cloud. Breakthroughs in nano-sensors demonstrate more possibility in developing built-in sensors such as Fibber Bragg Grating sensor for decoding the formation of solid electrolyte interphase and the evolution of crystal structure at cell level.<sup>30</sup>

Replacing the barcode on today's batteries with RFID will ensure that the recycling department can remotely record and continuously monitor the SOH value. Smart labeling and embedding information into the battery covering aspects such as electrochemistry materials, assembly procedure, and other pertinent information will facilitate fast sorting and disassembly



**Fig. 3 Intelligence-assisted predesign for ameliorated traceability and battery management system.** (a) Design of cognitive Digital Twin Battery with embedded IoT sensors, radio frequency identification (RFID) to extract and store health indicators value via digital twin; (b) Intelligent battery system gathers data transferred through IoT gateway to build cloud database for the health management via artificial intelligence; (c) Intelligence-guided decision making for the “3R” strategy.

and battery cells easily, how to separate electroactive materials in a green and effective manner, and how to achieve sustainable battery economy at the raw material level.

### Cognitive digital twin battery

The core of the cognitive digital twin battery lies in the real-time monitoring of the health state of batteries, which can be achieved through embedding an IoT sensor that interacts with the digital twin using radio frequency identification (RFID) or other communication protocols (Fig. 3a).<sup>24</sup>

The built-in sensor will be capable of decoding the chemical and thermal events in battery cells, networking with a digital twin, and enabling data to generate big global datasets covering a whole range of battery health-related information, such as charge/discharge profile, resistance, and open-circuit voltage (OCV), over their life cycle.<sup>25</sup> IoT has been popularly used to support smart cities,<sup>26, 27</sup> but also increasingly used for a smart waste management system.<sup>28</sup> To date, few studies are using IoT embedded sensors, software, and other technologies within physical objects to predict LIB's SOH. Kim and co-authors proposed the use of IoT sensors to measure cell voltage, current, and temperature that will be uploaded into the cloud and processed using ML.<sup>29</sup> Furthermore, a sensor called BMS-Slave consisting of multi-cell battery monitors to predict SOH and state of charge (SOC) was developed,<sup>24</sup> where the IoT component deployed Raspberry Pi is responsible for collecting

processes, and inform how the battery should be reused, refurbished, and recovered.<sup>31</sup> Nowakoski<sup>32</sup> has demonstrated the feasibility of establishing a cost-effective e-waste disassembly procedure through using RFID technology. However, more efforts should be committed to improving this concept since RFID presently only covers limited material data from suppliers.

Going forward, the growth in the number of batteries used for electric vehicles, electronic goods, etc., will be unprecedented. Manufacturers will expect transparency in the source tracing of each raw input material of a battery over its entire life cycle, including in any second life remanufactured or direct recovery operations. To this end, the cloud-based digital twin will facilitate ongoing tracking through IoT infrastructure, and the use of blockchains can enforce socially responsible supply chains and security.<sup>33</sup>

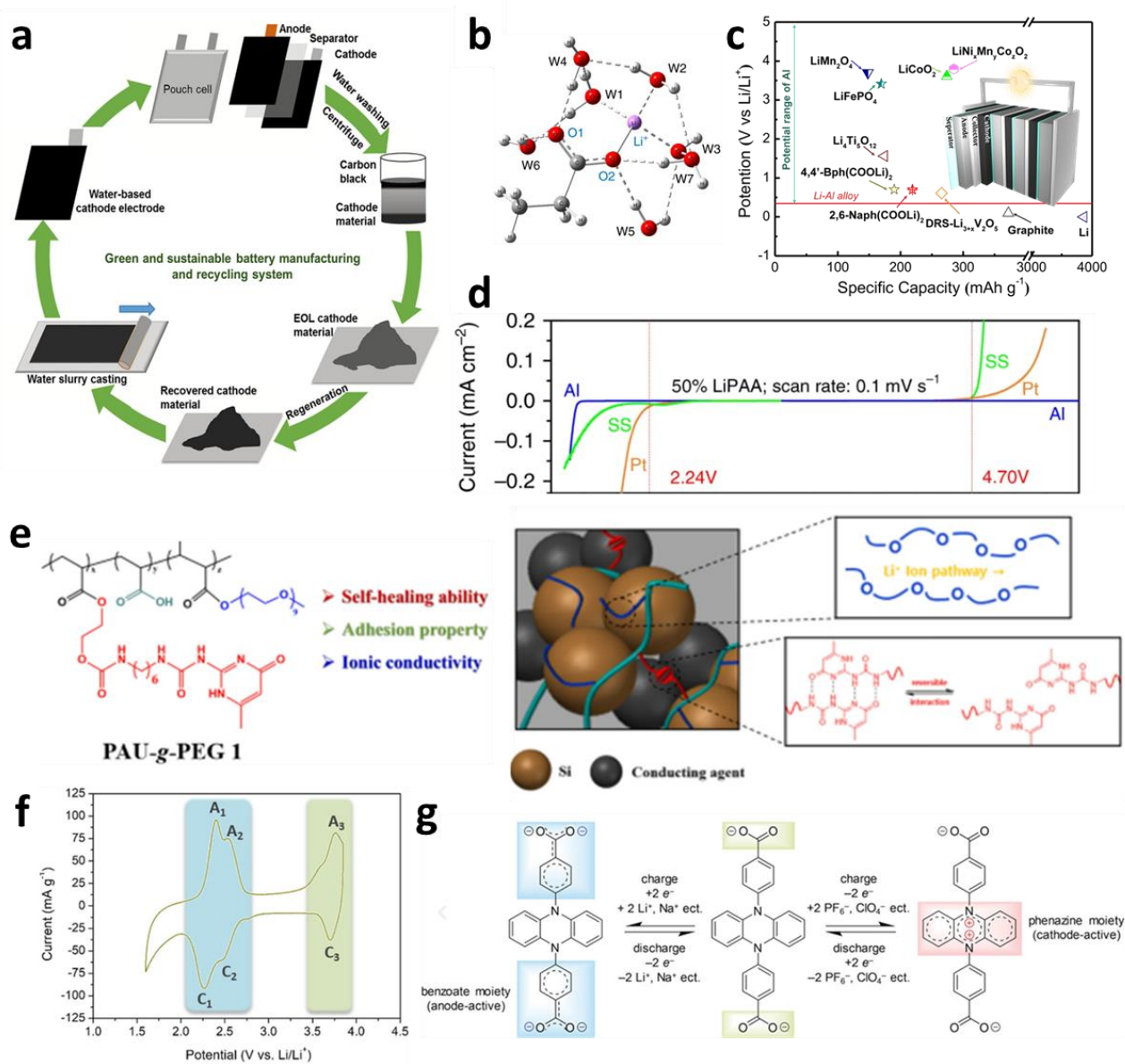
### Intelligent battery system

Intelligent battery systems connected with cognitive digital twin batteries through IoT gateway will create big global datasets covering a whole range of battery information over their life cycle, and AI technology can process and visualize the useful data for decision making (Fig. 3b).

Extracting health-related data is the first step. Utilizing the data to predict the value of health indicators (in this scenario, the SOH value) through AI will be the next stage. The embedded IoT

sensor, which interacts with a digital twin in a cognitive battery will transfer data through the IoT gateway, therefore creating a cloud database beneficial to real-time monitoring of battery health. Advanced algorithms play a vital role in fulfilling real-time predictions. Bayesian-based algorithms,<sup>34, 35</sup> Artificial Neural Network,<sup>36</sup> and Deep Learning<sup>25, 37, 38, 39</sup> have been used

or mathematic models to predict SOH, AI-assisted data-driven evaluation avoids the complexity of internal electrochemical reaction and failure mechanisms and therefore, dominated this research field recently.<sup>41, 42</sup> However, limited by current hardware and data storage technology, data-driven assessment is not widely applied. It is expected that SOH prediction could



**Fig. 4 Pre-design for electrode material separation.** (a) An ideal water-based recycling process, reprinted with permission from ref.65 Copyright 2020, Elsevier. (b) Binding order of water molecules with LiPAA and (d) the electrochemical stability windows of the 50 wt% LiPAA electrolytes measured on Pt, stainless steel (SS) and Al, reproduced with permission from ref.72. Copyright 2018, Nature publishing group. (c) Effective potential ranges of Al and promising electroactive materials for bipolar electrode structure (inset is the illustration of bipolar electrode structure). The data were collected from ref. 18 and ref.73-81. (e) Schematic illustrations of the polymeric binder PAU-g-PEG 1, with both reversible self-healing and Li<sup>+</sup>-conductive properties. Reprinted with permission from ref. 88. Copyright 2020, Nature publishing group. (f) CV at scan rate of 0.1 mV s<sup>-1</sup> showing the two redox active potentials of the homopolymer (PI1) synthesized using stoichiometric amounts of 3,7-diamino-methylphenothiazine (DAMPT) and naphthalene tetracarboxylic dianhydride (NTDA), reproduced with permission from ref.93. Copyright 2019, Wiley-VCH;<sup>91</sup> (g) Redox mechanism of the dual-ion 4,4'-(phenazine-5,10-diyl)dibenzonate (PZDB) anion. reproduced with permission from ref.94. Copyright 2019, Wiley-VCH.

in recent researches, providing valuable knowledge for developing intelligent battery systems. Normally, methods for SOH prediction can be categorized into four types, including model-based, data-driven, hybrid, and other assessments.<sup>40</sup> Compared to model evaluation, which employs electrochemical

be achieved by extracting simple features such as the charge/discharge profile and OCV. Follow this logic, the ampere-hour integral method or the OCV method, which calculates the battery capacity (or maximum usable capacity) within a certain period and then realizes the real-time

prediction of SOH through a series of control strategies, has been adopted in the industry.<sup>43-46</sup> Besides, due to the powerful real-time prediction function that only requires the voltage and charge/discharge capacity, breakthroughs in incremental capacity (IC) analysis are worthy of attention.<sup>44</sup> Hybrid assessments have complementary advantages and have achieved commercialization. For example, inaccuracy originated from environmental factors can be diminished through the combination of IC and data-driven evaluation,<sup>47</sup> and the combination of data-driven and model methods can help determine the model parameters.<sup>48</sup> It should be noted that breakthroughs in these technologies are remarkable, providing various choices for battery manufacturers. Despite this, there is still a big opportunity to improve existing research by improving prediction accuracy and economic viability. Battery health monitoring can also capitalize on Blockchain technology, which allows the IoT peer node to extract data from the battery and control the module and communicate with other nodes and cloud servers.<sup>49</sup> The combination of AI with IoT will ultimately lead to AI of things-based environment providing enhanced human-machine interactions and better decision making with respect to the complicated and dynamic battery system.<sup>50</sup>

With continuous cyber-physical updating from an array of IoT sensors with edge-processing capabilities, bigdata repositories can be set up, creating opportunities for real-time IC and data-driven analysis of SOH/SOC/RUL.<sup>41, 51</sup> Combined with RFID and cloud, such a system can achieve fully automated health prediction,<sup>52</sup> which will aid expand the life of existing battery fleets and guide the design and manufacture of new battery technologies. Over time, the collection of data repositories of both new and refurbished battery fleets will significantly leverage knowledge on a certain battery's full life cycle trajectory.

Cost-effective disassembly is the key aspect of EES recycling, and a centralized database of battery performance will offer significant cost savings to intelligence-guided sorting and disassembly.<sup>53</sup> Battery recycling cost-effectiveness is heavily dependent on the ability to automate the sorting and disassembly (Fig. 3c).<sup>14, 54</sup> Waste collection facilities must be able to read RFID tags of entering batteries that then enable authority for connection to cloud-based servers containing detailed information on the battery digital twin. Blockchain technologies can be used for secure authentication. After collection and automated sorting, the battery will enter the appropriate agile recycling chain, the digital twin will download precise disassembly instructions to robots that commence the disassembly process. Gradient utilization will be conducted during this process according to the SOH value of batteries. Battery disassembly robots are still in their infancy, but AI techniques and robotic technologies are advancing rapidly and will be able to conduct a range of critical tasks such as pack removal and disassembly, module removal, and cell separation.<sup>55</sup> To this end, some researches can be adapted to facilitate cost-effective sorting and disassembly. The reported digital-twin product-recovery framework for e-waste could be modified to spent battery recycling.<sup>56</sup> Cloud-based information repositories and RFID systems will be able to provide the necessary specifications for establishing cost-effective and

rapid sorting and disassembly. Ultimately, the processes related to recycling must use less energy than that necessary for mining.<sup>57</sup>

Overall, the cognitive digital twin battery and intelligent battery system provide SOH data for each battery cell, offering opportunities for intelligence-guided health diagnosis and gradient utilization. The recycling sector will also be able to identify the assembly and composition of the battery through reading the embedded information in the RFID. A cost-effective sorting and disassembly process for the following "Recover" stage is also established.

#### **Pre-design for electrode materials separation**

When it comes to the re-design of batteries for EES recycling, the first chemical design that needs to be addressed is how to access electroactive materials easily. The complex components and architecture of today's batteries place challenges in the opening of pack/cell and the separation of battery material. The standardization of the fixing type, fixing place, and glue used have been proposed to enable the fast pack opening. At the cell level, however, standardization is more challenging. How to achieve rapid cell opening troubles the recycling department. Thereby shredding and mechanical processes are predominantly adopted to avoid this issue. Rational material design will facilitate non-destructive disassembly to obtain pure electroactive materials.

Pre-design for electrode materials separation include the design of battery cell configuration and the use of smart materials, such as the materials with shape memory effect for the sealing, fixing, and packaging and for low-cost and rapid disassembly.<sup>58</sup> Pack and module design vary significantly, raising a recognized challenge for non-destructive disassembly.<sup>54</sup> Regardless of the number of cells and the connection configuration, cells in the module are permanently affixed and are not intended to serve individually. Standardization may partially alleviate the burden of recycling by automating dismantling. Endowing the pack and module an active disassembly ability might be more straightforward and promising. The manufacture of connection parts and the module are expected to apply shape memory polymer (SMP), which merits industrial viability, morphological versatility, and synthetic flexibility.<sup>59, 60</sup> SMP such as polyurethane, polyester, polysilamine, and some responsive hydrogels including poly(N-isopropyl acrylamide) (PNIPAAm) hydrogels, polythiophene gel can be easily adapted to develop reversible insulating rings and sealants for non-destructive pack and module opening.

Cell opening is another challenge facing the recycling sector. Shredding is a widely adopted method to tear down the robust battery cell casing, followed by a series of separation processes, such as seizing, froth flotation, magnetic separation, etc. The dilute concentration of valuable materials and the poor selectivity are difficult to overcome in such processes. A promising solution is designing a smart battery casing with robust mechanical strength but morphological versatility. Rapid development in lightweight shape memory alloy (SMA) such as Mg-based alloy<sup>61, 62</sup> can offer essential knowledge. Shape memory effects can be stimulated by temperature, light, electric field, and magnetic field.<sup>63</sup> As such, the outer casing and

constitutions can be feasibly separated from inner compositions. For pragmatic use, SMA or SMP should be carefully selected based on the products' requirements.<sup>58</sup> Unfortunately, current customer demand for higher performance and lower prices hamper the uptake of this technology.

Decoupling the electroactive materials from the current collector (CC) is expected to be accomplished in a more sustainable manner by EES recycling. Currently, heat treatment or organic solvents are applied to detach the electroactive materials. In a more sustainable scenario, the energy, chemicals inputs, and environmental footprints are expected to be significantly reduced. Substituting the organic solvents with the simplest and the most environmentally friendly solvent, which is water, is much more desirable. The development of water-soluble binders holds promise in fulfilling a water-based material separation processing line. Though the exploration of low-cost, safe, and environmentally benign binders for commercialized cathode materials has already become an imperative topic of practical significance, few studies have emphasized the importance of binders in recycling.<sup>23</sup> Binders play an important role in maintaining electrode integrity. Following this logic, they also contribute to the rapid separation of electroactive materials. Although there are concerns about the poor resistance of cathode materials in water and the relatively poor electrochemical stability under high pressure, it is feasible for water-based binders to be used with cathode materials (Fig. 4a). Guar gum, a natural non-ionic polysaccharide extracted from the seeds *Cyamopsis tetragonolobus*, has been successfully employed as a water-soluble binder for Li-rich cathode materials and has the function of reducing capacity degradation.<sup>64</sup> Li et al. demonstrated the feasibility of water-based manufacturing and recycling with the commercial water-based binder (CMC) and cathode materials (Fig. 4a).<sup>65-68</sup> Numerous efforts have been committed to developing suitable water-soluble binders for different cathode materials.<sup>69,70</sup> Still, the impacts of water-soluble binder in the performance of cathode and the feasibility of water-based manufacturing varied cathodes should be systematically studied. The development of water-soluble and the fluoride-free electrolyte is expected to overcome the use of toxic and expensive fluoride anions on the condition that the 1.23 V electrochemical stability window of water could be extended. Recent works on water-based electrolytes present opportunities regarding safe and recyclable LIBs.<sup>71</sup> A novel "water-in-ionomer" electrolyte based on lithium polyacrylic acid (LiPAA) and water developed by He et al.<sup>72</sup> opens a path for the development of non-toxic and more sustainable electrolytes with widened electrochemical stable stability window (Fig. 4b,d).

Simplifying battery configuration and components are theoretically favoured for convenient material separation. Considering the complexity in composition originated from the conventional prototype architecture, novel battery designs are expected to simplify the compositions and therefore, smoothing the material separation. Recently, a battery structure, which is the bipolar electrode structure (BE), can be adapted to assemble batteries using a unified current collector

for anode and cathode.<sup>73, 74</sup> Referring to the connection of multiple unit cells in series using external wiring, BE design not only eliminates the need for housing components and connecting essentials but also boosting electrochemical performances.<sup>74</sup> This simplification makes fewer components separated in battery recycling and cuts the cost in battery manufacturing since the elimination of expensive Cu foil provides the chance to achieve higher energy density. The effective potential window of Al will surge the researches in suitable electroactive materials (Fig. 4c). To avoid the side Li-Al alloying reaction with Li ( $\sim 0.3$  V vs. Li/Li<sup>+</sup>), the approach of using high voltage anode materials, such as Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> ( $\sim 1.5$  V vs. Li/Li<sup>+</sup>), presents a big opportunity in achieving bipolar LIBs with commercial electroactive materials.<sup>75</sup> Notwithstanding the excellent stability and safety of Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>, the significant voltage reduction of cells inevitably lost certain energy density. A trade-off between BE structure and high energy density can be achieved by exploring novel anode materials with discharge voltages of 0.5 to 1.0 V (vs. Li/Li<sup>+</sup>). The breakthroughs in n-type organic materials with lower redox potential hold promise for designing BE LIBs<sup>76</sup> and the reported materials such as 2,6-Naph(COOLi)<sub>2</sub> (discharge platform: 0.7 V vs. Li/Li<sup>+</sup>)<sup>77, 78</sup> and 4,4'-Bph(COOLi)<sub>2</sub> (discharge platform: 0.7 V vs. Li/Li<sup>+</sup>)<sup>79</sup> merit more attention. Beyond that, DRS-Li<sub>3+x</sub>V<sub>2</sub>O<sub>5</sub>, which delivers a reversible two Li<sup>+</sup> redistribution intercalation reaction at an average voltage of  $\sim 0.6$  V (vs. Li/Li<sup>+</sup>), represents a class of inorganic anode materials using Al foils.<sup>21</sup> This low-potential intercalation reaction mechanism sheds light on identifying other viable low-potential metal oxide anodes. Additionally, the investigations of inert substrates such as stainless steel mesh (SS) instead of Cu and Al foils have exemplified the feasibility of SS as a shared CC.<sup>80, 81</sup>

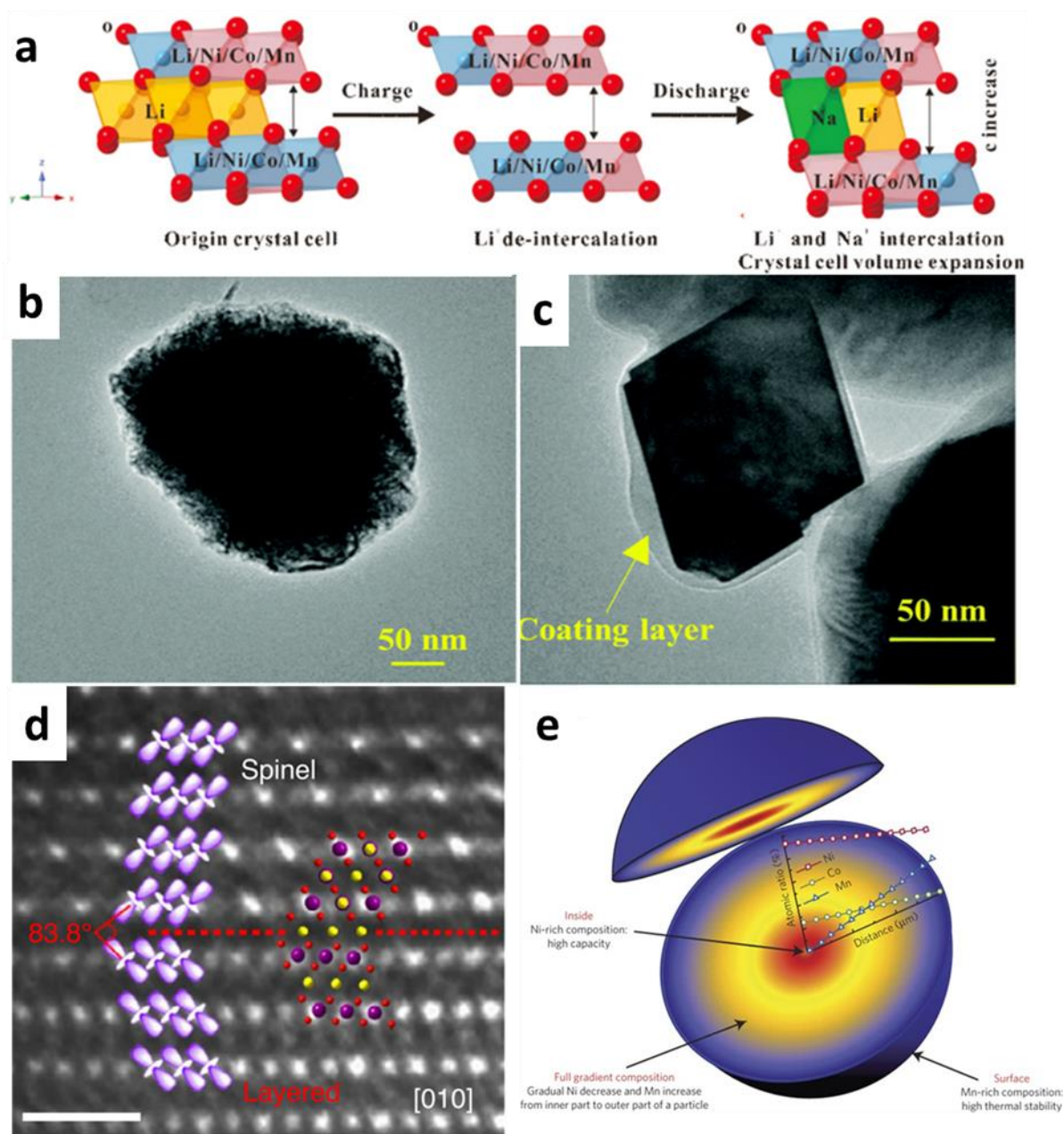
Furthermore, the sophisticated chemistry in today's LIBs inherently dilutes the recyclable contents within batteries, resulting in a reduction in the economic attractiveness of recycling. Design a battery with unified electroactive materials is highly promising in concentrating the cathode materials, simplifying separation, and therefore, increasing the economic viability. Notably, in light of the concept of the symmetric battery, batteries can be simplified by using electrode materials with two redox reactivity. Such an approach is expected to increase the ratio of electroactive materials, which will make recycling more economically worthwhile. Ideally, the two redox reactions happen in both low and high potential, which endows the materials to act as both anode and cathode. Such symmetric cell structure is readily transferred to bipolar cell configuration. Theoretically, the fundamental of designing these kinds of compounds is the synergistic effect of a transition metal element with low potential redox (Ti, V, Nb, and Mo) and other transition metal elements with high potential redox (Cr, Mn, Fe, Co, Ni, and Ru),<sup>82</sup> which is well exemplified by the works of other researchers.<sup>83-85</sup> Materials such as phosphate-based salt Li<sub>3</sub>V<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub><sup>82</sup> and Na<sub>3</sub>V<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub><sup>86</sup> take advantage of the redox reaction of V<sup>4+</sup>/V<sup>3+</sup> and V<sup>3+</sup>/V<sup>2+</sup> and hold promises in developing symmetric battery with cheap and abundant elements. The design of fully recyclable NIB with Na<sub>3</sub>V<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> as electroactive material and Al foil as CC is well demonstrated by Liu et al.<sup>87</sup> Based on the intrinsic property of Al foil, basic solution was used

to separate the electroactive materials and CC with a high solid-component recycling efficiency of ca. 99.7%. The design can be extended to other battery systems, which is promising to be practically transferred to the commercial scale.

On the account of the obvious advantages of aqueous polymeric binders regarding the limitations of PVDF, a foresight towards future EES recycling is to design a green and multifunctional binder for the emerging electrode materials with high energy density such as Si anode. A conductive self-healing polymeric binder consisting of a ureido-pyrimidinone (UPy) unit is capable of self-healing while ion-conducting

polyethylene glycol (PEG) was introduced onto the side chain of the poly(acrylic acid) (PAA) (Fig. 4e).<sup>88</sup> Such an approach offers a tested for exploring other sustainable binders. A further step towards simplified battery configuration is free of carbon black in the electrode, in which the feasibility has been demonstrated by the conductive composite binders such as PEDOT:PSS.<sup>89</sup>

Notably, polymeric electrode materials that show cathodic and anodic reactions with two redox peaks, hold promise in designing BE batteries with Li-based battery technology and beyond. More and more researches have proven the feasibility of constructing batteries with bipolar-type organic compounds,



**Fig. 5 Pre-design for sustainable battery materials.** (a) Schematic diagram of the ion-exchange of  $\text{Li}^+$  with  $\text{Na}^+$  provided by the CMC during cycling. Reproduced with permission from ref.96. Copyright 2016, Wiley-VCH. The TEM images of the electrode particles after 200 cycles using (b) PVDF and (c) CMC as the binder. Reproduced with permission from ref.97. Copyright 2016, Nature Publishing Group. (d) HAADF-STEM image at the spinel-layered interface for the SPL-LMO sample along the [010] zone axis. Scale bar, 1 nm. Reproduced with permission from ref.101. Copyright 2020, Nature Publishing Group. (e) Schematic diagram of the full concentration gradient lithium of manganese increasing accordingly. Reproduced with permission from ref.98. Copyright 2019, Nature Publishing Group.

such as polyparaphenylene (PPP;  $(C_6H_4)_n$ ),<sup>90</sup> poly(2,3-dithiino-1,4-benzoquinone) (PDB;  $(C_6O_2S_2)_n$ ),<sup>91</sup> 2,5-dihydroxyterephthalic acid ( $Na_4C_8H_2O_6$ ) with enolate and carboxylate groups.<sup>92</sup> For example, an asymmetric all-organic battery based on phenothiazine and naphthalene tetracarboxylic dianhydride units is designed, presenting two redox potentials at  $\sim 2.5V$  and  $3.7V$  (vs.  $Li/Li^+$ ) (Fig. 4f).<sup>93</sup> Integrating cathode-active moieties with anode-active moieties is another intelligent strategy to construct symmetric batteries. Such a strategy has been successfully applied to design 4,4'-(phenazine-5,10-diyl)dibenzoate (PZDB) containing cathode-active phenazine moiety covalently sandwiched between two conjugated anode-active benzoate moieties (Fig. 4g).<sup>94</sup> Moreover, it is claimed that this compound has excellent stability in common aprotic electrolytes and is promising for real-world use.

Embedding these chemical designs into future batteries will certainly smooth material separation, offering competitive efficiency and convenience over the complex shredding and material separation processes adopted in the current recycling system. Moreover, the purity of spent electroactive materials collected from such a process for regeneration will be ensured. Though there is still a long way to go before the realization of such designs for material separation, the strong driving force of global sustainable development will force the transformation of battery manufacturing.

#### Pre-design for sustainable electrode

The intrinsic sustainability of electrode materials is the prerequisite for EES recycling. A sustainable electrode that contributes to the referring scenario should merit the potential in cutting down the cost in recycling and manufacturing. Considering the high cost and supply risk of Co, it is expected to decouple the energy storage technology and the Co content.<sup>3</sup> <sup>95</sup> To this end, reducing the cobalt content in the electrode or replacing it with easily recyclable materials is a promising strategy.

Though Co-based batteries are considered a green energy technology, the supply risk of Co and its toxicity to the environment/humans diminishes its sustainability. Today's LIBs recycling can recover Co in various forms, which partially alleviated the demand for raw materials. However, the recycling efficiency and the purity of the products are the bottlenecks for a safe and long-term supply chain. Design of a cathode material with less Co content or even without Co would be an excellent contribution. Lithium-rich manganese-based materials (Li-rich Mn-based) and Ni-rich materials have a strong potential for commercial viability. Li-rich Mn-based and Ni-rich cathode materials could mitigate the supply risk of Co, while also increasing the sustainability of current LIBs technologies. More importantly, less Co content will significantly reduce the recycling costs. Reductive reagents are normally needed when recycling Co owing to the poor solubility of  $Co^{3+}$ . Comparatively, metal ions with lower valence, such as  $Ni^{2+}$ , have the intrinsic merit of better solubility, therefore avoiding the usage of costly reductive reagents.

Nevertheless, these materials suffer from the irreversible layered-to-spinel and cubic-to-tetragonal phase transition

associated with Jahn-Teller distorted  $Mn^{3+}$  ions and the dissolution of  $Mn^{2+}$  into the non-aqueous electrolyte, leading to a poor cycling performance with continuous voltage fading.<sup>96</sup> Utilizing a sustainable binder for the enhancement of cycling performance is a simple strategy. Commercial CMC-Na was utilized as a binder for Li-rich Mn-based materials and the proven ion-exchange of  $Na^+$  in CMC into the lattice of the cathode during the discharge process inhibits the phase transformation of layered structure into the spinel structure by suppressing the transition metal ion migration into the Li vacancies (Fig. 5a).<sup>96</sup> Guar gum has also been proposed as a binder for improved cycling performance, which attributes to the smooth and tightly coated layer containing both SEI and guar gum on the surface, leading to the stabilization of  $Ni^{2+}/Ni^{4+}$  during the charge/discharge process (Fig. 5b,c).<sup>97</sup> In addition to the implementation of a suitable binder, the design of the crystal structure<sup>98</sup> and nano-scaled interface modifications<sup>99</sup> were actively investigated to stabilize the crystal structure not only of Li-rich cathode materials but also Ni-rich materials. A structured Ni-rich oxide was developed with a linear increase concentration of Mn and a linear decrease concentration of Ni from the center towards the outer surface.<sup>100</sup> Such a design takes advantage of the high energy density of Ni and the high thermal and cycling stability of the Mn (Fig. 5e). A similar nano-design strategy, which is the Li-rich interior and a Li-poor surface was applied to ensure that Li-rich oxide cathode materials are immunized against oxygen release with a.<sup>98</sup>

Moreover, as a green cathode material for sustainable energy storage,  $LiMnO_2$  was actively investigated owing to its low cost, abundance, and environmental friendliness. Notwithstanding, the commercialization of  $LiMnO_2$  has been suppressed by the unstable structure associated with high-spin  $Mn^{3+}$ . To this end, the nano-scaled structural design holds promises in resolving these limitations. Zhu et al. proposed a hetero-structured layered and spinel domain with different orientations to stabilize the crystal structure of  $LiMnO_2$ .<sup>101</sup> The interfacial orbital ordering was observed by the HAADF-STEM taken at the spinel-layered interface along the [010] zone axis, which is originated from the almost perpendicular arrangement of the Mn  $d_{z^2}$  in these domains (shown in Fig. 5d). Such design helps suppress the cooperative Jahn-Teller distortion and Mn dissolution, thus accelerating the commercialization of these more sustainable energy materials.

Another sustainable material design that contributes to EES recycling is the implementation of polymeric electrode materials instead of inorganic compounds.<sup>11</sup> Though the improvement of organic electrode compounds is not as spectacular as inorganic materials in the past decades, a renaissance of organic electrodes is called upon since the intercalation compounds have reached their intrinsic limits.<sup>76</sup> The production of organic electrodes avoids the mining and high-temperature annealing process; hence a low  $CO_2$  footprint is expected. Coupled with its recyclability and low cost, organic electrodes hold promise to make rechargeable batteries and their recycling process more sustainable.<sup>11, 76</sup> More importantly, organic/polymeric compounds based on conversion reactions are also electroactive towards other ions. The solubilization of organic electrode materials in water and its insolubility in

organic solvents (such as ethanol), arguably, demonstrates a significant improvement in the environmental and economic sustainability of electrode material recycling processes. Compared to those processes currently used for inorganic electrode materials, which usually involves leaching with strong acid and solvent extraction using toxic organic solvents, water and ethanol can be used for recycling organic electrode materials such as Tetrakislawsonone<sup>102</sup> and dilithium benzenediacylate<sup>103</sup>. The potential of transferring this theory into practice is well demonstrated by other researchers.<sup>102-104</sup>

## Future perspectives

The exponential growth in consumption of LIBs to achieve green energy targets presents a new environmental challenge and accelerates natural resources depletion. Recycling spent LIBs has, therefore become a priority field of research since it can reduce the demand for mining new raw materials. Well-established recycling technologies including pyrometallurgy, hydrometallurgy, and their combinations are capable of recycling mixed spent batteries and recovering valuable metals, especially Co, Ni, and Mn in its various compounds such as Ni(OH)<sub>2</sub>. However, the simultaneous goal of high efficacy, high profit, and environmental sustainability has proven to be a challenging task for the current recycling sectors. Numerous efforts have been devoted to reducing the environmental impacts of metallurgical recycling technologies. The harmless treatment and recovery of toxic Li salt (such as LiPF<sub>6</sub>) and electrolyte (such as diethyl carbonate and dimethyl carbonate) have been achieved on a commercial scale. "Green solvents" such as deep eutectic solutions were developed to substitute conventional toxic solvents to extract critical metal elements. After optimization of the processing conditions and reagents used, the environmental sustainability of current recycling technologies seems to be improved. Unfortunately, considering their inherent energy intensity and chemical consumption, along with the more precise material extraction, separation, and purification processes, MBR technologies are not considered sustainable. Fortunately, as researchers and industries deepen their understanding of how to turn waste into resources, direct recycling was proposed and has proven to be a sustainable LIBs recycling technology.<sup>6</sup>

Direct recycling or direct regeneration is a non-destructive strategy to restore the electrochemical performance of cathode and anode, as well as recycling separator, electrolyte, and lithium salts, realizing fully recycling of spent LIBs. Direct recycling is often considered sustainable, but in reality, it is only as sustainable as its full processing line, including the pre-treatment process. If the pre-treatment process is highly labour-intensive and energy-intensive, the economic viability and sustainability of direct recycling diminish. This is the obstacle facing today's HADR, namely the high human intervention. Therefore, while there are many markets for recycling, it is believed that in the short term, current MBR technologies, including pyrometallurgy and hydrometallurgy, will continue to serve their respective roles in recycling market. That is because the number of recycling companies implementing the direct recycling strategy is negligible when

compared to the volume of literature published. This is most likely due to the major pitfalls of modern commercial Li-based batteries: sophisticated chemistry and poor traceability. The pre-treatment process for direct recycling is always understated, leading to a more costly recycling process on an industrial scale.

Obviously, resolving these two pitfalls is an essential requirement before realizing truly EES recycling of LIBs. Fortunately, revolutionary breakthroughs in intelligence technologies such as AI and ML, facilitate improved traceability and management of batteries. Furthermore, recent advanced materials science toward more sustainable rechargeable batteries also provides solutions that can be easily adapted into facilitating the sustainable recycling of LIBs and future batteries. Moreover, the chemical predesigns referring to simple and sustainable material separation and recycling proposed in this perspective are expected to convey the concept of embedding recycling into the materials selection and battery design towards closed loop and economically viable recycling. For the existing spent battery, we can still utilize the proposed "Reuse-Refurbish-Recover" EES recycling strategies, combining the state-of-the-art pre-treatment for the efficient separation of metal and non-metal components, and the direct regeneration of specific types of electroactive materials.

Beyond implementing predesign, some modifications, though still in their infancy, can be applied before retrieving damaged electroactive materials for the reduced need for disassembly. For example, at a macroscopic scale, dead batteries attributed to the drying of liquid electrolytes can be restored through adding a proper supplement of electrolytes without intricate disassembly. While from a nanoscale, 'inactive' Li species in the solid electrolyte interphase (SEI) might be rejuvenated and served as a supplementary Li source. Such a novel regeneration strategy has recently been proposed. Iodine species loaded by a biochar capsule host were used to release the Li<sup>+</sup> in Li<sub>2</sub>O-dominant SEI and thus rejuvenating the batteries back to acceptable electrochemical performances.<sup>105</sup> Similar approaches await exploration to reduce the workload of disassembly.

The superiority in GHG emission and economic offset from direct recycling in terms of LFP cathode given the more energetical efficiency in mining the iron precursor materials and the cost-effectiveness in manufacturing without premixing and drying steps to combine transition metals remain questionable.<sup>5</sup> However, infinite mining will not be the ultimate choice for environmental sustainability and the limited resources under the ground will someday become the bottleneck. Opportunities to increase the economic viability exist in the proposed intelligence-assisted "Reuse-Refurbish-Recover" recycling scenario. The recycling-friendly cell design and the artificial intelligence (AI)-assisted processes will help increase the cost-effectiveness in both recycling and manufacturing. For example, the waste olivine FePO<sub>4</sub> materials from selective recycling lithium from spent LiFePO<sub>4</sub> could be applied as a promising cathode material for NIBs.<sup>106</sup> Oxidative leaching reagents (such as Na<sub>2</sub>S<sub>2</sub>O<sub>8</sub>) can be used instead of strong acids to achieve the leaching of Li<sup>+</sup> while maintaining the

olivine structure. As a result, the spent batteries in converted into a resource, i.e., the "waste materials" within the spent batteries is transformed into another energy storage material.

Though more efforts need to be committed, the concept of intelligence-assisted predesigns in battery management systems, for material separation, simplifying the composition of batteries, and utilizing sustainable energy materials is promising to realize sustainable recycling of LIBs and beyond. More importantly, the intelligence-assisted predesign and "3R" concept could help the authorities set up and implement relevant regulations and even laws in the course of future battery design, thereby avoiding the widespread plastic catastrophe of the last century. LIBs product stewardship will help the government to establish regulations, standards, and incentives that embed innovative sustainable recycling principles into design and manufacturing. For example, the tax rebate could support recyclers to realize the recycling of low value spent electroactive materials, such as lithium iron phosphate. With such "carrots and sticks", the incorporation of intelligence-assisted predesign into battery management systems and battery chemistry will facilitate industrial sectors to implement the transitions towards a sustainable economy.

## Conflicts of interest

There are no conflicts to declare.

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## Notes and references

1. W. Marian, *PV Magazine*, 2020, **12**, <https://www.pv-magazine.com/2020/2008/2012/battery-manufacturing-capacity-set-to-increase-fourfold-to-2021-2023-twh-by-2030/>.
2. C. Vaalma, D. Buchholz, M. Weil and S. Passerini, *Nat. Rev. Mater.*, 2018, **3**, 18013.
3. *Nature Energy*, 2019, **4**, 253-253.
4. L. M. E. Cobalt, <https://www.lme.com/en-GB/Metals/Minor-metals/Cobalt#tabIndex=0>
5. R. E. Ciez and J. F. Whitacre, *Nature Sustainability*, 2019, **2**, 148-156.
6. P. Xu, Q. Dai, H. Gao, H. Liu, M. Zhang, M. Li, Y. Chen, K. An, Y. S. Meng and P. Liu, *Joule*, 2020.
7. D. H. S. Tan, A. Banerjee, Z. Chen and Y. S. Meng, *Nat Nanotechnol*, 2020, **15**, 170-180.
8. J. B. Dunn, L. Gaines, M. Barnes, J. L. Sullivan and M. Wang, *Material and energy flows in the materials production, assembly, and end-of-life stages of the automotive lithium-ion battery life cycle*, Argonne National Lab.(ANL), Argonne, IL (United States), 2014.
9. J. Xiao, J. Li and Z. Xu, *Environmental Science & Technology*, 2017, **51**, 11960-11966.
10. E. Fan, L. Li, Z. Wang, J. Lin, Y. Huang, Y. Yao, R. Chen and F. Wu, *Chemical Reviews*, 2020.
11. J. Piątek, S. Afyon, T. M. Budnyak, S. Budnyk, M. H. Sipponen and A. Slabon, *Advanced Energy Materials*, 2003456.
12. Y. Yang, E. G. Okonkwo, G. Huang, S. Xu, W. Sun and Y. He, *Energy Storage Materials*, 2021, **36**, 186-212.
13. E. Fan, L. Li, Z. Wang, J. Lin, Y. Huang, Y. Yao, R. Chen and F. Wu, *Chemical reviews*, 2020, **120**, 7020-7063.
14. G. Harper, R. Sommerville, E. Kendrick, L. Driscoll, P. Slater, R. Stolkin, A. Walton, P. Christensen, O. Heidrich, S. Lambert, A. Abbott, K. Ryder, L. Gaines and P. Anderson, *Nature*, 2019, **575**, 75-86.
15. T. Wang, H. Luo, Y. Bai, J. Li, I. Belharouak and S. Dai, *Advanced Energy Materials*, 2020, **10**, 2001204.
16. T. Wang, X. Yu, M. Fan, Q. Meng, Y. Xiao, Y.-X. Yin, H. Li and Y.-G. Guo, *Chemical Communications*, 2020, **56**, 245-248.
17. K. Liu, S. Yang, L. Luo, Q. Pan, P. Zhang, Y. Huang, F. Zheng, H. Wang and Q. Li, *Electrochimica Acta*, 2020, **356**.
18. H. D. Pham, M. Horn, J. F. S. Fernando, R. Patil, M. Phadatare, D. Golberg, H. Olin and D. P. Dubal, *Sustainable Materials and Technologies*, 2020, **26**.
19. Y. Shi, M. Zhang, Y. S. Meng and Z. Chen, *Advanced Energy Materials*, 2019, **9**, 1900454.
20. X. Li, J. Zhang, D. Song, J. Song and L. Zhang, *Journal of Power Sources*, 2017, **345**, 78-84.
21. H. Liu, Z. Zhu, Q. Yan, S. Yu, X. He, Y. Chen, R. Zhang, L. Ma, T. Liu and M. Li, *Nature*, 2020, **585**, 63-67.
22. E. Gies, *Nature*, 2015, **526**, S100.
23. D. L. Thompson, J. M. Hartley, S. M. Lambert, M. Shiref, G. D. Harper, E. Kendrick, P. Anderson, K. S. Ryder, L. Gaines and A. P. Abbott, *Green Chemistry*, 2020, **22**, 7585-7603.
24. W. Li, M. Rentemeister, J. Badedá, D. Jöst, D. Schulte and D. U. Sauer, *Journal of Energy Storage*, 2020, **30**, 101557.
25. Y. Zhang, Q. Tang, Y. Zhang, J. Wang, U. Stimming and A. A. Lee, *Nature Communications*, 2020, **11**, 1706.
26. A. Martín-Garín, J. A. Millán-García, A. Bãiri, J. Millán-Medel and J. M. Sala-Lizarraga, *Automation in Construction*, 2018, **87**, 201-214.
27. R. A. Stewart, K. Nguyen, C. Beal, H. Zhang, O. Sahin, E. Bertone, A. S. Vieira, A. Castelletti, A. Cominola, M. Giuliani, D. Giurco, M. Blumenstein, A. Turner, A. Liu, S. Kenway, D. A. Savić, C. Makropoulos and P. Kossieris, *Environmental Modelling & Software*, 2018, **105**, 94-117.
28. F. Gu, B. Ma, J. Guo, P. A. Summers and P. Hall, *Waste Management*, 2017, **68**, 434-448.
29. T. Kim, D. Makwana, A. Adhikaree, J. S. Vagdoda and Y. Lee, *Energies*, 2018, **11**, 125.
30. J. Huang, L. A. Blanquer, J. Bonafacino, E. R. Logan, D. A. Dalla Corte, C. Delacourt, B. M. Gallant, S. T. Boles, J. Dahn and H.-Y. Tam, *Nature Energy*, 2020, **5**, 674-683.
31. A. Condemi, F. Cucchiella and D. Schettini, *Applied Sciences*, 2019, **9**, 3422.
32. P. Nowakowski, *Journal of Cleaner Production*, 2018, **172**, 2695-2707.
33. Ö. Tozanlı, E. Kongar and S. M. Gupta, *Sustainability*, 2020, **12**, 5416.

34. S. S. Y. Ng, Y. Xing and K. L. Tsui, *Applied Energy*, 2014, **118**, 114-123.
35. D. Liu, J. Pang, J. Zhou, Y. Peng and M. Pecht, *Microelectronics Reliability*, 2013, **53**, 832-839.
36. P. Li, Z. Zhang, Q. Xiong, B. Ding, J. Hou, D. Luo, Y. Rong and S. Li, *Journal of Power Sources*, 2020, **459**, 228069.
37. L. Ren, L. Zhao, S. Hong, S. Zhao, H. Wang and L. Zhang, *IEEE Access*, 2018, **6**, 50587-50598.
38. Y. Fan, F. Xiao, C. Li, G. Yang and X. Tang, *Journal of Energy Storage*, 2020, **32**.
39. S. Shen, M. Sadoughi and C. Hu, 2019.
40. H. Tian, P. Qin, K. Li and Z. Zhao, *Journal of Cleaner Production*, 2020, **261**.
41. M.-F. Ng, J. Zhao, Q. Yan, G. J. Conduit and Z. W. Seh, *Nature Machine Intelligence*, 2020, **2**, 161-170.
42. Z. Deng, L. Yang, Y. Cai, H. Deng and L. Sun, *Energy*, 2016, **112**, 469-480.
43. Z. Zhou, Y. Cui, X. Kong, J. Li and Y. Zheng, *Journal of Energy Storage*, 2020, **32**.
44. X. Li, J. Jiang, D. Chen, Y. Zhang and C. Zhang, *Applied energy*, 2016, **177**, 537-543.
45. X. Bian, L. Liu and J. Yan, *Energy*, 2019, **177**, 57-65.
46. X. Chen, H. Lei, R. Xiong, W. Shen and R. Yang, *Applied Energy*, 2019, **255**, 113758.
47. X. Li, Z. Wang and J. Yan, *Journal of Power Sources*, 2019, **421**, 56-67.
48. F. Li and J. Xu, *Microelectronics Reliability*, 2015, **55**, 1035-1045.
49. T. Faika, T. Kim, J. Ochoa, M. Khan, S. Park and C. S. Leung, 2019.
50. T. Jin, Z. Sun, L. Li, Q. Zhang, M. Zhu, Z. Zhang, G. Yuan, T. Chen, Y. Tian and X. Hou, *Nature communications*, 2020, **11**, 1-12.
51. L. Song, K. Zhang, T. Liang, X. Han and Y. Zhang, *Journal of Energy Storage*, 2020, **32**, 101836.
52. A. Ran, Z. Zhou, S. Chen, P. Nie, K. Qian, Z. Li, B. Li, H. Sun, F. Kang, X. Zhang and G. Wei, *Advanced Theory and Simulations*, 2020, **3**, 2000109.
53. A. Y. Alqahtani, S. M. Gupta and K. Nakashima, *International Journal of Production Economics*, 2019, **208**, 483-499.
54. D. L. Thompson, J. M. Hartley, S. M. Lambert, M. Shiref, G. D. J. Harper, E. Kendrick, P. Anderson, K. S. Ryder, L. Gaines and A. P. Abbott, *Green Chemistry*, 2020, **22**, 7585-7603.
55. K. Wegener, W. H. Chen, F. Dietrich, K. Dröder and S. Kara, *Procedia CIRP*, 2015, **29**, 716-721.
56. X. V. Wang and L. Wang, *International Journal of Production Research*, 2019, **57**, 3892-3902.
57. M. Arbabzadeh, G. M. Lewis and G. A. Keoleian, *Journal of Energy Storage*, 2019, **24**, 100779.
58. H. Abuzied, H. Senbel, M. Awad and A. Abbas, *Engineering Science and Technology, an International Journal*, 2020, **23**, 618-624.
59. A. Lendlein and O. E. Gould, *Nature Reviews Materials*, 2019, **4**, 116-133.
60. F. Zhang, Z. Zhang, Y. Liu and J. Leng, *Fibers and Polymers*, 2014, **15**, 534-539.
61. K. Yamagishi, Y. Ogawa, D. Ando, Y. Sutou and J. Koike, *Scripta Materialia*, 2019, **168**, 114-118.
62. Y. Ogawa, D. Ando, Y. Sutou and J. Koike, *Science*, 2016, **353**, 368-370.
63. Y. Liu, H. Du, L. Liu and J. Leng, *Smart Materials and Structures*, 2014, **23**, 023001.
64. Z.-W. Yin, T. Zhang, S.-J. Zhang, Y.-P. Deng, X.-X. Peng, J.-Q. Wang, J.-T. Li, L. Huang, H. Zheng and S.-G. Sun, *Electrochimica Acta*, 2020, **351**, 136401.
65. J. Li, Y. Lu, T. Yang, D. Ge, D. L. Wood III and Z. Li, *iScience*, 2020, 101081.
66. A. Guerfi, M. Kaneko, M. Petitclerc, M. Mori and K. Zaghib, *Journal of Power Sources*, 2007, **163**, 1047-1052.
67. J.-T. Li, Z.-Y. Wu, Y.-Q. Lu, Y. Zhou, Q.-S. Huang, L. Huang and S.-G. Sun, *Advanced Energy Materials*, 2017, **7**, 1701185.
68. M. Sun, H. Zhong, S. Jiao, H. Shao and L. Zhang, *Electrochimica acta*, 2014, **127**, 239-244.
69. P. Jakóbczyk, M. Bartmański and E. Rudnicka, *Journal of Applied Electrochemistry*, 2021, **51**, 359-371.
70. H. Isozumi, T. Horiba, K. Kubota, K. Hida, T. Matsuyama, S. Yasuno and S. Komaba, *Journal of Power Sources*, 2020, **468**, 228332.
71. D. Kundu, B. D. Adams, V. Duffort, S. H. Vajargah and L. F. Nazar, *Nat. Energy*, 2016, **1**, 16119-16126.
72. X. He, B. Yan, X. Zhang, Z. Liu, D. Bresser, J. Wang, R. Wang, X. Cao, Y. Su, H. Jia, C. P. Grey, H. Frielinghaus, D. G. Truhlar, M. Winter, J. Li and E. Paillard, *Nature Communications*, 2018, **9**, 5320.
73. K.-N. Jung, H.-S. Shin, M.-S. Park and J.-W. Lee, *ChemElectroChem*, 2019, **6**, 3842-3859.
74. T. Liu, Y. Yuan, X. Tao, Z. Lin and J. Lu, *Advanced Science*, 2020, **7**, 2001207.
75. S.-H. Kim, K.-H. Choi, S.-J. Cho, J. Yoo, S.-S. Lee and S.-Y. Lee, *Energy Environ. Sci.*, 2018, **11**, 321-330.
76. Y. Lu and J. Chen, *Nature Reviews Chemistry*, 2020, **4**, 127-142.
77. T. Yasuda and N. Ogihara, *Chemical Communications*, 2014, **50**, 11565-11567.
78. N. Ogihara, N. Ohba and Y. Kishida, *Science advances*, 2017, **3**, e1603103.
79. Y. Ozawa, N. Ogihara, M. Hasegawa, O. Hiruta, N. Ohba and Y. Kishida, *Communications Chemistry*, 2018, **1**, 1-11.
80. V. Livshits, A. Blum, E. Strauss, G. Ardel, D. Golodnitsky and E. Peled, *Journal of power sources*, 2001, **97**, 782-785.
81. Z. Wei, S. Chen, J. Wang, Z. Wang, Z. Zhang, X. Yao, Y. Deng and X. Xu, *J. Mater. Chem. A*, 2018, **6**, 13438-13447.
82. S. Guo, J. Yi, Y. Sun and H. Zhou, *Energy & Environmental Science*, 2016, **9**, 2978-3006.
83. H. Wang, T. Zhang, C. Chen, M. Ling, Z. Lin, S. Zhang, F. Pan and C. Liang, *Nano Research*, 2018, **11**, 490-498.
84. H. Gao and J. B. Goodenough, *Angewandte Chemie*, 2016, **128**, 12960-12964.
85. C. Wang, L. Zhang, M. Al - Mamun, Y. Dou, P. Liu, D. Su, G. Wang, S. Zhang, D. Wang and H. Zhao, *Advanced Energy Materials*, 2019, **9**, 1900909.
86. B. L. Ellis, T. N. Ramesh, L. J. M. Davis, G. R. Goward and L. F. Nazar, *Chemistry of Materials*, 2011, **23**, 5138-5148.
87. T. Liu, Y. Zhang, C. Chen, Z. Lin, S. Zhang and J. Lu, *Nature Communications*, 2019, **10**, 1965.
88. J. Nam, E. Kim, K. R. K. Y. Kim and T. H. Kim, *Sci Rep*, 2020, **10**, 14966.
89. H. Zhong, A. He, J. Lu, M. Sun, J. He and L. Zhang, *Journal of power sources*, 2016, **336**, 107-114.
90. L. Zhu, A. Lei, Y. Cao, X. Ai and H. Yang, *Chemical Communications*, 2013, **49**, 567-569.
91. J. Xie, Z. Wang, Z. J. Xu and Q. Zhang, *Advanced Energy Materials*, 2018, **8**, 1703509.

92. S. Wang, L. Wang, Z. Zhu, Z. Hu, Q. Zhao and J. Chen, *Angewandte Chemie*, 2014, **126**, 6002-6006.
93. N. Casado, D. Mantione, D. Shanmukaraj and D. Mecerreyes, *ChemSusChem*, 2020, **13**, 2464-2470.
94. G. Dai, Y. He, Z. Niu, P. He, C. Zhang, Y. Zhao, X. Zhang and H. Zhou, *Angew Chem Int Ed Engl*, 2019, **58**, 9902-9906.
95. *Nature Energy*, 2020, **5**, 825-825.
96. S. Zhang, H. Gu, H. Pan, S. Yang, W. Du, X. Li, M. Gao, Y. Liu, M. Zhu, L. Ouyang, D. Jian and F. Pan, *Advanced Energy Materials*, 2016, **7**.
97. T. Zhang, J. T. Li, J. Liu, Y. P. Deng, Z. G. Wu, Z. W. Yin, D. Guo, L. Huang and S. G. Sun, *Chem Commun (Camb)*, 2016, **52**, 4683-4686.
98. Z. Zhu, D. Yu, Y. Yang, C. Su, Y. Huang, Y. Dong, I. Waluyo, B. Wang, A. Hunt and X. Yao, *Nature Energy*, 2019, **4**, 1049-1058.
99. B. Qiu, M. Zhang, L. Wu, J. Wang, Y. Xia, D. Qian, H. Liu, S. Hy, Y. Chen and K. An, *Nature communications*, 2016, **7**, 1-10.
100. Y. K. Sun, Z. Chen, H. J. Noh, D. J. Lee, H. G. Jung, Y. Ren, S. Wang, C. S. Yoon, S. T. Myung and K. Amine, *Nat Mater*, 2012, **11**, 942-947.
101. X. Zhu, F. Meng, Q. Zhang, L. Xue, H. Zhu, S. Lan, Q. Liu, J. Zhao, Y. Zhuang, Q. Guo, B. Liu, L. Gu, X. Lu, Y. Ren and H. Xia, *Nature Sustainability*, 2020, DOI: 10.1038/s41893-020-00660-9.
102. M. Miroshnikov, K. Kato, G. Babu, N. K. Thangavel, K. Mahankali, E. Hohenstein, H. Wang, S. Satapathy, K. P. Divya and H. Asare, *ACS Sustainable Chemistry & Engineering*, 2019, **7**, 13836-13844.
103. S. Renault, A. Oltean and D. Brandell, 2015.
104. S. Renault, D. Brandell and K. Edström, *ChemSusChem*, 2014, **7**, 2859-2867.
105. C. Jin, T. Liu, O. Sheng, M. Li, T. Liu, Y. Yuan, J. Nai, Z. Ju, W. Zhang and Y. Liu, *Nature Energy*, 2021, 1-10.
106. W. Tang, X. Song, Y. Du, C. Peng, M. Lin, S. Xi, B. Tian, J. Zheng, Y. Wu, F. Pan and K. P. Loh, *Journal of Materials Chemistry A*, 2016, **4**, 4882-4892.