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Rechargeable Batteries of the Future—The State of the Art from a BATTERY 2030+ Perspective

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The development of new batteries has historically been achieved through discovery and development cycles based on the intuition of the researcher, followed by experimental trial and error—often helped along by serendipitous breakthroughs. Meanwhile, it is evident that new strategies are needed to master the ever-growing complexity in the development of battery systems, and to fast-track the transfer of findings from the laboratory into commercially viable products. This review gives an overview over the future needs and the current state-of-the art of five research pillars of the European Large-Scale Research Initiative BATTERY 2030+, namely 1) Battery Interface Genome in combination with a Materials Acceleration Platform (BIG-MAP), progress toward the development of 2) self-healing battery materials, and methods for operando, 3) sensing to monitor battery health. These subjects are complemented by an overview over current and up-coming strategies to optimize 4) manufacturability of batteries and efforts toward development of a circular battery economy through implementation of 5) recyclability aspects in the design of the battery.

1. State of the Art: Introduction

1.1. Introduction

The battery research field is vast and flourishing, with an increasing number of scientific studies being published year after year, and this is paired with more and more different applications relying on batteries coming onto the market (electric vehicles, drones, medical implants, etc.). The large interest arises from the fact that quality and performance of rechargeable batteries (and primarily lithium ion batteries) have increased in the latest years while at the same time the prize has decreased dramatically.[1] "Batteries are a key enabler for European competitiveness and decarbonization" as stated in the strategic agenda of the European Battery Partnership[2] and will be one necessary tool to make Europe "fit for 55 within 2030."[3]

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The questions that need to be answered to allow the expectations on the batteries of the future to become reality are many. What battery chemistries are we to expect in the future? Will enough raw materials be available to realize a battery-based true decarbonization of our society? Will recycling be sufficiently efficient to recover most of the components of a battery cell? Will battery manufacturing processes be sustainable enough not to counteract the introduction of more batteries?

The purpose of this paper is to define the state of the art of necessary future battery research fields which can, at least partly, support the answers to these questions. These fields are selected to directly match and form the platform for the vision described in the long-term research-oriented Battery 2030+ roadmap.[4] This roadmap has a "chemistry neutral" stance which is employed for the design of long-term ideas well beyond 2030, with clear expectations on what to achieve and what actions and measures are needed to reinvent the way we invent the batteries of the future (a scientifically deeper roadmap by Kristina Edström et al. is published separately in this special issue). Battery 2030+ is the "European large-scale research initiative for future battery technologies"[4] with an approach focusing on the most critical steps that can enable the acceleration of the findings of new materials and battery concepts, the introduction of smart functionalities directly into battery cells and all different parts always including ideas for stimulating long-term research on sustainable manufacturing and recycling. This will be reflected in the different chapters within the paper.

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1.2. History

Electrochemical energy storage has become an increasingly important and growing topic which started already in the 18th century, when Alessandro Volta built his "pile" consisting of alternating cathode and anode layers, separated by a tissue and connected by an electrolyte. While the original aim of Volta was to perform biological experiments rather than energy storage, the basic setup of the pile is still the template for any modern battery.

Driven by the technical progress and the development of electrical applications in the 19th and 20th century, electrical power sources moved more and more into the focus of research and a series of rechargeable (i.e., "secondary") and non-rechargeable (i.e., "primary") batteries was developed, see **Figure 1**. Among these, the lead-acid battery was a major and successful breakthrough. Still today, the Pb-acid battery holds a major share on the battery market. Already 150 years ago, it enabled the first electric vehicles which dominated the market long before the combustion engines led to a technology change after having sufficiently improved in the early 20th century.

With the technological progress and the diversification of electronic and mechanical applications in the second half of the 20th century the demand rose for batteries in consumer applications with longer operation times, smaller size, lighter weight, rechargeability, high safety and low cost. In turn, this has inspired chemists, physicists, materials scientists and engineers to develop new designs and new concepts of batteries which can satisfy these demands and offer viable solutions.

Since the 1960s, the so far most successful type of batteries is under development: rechargeable batteries which are based on lithium ions as internal charge carriers.[6,7] The first Li-batteries used metallic lithium in the anode, together with a liquid electrolyte—a concept which has later been dropped for safety reasons.[6,7] Therefore, since the late 1980s, the Li metal has been replaced by graphite, which can safely intercalate and de-intercalate Li ions (i.e., the "Li-ion battery"), see also **Figure 2**. However, the Li is diluted by a factor of 10 by this means, which leads to a considerable lowering of the energy density of the battery. Considerable improvements were made on the cathode side. First electrodes were based on layered compounds such as TiS_2,[8] which intercalate Li ions at comparably low potential (≈2 V vs Li/Li^+) and offered only moderate storage capacities. A major advance was realized by switching to layered transition metal oxides such as CoO_2[9,10] which, due to the stronger interaction of the oxidic groups to lithium and the higher packing density enabled both higher potential and higher storage capacity of the cathode. Thus, the first commercial Li ion battery was built and sold by SONY in the early 1990s. Since its market introduction, the Li-ion battery has increased its energy density by a factor of three to four while the prize has dropped by a factor of 18, including a 90% drop in the last 10 years.[1] A series of efforts is underway worldwide to further increase the performance by developing better storage materials, electrolytes, and cell concepts.

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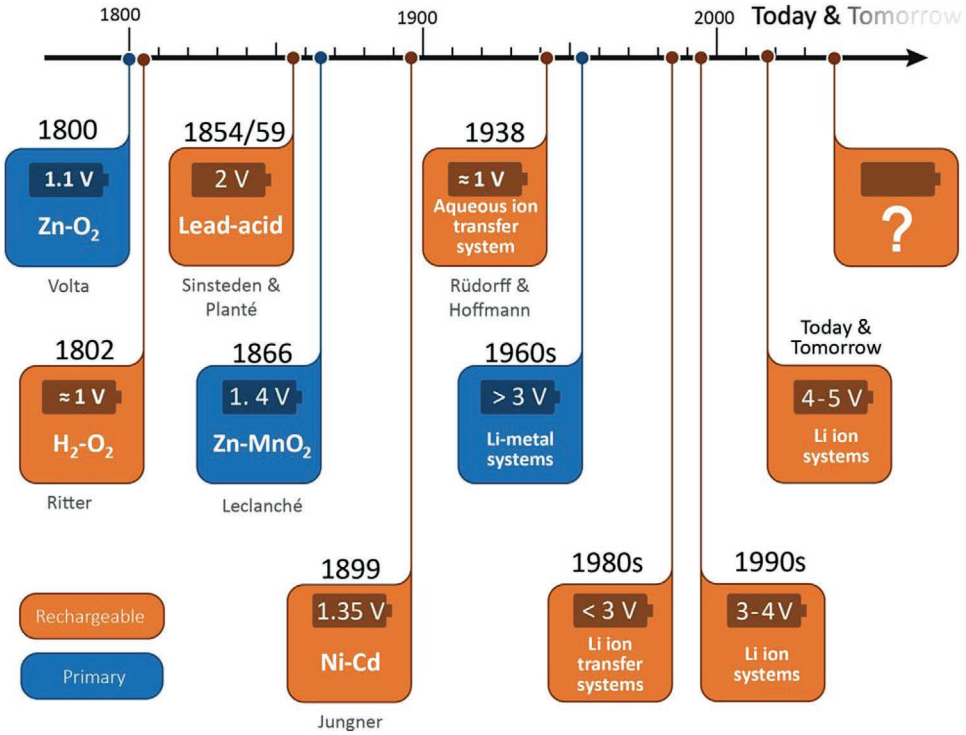

Figure 1. Historical overview of the development of battery types by Placke et al. Reproduced with permission.[5] Copyright 2021, Springer Nature.

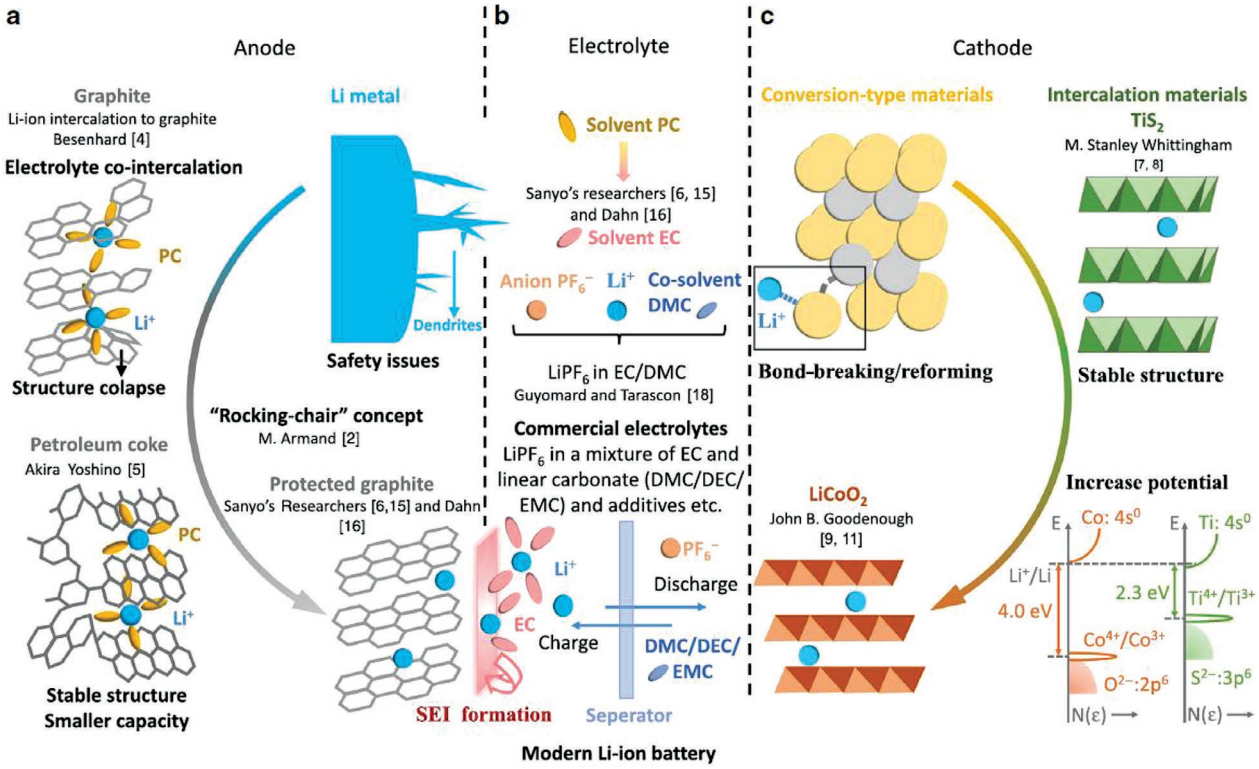

Figure 2. Milestone discoveries that shaped the modern lithium-ion batteries. The development of a) anode materials including lithium metal, petroleum coke and graphite, b) electrolytes with the solvent propylene carbonate (PC), a mixture of ethylene carbonate (EC) and at least one linear carbonate selected from dimethyl carbonate (DMC), diethyl carbonate (DEC), ethyl methyl carbonate (EMC) and many additives, c) cathode materials including conversion-type materials, intercalation materials titanium disulfide (TiS_2) and lithium cobalt oxide ($LiCoO_2$). Reproduced under the terms of the CC-BY open access license.[14] Copyright 2020, The Authors.

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1.3. Situation

Meanwhile, electrochemical energy storage in batteries is regarded as a critical component in the future energy economy, in the automotive- and in the electronic industry. While the demands in these sectors have already been challenging so far, the increasingly urgent need to replace fossil energy by energy from renewable resources in both the stationary and the mobile sector adds further challenges. For 2030, a globally installed storage capacity of more than 1 TWh in batteries is foreseen.[11,12] This massive expansion of storage capacity generates extra challenges not only with respect to energy density and fast charging. Rather, the mass application of batteries requires additional focus on aspects such as the sustainability of resources, the recyclability of components, cycle- and calendar lifetime of the system, safety and costs. In fact, the need for viable solutions is accelerating rather than slowing down at the moment. The reasons for this are on the one hand the need to reduce the CO_2 emissions in order to slow down global warming, and the necessity to supersede our reliance on fossil fuels, the resources of which are expected to dwindle from the mid-2020s onward, according to the International Energy Agency.[13]

The recent developments and the technological status in the field are summarized in Figure 2 wherein the development steps are indicated for the cathode, the anode, and the electrolyte of a Li ion battery. From the current state of knowledge, it will be difficult or even impossible to satisfy the future requirements with solutions that are based on this actual technological status. Rather, new approaches will be necessary to accelerate research and find better materials, build more reliable and self-controlling battery systems and integrate later demands from recycling already at the beginning of the R&D process and in manufacturing.

1.4. Current Strategy

In order to tackle these challenges, the battery community is currently focusing on several major pathways of research which shall put forth systems that can fulfil either all or at least several of the demands listed above. A major trend is to replace critical elements in the battery by more sustainable solutions, while still improving the properties of the battery. In general, the following development trends can be noticed:

- Replacement of critical elements in the cathode by more sustainable elements with a higher natural abundancy. This is done either by development of low Co-containing NMC (nickel-manganese-cobalt oxide) materials[15,16] and/or by developing new Co-free cathode materials with high capacities such as Li–Ni–Mn–oxide systems or battery cathodes based on Li-iron phosphate (LFP). Nickel- and manganese-rich cathode materials have already reached the commercial stage.[15,16] Improvement of rate performance and long-term stability through functional coatings is still very actively pursued, however.[17] The likewise commercialized LFP, on the other hand, offers high safety and long cycle life, albeit at lower specific capacity.[18,19]
- On the anode side, solutions are being developed for so-called post-Li systems which do not rely on Li as charge carrier any more. Such batteries are based on Na, Mg, Al, Zn, Ca, or Cl, use globally abundant and recyclable materials and can provide batteries with a more sustainable perspective. The sodium ion battery is first of these new "beyond" technologies to reach commercially viability, even though mainly in the area of stationary energy storage systems energy where energy density and charging rate impose less stringent limitations.[20–22]
- Conversion-based systems based on Li-chalcogenide combination (Li–sulfur,[23–25] Li–O_2[26,27]) are being developed. These technologies still face challenges, but also promise to deliver very high capacities while necessitating only unproblematic resources.
- The development of all-solid-state batteries (ASSB) shall enable higher storage capacities and higher safety by replacing the so far liquid electrolyte in batteries by a solid ion conductor. This shall allow the use of metallic lithium in the anode which would considerably enhance the storage capacity of the battery. The realization of lithium-metal batteries is making progress, but the challenges are enormous. This has recently prompted the development of resource-saving lithium metal-free anodes wherein Li is plating on the current collector itself.[28–30]

These approaches are also reflected in the general research strategies in the automotive sector of major players, which is depicted in **Figure 3**.

1.5. The Challenge

The research in these fields is currently done by a classical Edisonian sequential battery discovery and development cycle which is essentially based on the intuition of the researcher, followed by trial and error in the experiment. This approach, together with fortune in some cases has enabled virtually all successful developments so far. However, from what has been stated above, the crucial need for much more powerful and sustainable solutions has become so big, that it is questionable whether the Edisonian approach can solve the problem. In particular, the rising complexity, from the various battery materials to the entire battery system makes it difficult to transfer findings from the laboratory scale into commercially viable products. The following chapters will address these needs and present a unique collection of the specific state-of-the art in the five research pillars of the European Large-Scale Research Initiative BATTERY 2030+:

1) Battery Interface Genome in combination with a Materials Acceleration Platform (BIG-MAP)
2) Self-healing
3) Sensing
4) Manufacturability
5) Recyclability

2. State of the Art: (BIG-) MAP

2.1. Autonomous Synthesis and Experimentation

A core element in the development of Materials Acceleration Platforms (MAPs)[32] to accelerate the advancement of new and

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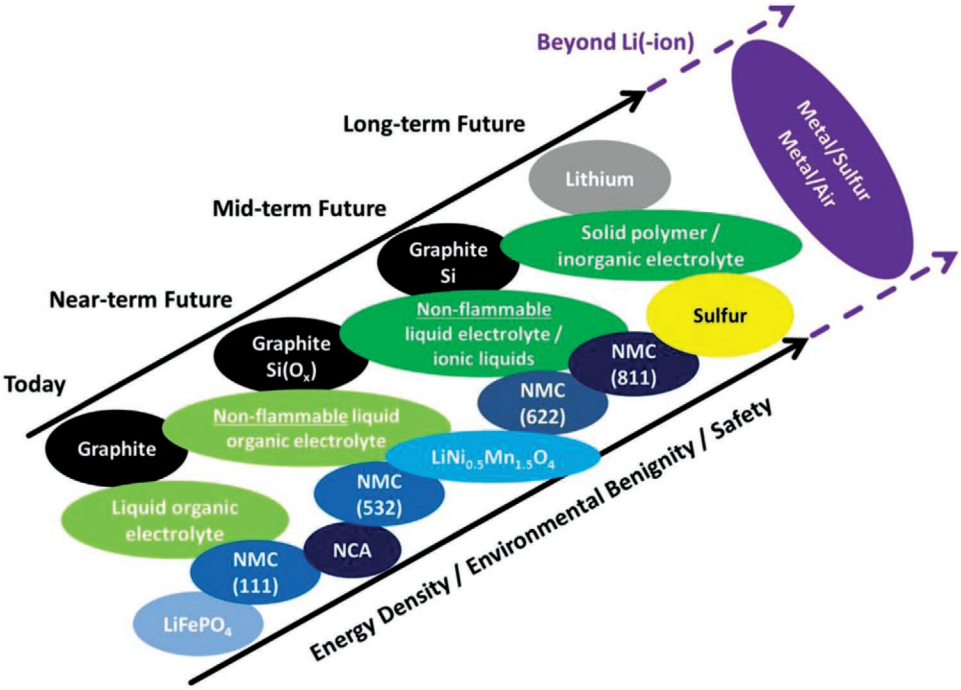

Figure 3. Joint perspectives of automotive battery R&D in China, Europe, Germany, Japan, and the USA. Reproduced with permission.[31] Copyright 2018, Elsevier.

improved battery chemistries and cells is the efficient exploration of electrode materials,[33–36] electrolyte formulations,[33] and processes.[37] A more efficient conduction of experiments hinges on the availability of automatization[38,39] of characterization tools and AI-enhanced analysis tools (AI: artificial intelligence, see Bhowmik et al. in this issue, AI). These research steps need to be accelerated individually, but also integrated to conduct combinatorial synthesis[40,41] and characterization experiments in an, at least partially, autonomous fashion. Building upon the advancements in the combinatorial materials science community,[38,42] different research groups have endeavored to transition from high-throughput characterization and combinatorial synthesis of bespoke (mostly) organic and thin-film materials toward autonomous synthesis[43,44] (see also references therein). Generally speaking, there is an established toolbox of combinatorial synthesis of thin-film materials, a few studies on combinatorial formulation of electrolytes,[33,45] and few studies on the combinatorial synthesis of powders,[46] but still none for a complete autonomous workflow from discovery toward devices. Methods for combinatorial synthesis (i.e. high-throughput screening, HTS) of battery related materials like anodes, cathodes, and electrolytes (beyond those cited above) can be found in this special issue's dedicated paper (Laskovic et al. in this issue, HTS). High-throughput battery assembly has largely been done on batteries sharing a common electrode, i.e., multiple cathode materials are tested against the same anode.[34–36,47] Applications of high-throughput electrochemistry and related electrochemical microscopy methods were reviewed in a recently by Daboss et al.,[48] highlighting especially the potential of scanning electrochemical microscopy (SECM) and scanning droplet cells (SDC) for battery research.

Acceleration by means of AI-enhanced methods are discussed in the dedicated paper in this issue (Bhowmik et al., AI). Recent advances in automated analysis and translation of results across instruments specifically designed for battery related applications play an import role, for example, translation of battery cycling data as published by Herring et al.[49] One of the largest bottlenecks in transitioning from conventional research methods toward accelerated approaches is the automation of workflows[38,43,44,50] and the automated data analysis. Seemingly simple problems like background subtraction[50] or spectral deconvolution[51] under physical constraints do, however, constitute complex mathematical problems.[52] This includes recent advancements in automated X-ray diffraction data analysis.[52–54] All of the above mentioned steps along the research value chain that hinge on interoperable data management and data management plans.[55] When data analysis and data management are implemented in a frictionless workflow, interesting non-convex optimization problems like the lifetime extension for fast-charging protocols become possible.[56,57]

2.2. Understanding the SEI from a Multi-Scale and Multi-Domain Approach

Conversion between electric and chemical energy inside batteries takes place at the interfaces between electrodes and electrolytes. Structures and processes at these interfaces determine their performance and degradation. However, one cannot simply identify a single relevant process, since one observes several interplaying phenomena, for example, solid–electrolyte interphase (SEI) dynamics and lithium plating. While the fundamental understanding of the SEI has evolved over the last five decades (**Figure 4**), recent developments in experimental operando techniques (see paper by Lyonnard et al. in this issue, LSF),

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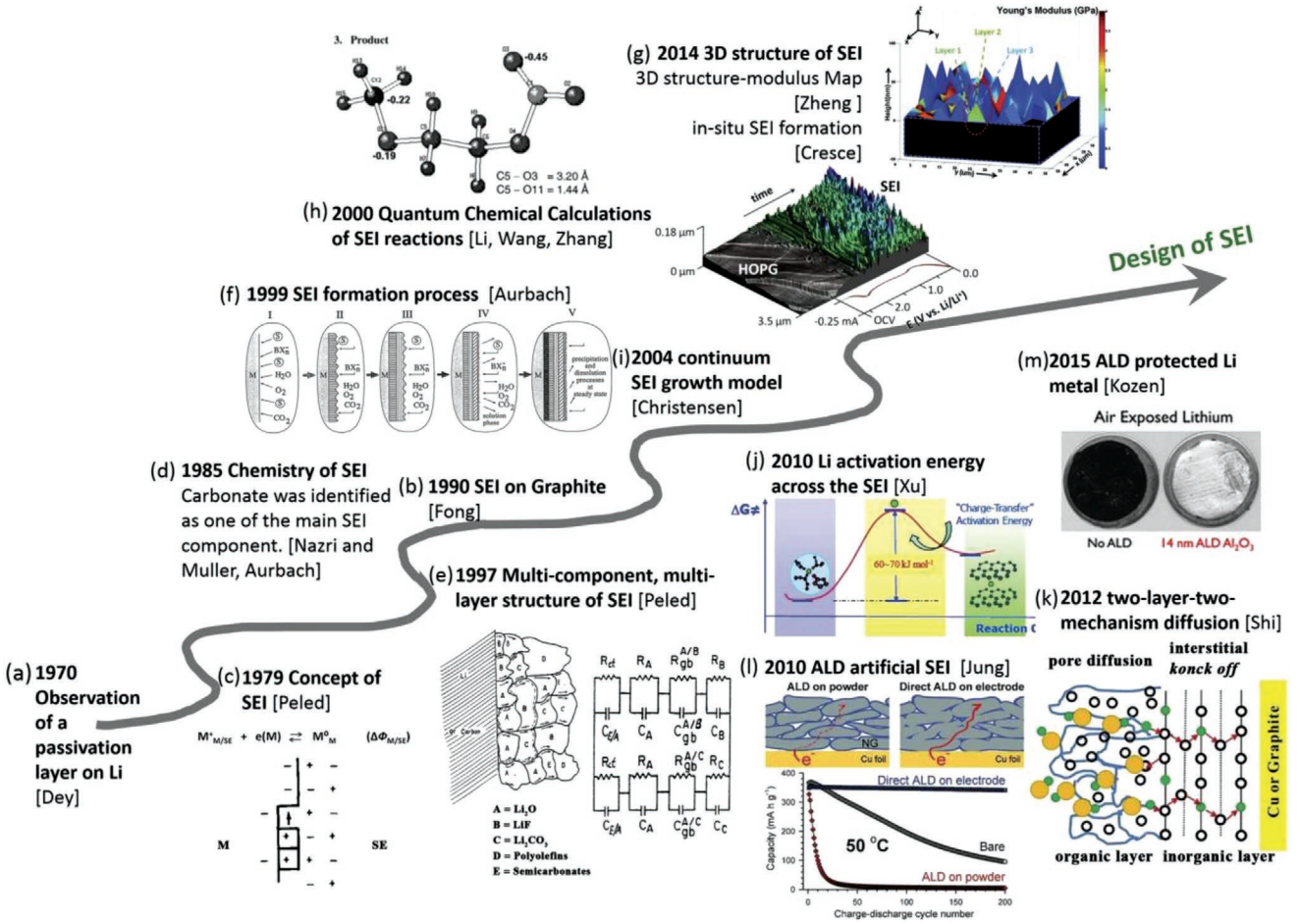

Figure 4. Four decades of experimental research and modelling of the SEI on negative electrodes: From discovery to understanding and artificial design. Reproduced under the terms of the CC-BY license. Copyright 2018, The Authors.

quantum calculations and machine learning (ML) have accelerated this understanding.

While the interfacial properties depend critically on the specific battery materials and interfaces, transfer and active learning from different battery materials and designs can pave the way for inverse design[59] (also see Bhowmik et al. in this issue, AI). The Battery Interface Genome (BIG) has taken up this challenge and aims at deciphering the complexity of interfacial phenomena and processes for relevant battery chemistries[60] (also see roadmap paper by Edström et al. in this issue).

In this section, we discuss the state of the art in describing the physico-chemical phenomena at the electrode-electrolyte interface, where the electrochemical energy conversion takes place (see also INTER paper by Grimaud et al. in this issue). Besides the wanted half-cell reactions, degradation reactions also proceed at the interface. Most importantly for lithium-ion batteries, the electrolyte is unstable at the typical working potentials of the negative electrodes. Luckily, the decomposition products of the electrolyte form a solid, passivating SEI layer on the electrode surface limiting further degradation. We discuss continued SEI formation as important degradation mechanism in lithium-ion batteries.

Processes at the battery interface are determined by atomistic reaction processes on the one hand and the emergent structures up to the micron scale on the other hand. Thus, BIG requires a multi-scale and multi-sourced approach for theory[58] and experiments. Electron configurations are typically calculated by density functional theory (DFT),[61] the atomistic motion is simulated with ab-initio and classical molecular dynamics simulations (AIMD/MD),[62] amorphous structures are sampled with Monte Carlo simulations (MC),[62] structures, transport, and mechanics are captured in continuum simulations (CS).[63] Similarly, a plethora of methods is used for measuring interfacial properties on the different length scales, for example, X-ray photoelectron spectroscopy (XPS) determines chemical compositions,[64–66] time-of-flight secondary mass spectroscopy (TOF-SIMS) captures the inhomogeneity of chemical SEI composition,[65] electron energy-loss spectroscopy (EELS) show the heterogeneity of the SEI throughout the electrode,[67] in situ X-ray diffraction (XRD) can reveal structures of crystalline phases,[68] scanning electron microscopy (SEM) can provide nanoscale resolution under vacuum conditions,[69,70] Raman Spectroscopy contains information about the chemical composition of the probe,[71] and electroanalytical methods determine SEI thickness.[72,73] Cutting-edge methods include

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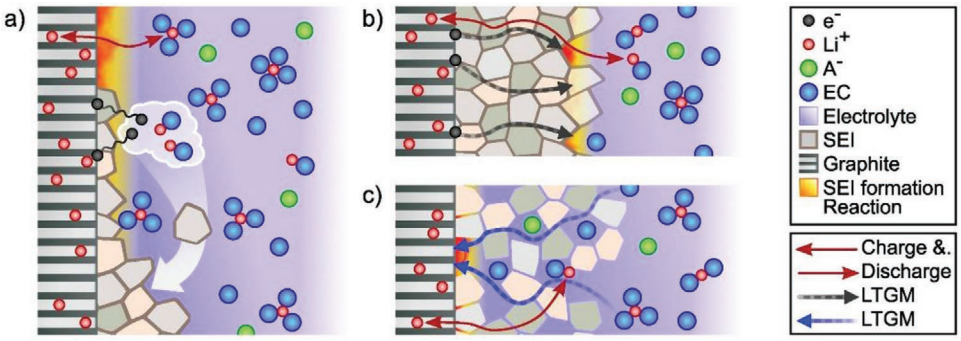

Figure 5. Cross-section through the negative electrode, the SEI, and the electrolyte. Solvent, Li ions and electrons are mobile species and move as indicated by the corresponding arrows. a) Initial SEI formation: Electrons tunnel, electrolyte is reduced and reduction products precipitate as solid film. b) Long-term SEI growth proceeds via a mechanism that transports negative charge to the SEI/electrolyte interface. c) Alternatively, long-term SEI growth is caused by electrolyte diffusing toward the electrolyte/SEI interface. Reproduced with permission.[63] Copyright 2018, Elsevier.

nuclear magnetic resonance (NMR) spectroscopy which observes chemical bonds[74] and atomic-resolution imaging like cryo-transmission electron microscopy (TEM) that visualizes the crystalline structure of the SEI.[75,76]

The SEI contains several chemical compounds, which Peled et al. originally depicted as a mosaic SEI structure.[77] Further analysis revealed the regularity in the SEI structure. The classification into an inner inorganic and an outer organic layer prevails today,[65] but the spatial-temporal details remain a topic of current research. Continuum modeling shows that the interplay of formation reactions at different reduction potentials leads to a multi-layer SEI with a compact inner layer and a porous outer layer.[78] SEI structure and composition depends on the electrode surface and can vary on a single electrode particle.[67] Recent cryo-TEM experiments indicate that large porous outer layers might be a failure mode[79] and clarify that inorganic compounds are not only present near the electrode, but can be embedded in an amorphous matrix or aligned near the electrolyte depending on the electrolyte.[75] This hints at the role of grain boundaries for transport in the SEI.[80]

In the first few cycles, a compact SEI is formed on graphite electrodes governed by electron tunneling and reaction kinetics.[80,81] However, SEI growth does not stop after this formation phase. Under long-term storage conditions, SEI thickness grows with the square-root-of-time, clearly signaling transport limited growth.[82] The nature of the relevant transport mechanisms is still being debated (**Figure 5**).[63] Can solvent molecules diffuse through the SEI pores,[83] or do electrons move through the compact SEI, either via a conduction band[84,85] or via diffusion through localized states?[86] The measured dependence of capacity fade on electrode potential hints at the latter.[72] In this case, the electron density in the SEI and its growth rate depend exponentially on the electrode potential. Such a dependence can explain[81,87] that SEI is mainly growing during intercalation as seen by differential capacity analysis.[73] Microstructure-resolved 3D simulations of porous electrodes have revealed that the exponential dependence of SEI growth on electrode potential leads to inhomogeneous SEI growth throughout the electrode.[88,89] These results of physical-chemical models give important input for improved battery management systems that have to deal with the continued loss of battery capacity.[90]

Current research is directed toward high-capacity electrodes like silicon and lithium metal which undergo a large volume change during intercalation/deintercalation. This exerts stress on the SEI at the electrode surface. Atomic force microscopy and focused ion beam transmission electron microscopy can quantify the regimes of elastic deformation,[91,92] plastic flow, and fracture. Mechanical models make clear the role of plastic flow in the SEI on silicon electrodes.[93] The outer SEI is prone to failure.[92] Its continued reformation leads to linear-in-time capacity fade increasing with applied current.[83,94] Understanding and mitigating this effect is crucial for the development of high-energy silicon electrodes.

Plating lithium on the negative electrode can become thermodynamically and kinetically favorable at large current densities and low temperatures.[95,96] It is important to note the heterogeneity of this phenomenon in porous electrodes[97] and its coupling to inhomogeneous SEI growth deserves future attention. A further challenge, especially for lithium metal electrodes, is the tendency of electrochemically deposited lithium to form structures with high surface areas, e.g., nano-scale whiskers and micro-scale dendrites,[98] which entails a continued formation of fresh SEI and lithium loss. Modelling can classify the emergent structures.[69] Dendrites are limited by transport properties,[69] whereas the theoretical description of whiskers is based on surface properties like SEI elasticity.[99]

2.3. Deep-Learned Models and Explainable AI

Deep generative models are capable of learning the underlying probability distributions in the input space[100] and can thus be used to encode physics driven constraints in battery materials from large validated datasets. This is fundamentally different from more popular machine learning models that predict battery materials properties like energy,[101,102] electron density,[103] intercalation potential,[104] etc. from its structure which has been covered in detail recently.[105] A range of algorithms have been tried out with varying level of success, among which most popular and versatile have been, variational auto encoders[106] and generative adversarial networks.[107] Both generative schemes can operate on image (pixels and voxels),[108–110] graph (nodes and edges)[111–113] (e.g., SMILES) and fingerprint-vector based representations[114]

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of materials or molecules. Full diversity of representations are usable for atomic scale structures, but at the continuum scale, image-based representations are most useful.[109,115] Beyond constraints learnt from data, additional known physical laws and constraints can be imposed on the generation.[116,117] Generation of new structures at the atomic scale can also be defined as a Markov decision process and deep reinforcement learning[118,119] working with a physics simulation environment can be used to train a structure generator. Finally, if the underlying correlations (that define the physical viability of the structure) are simple enough to be defined as heuristic rules, a logic-based generative scheme can be defined using them.[120]

This is an emerging field[59] and the application of generative models to explore structural space has so far been focused on small molecules, which are easy to represent in many possible ways and can quickly be tested for goodness of fit. Solid state crystals structures owing to periodic boundary condition and inherent symmetries, are more challenging and thus only few practical cases have been demonstrated.[108,121,122] On the other hand, at the microstructural scale, generative models of solid materials have been very successful even for complex heterogeneous systems and limited datasets.[109,110,115–117,123–125] Outstanding developments in convolutional networks in image processing and generation have been key to that success.

Developments in the area of "explainable AI (XAI)"[126,127] methods have provided a new path toward utilization of big data[55] in scientific discovery via data driven realization of chemical laws and design principles. This approach is fundamentally different from the originally envisioned direction for big data,[128] where hypothesis driven research would be displaced by pure algorithmic approaches. Instead, XAI can usher in a quantum leap in hypothesis-driven research where machine learning autonomously generates different hypotheses and tests, leading to new scientific intuitions—following the current best practices but bringing enormous speedup relying on exa-scale computing. This can be achieved as autonomous agents, not just human researchers, can now do the generation of scientific understanding.[129] The capability of deep learning to go beyond numerical prediction and enter the realm of scientific insight and hypothesis generation opens the door to accelerate the discovery of conceptual understanding in some of the most challenging areas like battery systems. Furthermore, the same autonomous analysis can be performed for high dimensional correlations that are beyond human intuition. For example, moving away from simple linear descriptors[130] to complex ones combining multiple features.[131]

Descriptor search for key properties have been central to chemical and materials sciences as they guide exploration of design space efficiently for target materials properties with limited experimental/computational cost.[132–135] Simple descriptors can also be seen as input to human-understandable design principles, which are a simple, actionable form of the underlying theoretical paradigm. For example, electronegativity of neighboring atoms might be a simple descriptor to optimize ion intercalation potential in battery cathode materials,[136] although the electronic structure effects causing the correlation are highly complex and nonlinear. Even approximate property estimation through descriptors are valuable as a multi-step screening[137] based design protocol is computationally inexpensive using semi-accurate descriptors for target properties. Descriptors can be discovered relatively easily from statistical/symbolic models or ML models with limited complexity.[138–141] These approaches have become popular in last few years in the materials science community specifically for batteries as well toward data driven descriptor search.[134,142–144]

2.4. Impact of the SoA on the Implementation of BIG-MAP

The methodological developments in the areas autonomous synthesis and experimentation, multi-scale and multi-domain approach for battery interfaces, and deep-learned models and explainable AI, pave the way for materials acceleration platforms like BIG-MAP that depend critically on the availability, modularity, and interoperability of new approaches.

3. State of the Art: Self-Healing

3.1. Introduction

Self-healing in materials science is a relatively new research direction with some examples such as self-healing fiber-reinforced polymer composites, self-healing coatings, self-healing cementitious materials, self-healing ceramics, self-healing organic dyes, self-healing concrete molecules[145–148] and with known examples developed for different battery chemistries summarized in recent comprehensive review papers.[149,150] A research direction of the Battery 2030+ is well explained in the original roadmap [4] and the new roadmap paper by Edström et al. in this issue. while within the perspective paper, authors are providing their view in the field of self-healing functionalities in Li-ion batteries.[151] Batteries are composed of components and interphases which are operating in thermodynamically metastable environment and are prone to different degradation. Use of the preventive steps (coatings, doping, additives, etc.) can improve the quality, reliability, lifetime and safety (QRLS) of cells, but since they are operating in diverse conditions (temperature, current densities, mechanical stress, etc.) which have an important impact on ageing processes and consequently on their QRLS, battery cells undergo different ageing processes during their lifetime. Ageing processes can be divided into two larger groups: a) mechanical degradation and b) chemical and electrochemical degradation. Degradation mechanisms are strongly connected with battery chemistry (structure and quality of components and materials) and that calls for designed self-healing functionalities for each degradation process with a possibility of their vectorization within battery cells. Self-healing functionalities are divided into intrinsic (autonomous) and extrinsic (non-autonomous) with a need for external stimulus for triggering. Here we provide an overview over the state of the art of degradation mechanisms in Li-ion batteries and related self-healing functionalities.

3.2. Mechanical Degradation and Known Self-Healing Approaches

Mechanical fracture of high energy density materials for the next-generation Li-ion batteries is based on the large volumetric

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changes during lithium insertion/alloying and removal/de-alloying.[152–154] Silicon (Si) is an excellent example for testing different self-healing approaches since its degradation is a combination of different mechanisms. The major degradation occurs due to large volume expansion (≈300%).[155] Two major self-healing approaches are well described in the literature: a) the use of liquid metal alloys with a low melting point and b) the use of self-healing binders. The concept of self-healing metallic alloys was demonstrated by Deshpande et al.,[156] by using gallium. They showed that lithium gallium alloy (Li_2Ga) spontaneously heals micro-cracks in the Li-ion batteries. Other low-melting-point elements like indium (In) and tin (Sn) were studied due to their relatively high stability under ambient conditions and their environmental acceptability.[157,158] Combinations of different metals yield binary or ternary alloys with different melting points. An example of a room-temperature liquid metal alloy based on an eutectic mixture of Ga-Sn showed improved cyclability and rate performance.[159] Although room temperature alloys provide a good direction for healing cracks, unstable SEI (solid electrolyte interface) remains a problem which can be mitigated with the combination of liquid metal alloy and polymer electrolyte.[160] The use of self-healing binders offers a promising way to maintain the integrity of the silicon electrode. This approach is using a strong chemical bonding of various functional binders such as sodium carboxymethyl cellulose (NaCMC),[161,162] poly(acrylic acid) (PAA),[163,164] poly(ethylene glycol) (PEG)[165] polyamide imide, polyamide (PAI),[166] guar gum (GG)[167] and sodium alginate[168] with silicon surface. Most of the studies have been obtained with nanosized silicon and some of the proposed binders cannot be used with micron-sized Si because of the lack of oxygen on the freshly pulverized Si surface.[167] Another important direction for the development of self-healing binders is the performance of Si electrodes having high areal capacity (>4 mAh cm^{-2}). In this direction, an elastic self-healing CA-PAA binder shows long-term stable cycling[155] due to the multiple hydrogen bonds formed by in situ cross-linking of water-soluble citric acid (CA) and poly (acrylic acid) (PAA) which can establish a reversible network for silicon particles.

The strategy of preventing the degradation processes in the layered cathode materials $LiNi_xMn_yCo_zO_2$ (NMC, $x + y + z = 1$) is based on the bulk structure/surface modifications and microstructure design engineering. Such an approach is considered rather as a preventive step than a curative self-healing approach. A possible strategy would be a design of binders with robust mechanical and adhesive properties, namely properties of PVdF are not sufficient to maintain integrity caused by internal stress at high voltages.[169] Replacing the PVdF with highly viscoelastic, branched polymers capable of strong supramolecular interactions[170] is highly desirable.

3.3. Chemical and Electrochemical Degradation Mechanisms

Electrolyte degradation / formation of the SEI: Formation of the SEI enables cyclability and is responsible for the additional resistance which changes during battery aging. Changes in the thickness and composition are potentially caused by thermal decomposition, by conversion of the organic part of SEI leading to inorganic salts and by cathode degradation products.[171] Due to this continuous growth, the impedance rise is responsible for irreversible consumption of up to 10 at% of lithium in the cell and for electrolyte degradation.[172] To address problems with the SEI growth and evolution, three different types of self-healing electrolytes have been proposed. The self-healing functionalities in gel polymer are provided by the polymer matrix and ionic liquids act as plasticizers, while the additive, lithium salts, and liquid solvent ensure ionic conductivity.[173,174] Self-healing single-ion conducting polymer electrolytes[175,176] are a viable alternative to cross-linked polymers due to improved mechanical stability compared to cross-linked polymers. A second option are composite polymer electrolytes[177] with grafted ceramic particles that provide a fast conduction channel for lithium. The third option are crosslinked solid polymer electrolyte networks capable of fast intrinsic self-healing and with high stretchability.[178] For instance, the recovery of conductivity and rheological properties following damage in polymer electrolyte networks with dynamic covalent cross-links has recently been investigated. These polymer networks act as solids at room temperature but can flow at high temperatures.[179]

Gas evolution in batteries is generally connected with SEI formation, SEI ageing and with electrolyte oxidation at high voltage followed by oxygen release from the oxide structure and metal dissolution. Among the transition metals used in batteries, manganese dissolution is the most known problem[180,181] and the mechanism of dissolution has been the subject of many different studies which are very well summarized in a recent review.[182] Although, the direct capacity drop due to dissolution of Mn is only 20–33%,[183] increased polarization in the cell caused by the growth of SEI and dropped capacity cause additional capacity fading. Self-healing solutions are related to the scavenging of parasitic species, for instance HF, which not only prevents metal dissolution but also inhibits the chemical crosstalk with the anode and prevents oxidation of the electrolyte. The structure, homogeneity and stability of the cathode electrolyte interface (CEI) are important.[184] It can be formed by using functional electrolyte additives, that support the construction of a robust CEI and provide scavenging functions.[185] The disadvantage of this approach is the consumption of additives and poor control of CEI growth. Another possible direction is the use of designed binders and separators, which can act as scavengers and prevent cross-communication between electrodes.[186] This has been demonstrated through chelating agents like crown ethers (18-crown-6,[187] aza-15-crown-5[188]), disodium iminodiacetate,[189] etc. Other additives, such as tetraethyl orthosilicate (TEOS),[190] poly(4-vinyl pyridine) (P4VP),[191] terpyridine/polyvinylpyrrolidone (TPY/PVP),[192] and dilithium maleate[193] enable scavenging of acidic species like HF when added to separators or binders.[194,195]

Lithium plating/dendrite formation in Li-ion batteries depends on the cell quality and its use.[196,197] The charging rate is critical since it has to be adopted by the rate of lithium-ion diffusion into graphite. To design preventive and curative functionalities which can help to avoid the formation of dendrites on the surface of graphite or metallic lithium anodes, different groups are studying approaches how to stabilize the use of metallic lithium. Here the common agreement is that a stable SEI should enable suppression of dendrites and be

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able to repair damages, that is, it should combine preventive and curative functionalities.[198] Most of the studies are using thick Li metal anodes (>250 µm) with an excess of electrolyte in a symmetric cell configuration applying low current densities with low capacity. For practical applications, it is necessary to demonstrate the applicability of the protection layer by using thin lithium foil over several hundreds of cycles with a capacity over 1.0 mAh cm^{-1}[2] and by using a lean electrolyte amount. Even though these requirements are demanding, some results obtained in the asymmetric cell configuration (using Cu as a counter electrode) show high Coulombic efficiency (CE) and good stability. For instance, alucene-coated lithium was tested over 160 cycles with a CE of 99.5%.[199] Wang et al. demonstrated 99.1% CE on a coated 3D carbon host at high current density (2 mA cm^{-1}[2]) and high capacity (4 mAh cm^{-1}[2]).[200] When combined in a full cell design with a NMC523 cathode, a cell was cycled over 200 cycles under lean electrolyte (7 µL $(mAh)^{-1}$), limited Li amount (1.9-fold Li-excess) and high areal capacity (3.4 mAh cm^{-1}[2]) conditions. Those are only two examples from the literature since there is still a need for further improvements using practical and saleable approaches to demonstrate the safe use of lithium metal and other anodes at ambient temperature conditions.

Yet another important issue in the Li-ion battery ageing is electrical contact loss, which is related to the loss of the internal electrical contacts in the electrode.[201] The reasons are manifold, migration and agglomeration of conductive carbon, particle fracture,[202] formation of a thick SEI[203,204] and gas evolution.[205] The most elegant solution is the use of microcapsules which can deliver conductive additives upon external stimulus.[206,207]

This short overview of degradation mechanisms and related self-healing approaches points out the complexity of the field since most of approaches were demonstrated in the half-cell combination using laboratory cell design. Different degradation processes call for vectorization of self-healing functionalities with the focus on their manufacturability, sustainability, and recyclability.

4. State of the Art: Sensing

4.1. Introduction

Physical and chemical characterizations, either ex situ or operando, have long been used to examine battery chemistries. However, characterization techniques can hardly be implemented into commercial battery cells. Novel approaches are thus needed to probe the battery chemistry and physical properties during operation and to infer its health status. Among these strategies, electrochemical sensing, thermal and strain sensing, optical sensing as well as acoustic sensing are currently pursued.

4.2. Electrochemical Sensors

Sensing technologies have first been implemented into Pb-acid technology with great advances made with the implementation of electrochemical impedance spectroscopy (EIS) which allows tracking the evolution of cell resistance during operation, thus accessing their state of health (SoH).[208] Indeed, battery chemical effects such as the growth of resistive layers (i.e., SEI) and the shuttling of dissolved redox species between both electrodes, which is often associated with the dissolution of metals, can be probed by recording the electrochemical behavior of the cells, which act as a sensor.[208] However more widespread use in battery sensing outside the laboratory is being hampered by the challenging task of miniaturizing probes and sensors, which could however benefits from recent advances in the field of biophysics/chemistry. Hence, miniaturization down to micro or even nano dimensions is now envisioned for electrochemical sensors using several mechanical, chemical, and electrochemical protocols to prevent environmental artefacts (e.g., convection).[209] Combining miniaturization with advanced electrochemical techniques (pulse), the incorporation of new detection microsystems into batteries is now achievable.

Nowadays, a combination of electrochemical measurements that include EIS and resistance measurements, current pulse measurements, coulomb counting, and open circuit voltage-based estimations is used to estimate the battery state-of-charge (SoC).[210] The major challenge in electrochemical battery diagnostics resides in the design of (electro)chemically stable (quasi-)reference electrodes (REs). Indeed, such reference electrodes are necessary to decouple effects at both electrodes in voltammetric/amperometric and/or potentiometric detection regimes. Mainly, reference electrodes suffer from the corrosive environment of the batteries, related to the use of carbonate-based electrolytes in Li- or Na-ion batteries or acid/alkaline for aqueous batteries.[211] Also, to be accurate, an RE must be perfectly placed in relation to the other components, to avoid current lines to disturb the signals and prevent experimental artifacts.[212] Therefore, efforts are made to develop reliable, user-friendly, chemically stable electrochemical sensors relying on RE.

4.3. Thermal, Strain, and Gauge Sensors

Thermal battery management system (TBMS) models have long been validated with knowledge of thermal mapping of cells.[213] Different technologies for temperature sensors for use in TBMS exist, that are differing in their accuracy and in the convenience with which they can be inserted within the cell. The main four leading types being resistance temperature detectors (RTDs),[214] thermally sensitive resistors (thermistors),[214] thermocouples,[215] and fiber Bragg grating (FBG) optical sensors.[216] Due to their thickness (1 mm), thermistors are positioned only on the top of the cell (not on the surface), unlike RTDs that are much thinner (100 µm).[214] For instance, by screen printing thermal sensor arrays on the surface of 18650 cell casings, mapping of the longitudinal surface variation in cell temperature could be achieved with an accuracy of ±1 °C.[214] Nevertheless, the predictive power and the accuracy of current TBMS model suffers from the lack of information regarding the inside of the cell. To tackle this issue, attempts to implement thermocouples within the battery cell (it being 18 650 and/or pouch cells) were thus made. Hence, a successful

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electrocardiogram of a 25 Ah battery with temperature contours within the cell was realized by combining the use of thermocouples embedded into the cell with some outside of the cell casing.[215] Such valuable information are therefore needed to validate thermo–electrochemical models, with nevertheless limitations associated with positioning and wiring all the thermocouples without affecting the performance and the air-tightness of the cell. Toward that goal, infrared thermography could be envisioned. However, poor spatial resolution, limited temperature accuracy and susceptibility to background noise limit the implementation of such technique.[213]

More recently, Dahn and coworkers have introduced differential thermal analysis (DTA) to measure in a non-destructive way the melting point of liquid electrolyte and correlate it with the SoH of the battery.[217] However, this technique remains ex situ, and cannot be miniaturized to be used in commercial cells.

Critically important is also the development of methods to sense intercalation strain and cell pressure.[218] Indeed, such techniques provide uniquely important information regarding the SEI dynamics, which impacts the SoC and SoH of batteries. Early on, in situ strain gauge measurements were developed to probe the total volume change of Ni-Cd batteries. This and was then extended to the study of commercial Li-ion ($LiCoO_2$/C and similar cells). In the latter case, it allows measuring the strain associated with phase transition in intercalation materials. Volume variation in the cell could also be detected allowing for instance, as recently demonstrated by Dahn, to detect SEI growth owing to its associated irreversible volume growth by simply using external sensors. Hence, a correlation between capacity retention and irreversible pressure increase could be made, providing valuable insights into the battery state-of-life.[219] The use of strain sensors however fails at providing spatial information owing to their location outside the cell. Hence, limited information is collected in order to improve SoC and SoH of batteries.

4.4. Optical Sensors

The most studied optical sensors are the so-called fiber Bragg grating (FBG) sensors, as they can access local temperature.[216,220] The main demonstration of using FBG to monitor Li-ion batteries was made by PARC (a Xerox company) which, by using embedded FBG sensors attached to the electrode, were able to estimate the SoC with less than 2.5% error of the batteries following the strain measured by the sensors.[221]

Nevertheless, one limitation of FBGs is that they rely on changes of the refractive index between the core and cladding to decouple pressure and temperature. This can be achieved by the use of micro-structured optical fibers (MOFs), also known as photonic crystal fibers (PCFs).[222] Indeed, MOFs embed air holes inside the fiber core, the patterning allowing to manipulate the waveguide structure and thus achieve total internal reflection and to measure temperature and pressure independently with a single fiber.

Despite still being in its infancy, FBGs were recently incorporated into commercial 18 650 sodium-ion cells.[223] By inscribing the FBGs both as single-mode fiber (SMF) and as MOF, the temperature and pressure evolutions inside the cells were tracked with high resolution. Further addition of surface/ambient FBGs together with a thermal model enabled the operando monitoring of heat generated during cell operation.[223] Such approach gives additional access to heat capacity contribution, which is inaccessible via conventional isothermal calorimetry, and thus benefits the designing of battery thermal management systems. The deeper analysis of heat information deciphered the thermodynamic parameters such as enthalpy and entropy, pertaining to the phase transitions and cell ageing. Furthermore, the simultaneous monitoring of heat and pressure decoded the exothermic reactions and gas evolutions during the SEI and CEI formations,[223,224] and thereby accelerated the screening of the electrolyte additives. This work was further extended by showing that Rayleigh back scattering could be used to monitor the temperature in 18 650 cells along its axial dimension with a resolution up to 0.05 pixel mm^{-1} Rayleigh sensors, unlike FBGs, are less expensive to manufacture, but require a more expensive interrogation system and greater calculation resources to analyze the large amount of data generated.[225]

Finally, nano-plasmonic sensing (NPS) was introduced in 2017 to the field of batteries. Indeed, shifts in the wavelength of the plasmon resonance peak, that is, electron oscillation, can be correlated to changes in the refractive index (RI) near the sensor surface (<100 nm).[226] Owing to their ability to focus, amplify, and manipulate optical signals, these sensors can be used for monitoring operando phenomena, including the growth of SEI, the lithium intercalation/deintercalation from electrodes, or the variation in ion concentrations at the local scale. Nevertheless, these sensors suffer from low stability upon cycling and exposure to liquid electrolyte associated with the deposition of a metallic plasmonic nanostructure on top of the fiber.

4.5. Acoustic Sensors

Upon cycling, batteries undergo variations in volume as great as 10%, resulting in mechanical stress (i.e., cracking) in the battery's materials. Hence, listening to the elastic acoustic waves generated during volume changes of battery materials before to analyze these waves[227] was used early for the Pb-acid and Ni-MH technologies[228] before being adapted for LIBs.[229,230] Indeed, AE is very effective for monitoring the different steps associated with battery operation, which includes the formatting step, as well as for detecting abusive operation conditions resulting in excessive stress in materials and potential safety issues. However, the main limitations of the acoustic emission (AE) technique are the lack of spatial resolution and the minimum stress undergone by materials for them to generate acoustic waves, which can be partially addressed by generating ultrasonic acoustic waves with piezoelectric transducers, and measuring their speed of propagation across the battery to estimate the SoC of LIBs.[231]

The development of high-sensitivity, high-reproducibility, high accuracy, and low-cost sensors in the coming years is critical in order realize the smart batteries of the future. This will be made possible by the development of novel sensing techniques, bearing in mind that their manufacturability must be ensured without significant increase of cost. Furthermore, the development of an efficient battery management system (BMS)

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must be made to ensure communications between the novel smart functionalities of future batteries.

5. State of the Art: Manufacturability

5.1. Introduction

Battery manufacturing is a concept covering a large area. In the present context, it may refer to battery cells, modules or battery packs. Accordingly, this section will be focused at the cell level, understood as the physical place where any future battery technology will take its basic and unmistakable form.

The availability of a new generation of advanced battery materials and components will open a new avenue for improving battery technologies. These new battery technologies will need to face progressive phases to bring new ideas from concept to prototypes through validation before putting them in place in a full industrial implementation. First, they will need to prove their potential at first stage of the lab scale, and second, the feasibility of their upscaling will need to be addressed for ultimate deployment of the cell into industrial level.

Manufacturing of benchmark battery technologies is addressed in this section from the perspective of digital manufacturing. The potential of physics-based modelling and of AI are discussed to apply these computational tools to support the development of digital twins both for innovative cell design (including geometry) and cell manufacturing routes, avoiding or reducing the classical trial and error approaches. Such digital twin concepts also bring a promising path toward a more sustainable and efficient the manufacturing methodologies, thanks to their capabilities to advance the understanding and optimization of parameters and their influence in the final battery cell product, prior to its physical implementation. A detailed description of the horizon in the digital battery manufacturing can be found in the chapter manufacturability of the BATTERY 2030+ Roadmap.[4]

5.2. Current Status

Lithium ion batteries are today's battery technology of reference. Other battery technologies exist as well, sharing the basic underlying electrochemical and structural concepts, but they may differ substantially in their physical disposition of their elements.

Automated battery cell manufacturing is well established today in Lithium ion batteries. Lithium ion batteries currently comprise a wide range of technological approaches, ranging from so-called generation 1 to generations 2 (a and b) and 3 (again both in its a and b versions) based on classifications published by National Platform Electromobility and adopted by JRC,[232] all of them sharing a basic layout based on electrodes coated on top of thin metal sheets.

5.3. Battery Cell Manufacturing

The battery cell manufacturing process can be divided into the categories: electrode production, cell assembly and cell finishing as can be seen in **Figure 6**.[233] This distinction is valid across the myriad battery chemistries. For simplicity and clarity, the discussion in this section will focus on Li-ion cell manufacturing.

In electrode manufacturing, raw active materials and additives are combined to create electrode structures tailored to the needs of the cell. Electrodes are primarily composed of active materials, which account for roughly 90% of the electrode by mass. However, many active materials must be mixed with other non-active components, such as conductive carbons and polymeric binders, to enhance electronic conductivity and to ensure good contact and adhesion.[234]

The electrode manufacturing process begins by dispersing the dry materials into a liquid solution to create a slurry. Current manufacturing processes use an aqueous slurry for the negative electrode materials (e.g., graphite). The solvent of choice for cathode slurry preparation is the organic solvent *N*-Methyl-2-pyrrolidone (NMP). This is problematic, however, since NMP it toxic to reproduction,[235–239] while processes involving alternative organic solvent are still at a research stage.[240,241]Some processes have been developed featuring aqueous slurries for positive electrode manufacturing. The slurries are thoroughly mixed and the properties including viscosity and pH are carefully controlled.[242–250]

After uniformly mixing, the resulting slurry is cast onto a metal foil current collector and dried in a convection oven.

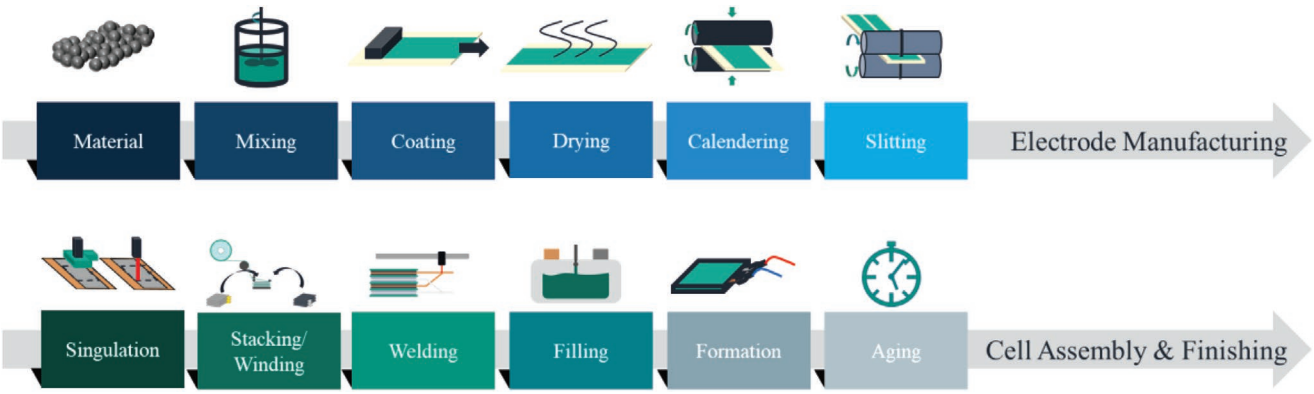

Figure 6. Simplified overview of the Li-ion battery cell manufacturing process chain. Figure designed by Kamal Husseini and Janna Ruhland.

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As the solvent evaporates from the slurry, a porous solid active layer is formed. Electrode drying under vacuum can take a wide range of time with some electrodes taking 12–24 h at 120 °C to be completely dry.[251] In commercial applications, an NMP recovery system must be in place during the drying process to recover evaporated NMP due to the high cost and potential environmental pollution.[252,253] On-going efforts to reduce the production cost of Li-ion batteries include the pursuit of positive electrode slurries based on water or removing the need for a solvent altogether.[254–259]

The dried electrodes are calendared to guarantee the homogeneous thickness of the active layer, improve electronic conductivity, and increase electrode energy density.[260–263] In a roll-to-roll process, multiple rows of electrodes can be coated onto a single large current collector roll. In this case, the larger roll of calendared electrodes (the mother roll) is then slit into smaller rolls (the daughter rolls) each containing one row of electrodes. The completed electrode rolls may be vacuum dried to remove any remaining solvent prior to cell assembly.

Li-ion battery cells can be manufactured in a variety of formats: cylindrical, prismatic, and pouch type. For each cell format, electrodes must be cut from the roll to the appropriate dimensions. Electrode cutting may be done mechanically (e.g., stamping) or using laser-cutting. For cylindrical and pouch cells, long strips of electrodes are wound together with a porous separator. The resulting electrode and separator assembly, sometimes referred to in the industry as a jelly roll, is then inserted into a container. In pouch cells, electrodes may be Z-folded from a continuous sheet or cut into individual sheets and stacked together. For stacked pouch cells, an uncoated section of the current collector, known as the tab, is left in each layer. These tabs are then welded together to create single contacts for the positive and negative electrodes before the cell is inserted in the pouch and injected with electrolyte. Pouch cell casings leave some extra space known as a gas bag to collect and remove gas that can evolve during the remainder of the manufacturing process.[264–267]

To finish the cell, it is cycled under precisely defined conditions to support the formation of the solid-electrolyte interphase in the negative electrode. This formation process can last up to 24 h and may evolve significant amounts of gas in the cell. The gas can be removed from cylindrical and prismatic cells through a unidirectional valve. In pouch cells, the gas is collected in the gas bag, sealed off from the cell, and the gas bag is cut off. After the formation process the ageing process is finalizing the battery cell.

Finally, some quality control measurements may be applied to the manufactured cell before delivery. Capacity and resistance are measured, and it is stored several days in open circuit to monitor self-discharge.[268,269]

Additive manufacturing has been used in the past to make these porous electrodes for lithium-ion batteries but because of the manufacturing process their design has been limited. A new method of 3D printing battery electrodes that create a micro lattice structure with controlled porosity was recently developed[270] which demonstrated vastly improved capacity and charge-discharge rates for lithium-ion batteries. Nevertheless, these concepts and techniques are still in their infancy and therefore will certainly need to be explored in the near future.

5.4. Multiphysics Modelling and Artificial Intelligence in Cell Design and Manufacturing

Multiphysics modelling approaches can be of a great importance in battery design and manufacturing to:

1) Accelerate new cell designs in terms of the required targets (e.g., cell energy density, cell lifetime) and efficiency (e.g., by ensuring the preservation of sensing and self-healing functionalities of the materials being integrated in future batteries).
2) Accelerate the optimization of existing and future manufacturing processes in terms of cell chemistry, manufacturing costs and sustainability/environmental impact (e.g., in terms of amount of energy consumed) and assist in the development of new manufacturing processes.

Li-ion cell design has been the subject of study for decades to improve not just energy density but also cycling stability. Currently, conventional cell design uses ad-hoc rules to link design variables and manufacturing parameters with cell performance, requiring a time-consuming experimental work. This is the reason why researchers demand characterization and computational simulations to increase the understanding of the phenomena happening at the cell in different scales and physical domains in an efficient way. The main barriers to effectively implement them as a work basis in the industry are the high computational costs of the most accurate computational models and the lack of understanding on how the phenomena at different scales and physical domains interact and finally influence in battery performance. Simulation tools with advanced numerical algorithms are required to give accurate results at reasonable computational costs, therefore unlocking its generalized use. Therefore, coupling traditional physics-based models to advanced optimization algorithms in AI frameworks becomes crucial to optimize cell design in a cost-effective way.

As mentioned above, conventional cell designs have relied on ad-hoc rules. However, to identify the optimal designs of a cell, the design process has to include all relevant design variables, and the interactions between various parameters on cell performance need to be considered. Dealing experimentally with such a huge set of parameters can become a “nightmare” and the simulation-based approaches are of great importance as provides understanding of the whole mechanisms at different scales and domains.

Most of the physics-based models used for battery cell optimization are supported on the Doyle's pseudo 2D (p2D) approach,[271] combining the porous electrode theory proposed by Newman et al.[272] and the concentrated solution theory. The P2D model has been used to optimize cell components such as the cathode and anode thickness, porosity, particle size and many other important electrode parameters.[273–278] Another example of the application of the model-based design is presented by Ayerbe et al.,[279] where a set of dimensionless parameters is derived and demonstrated the represent an effective and inexpensive tool to guide on cell designs in an early stage of the development of a new cell chemistry.

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However, the computational cost can be prohibitively high if a p2D model is directly applied for battery design. In simulation-based battery design, thousands of simulations are often required to determine the optimal design variables. Moreover, the complex non-linear nature of the battery model may result in convergence problems under some sets of design variables. Besides, sensitivity of the design variables is also difficult to analyze due to the very high computational cost. Without sensitivity analysis the possible reduction of design space through elimination of insensitive design variables becomes inapplicable. Surrogate models based on ML or reduced order models have the potential to reduce the computational burden of battery design by several orders-of-magnitude.[280–284]

The manufacturing process of lithium ion batteries involves physicochemical mechanisms occurring at multiple spatial and temporal scales. For instance, as it was mentioned previously the rheology and self-organization phenomena of slurries depend on interaction mechanisms between their constituents at the molecular/particle levels (solvent, active material, additives, binder) while the calendaring depend on the local but also overall mechanical properties of the composite (electrode) material. The optimization of such a process only based on trial-error is highly inefficient and one cannot guarantee that the optimal process parameters with regard to energy consumption are adopted. It is therefore important to dispose also of computational models able to predict of how the adopted fabrication parameters impact the final fabricated cell and associated performance and durability: recent works are moving forward into this direction.[285,286]

Among the modeling efforts carried out, Brownian Dynamics simulations in the three spatial dimensions have been applied to simulate suspensions of active material particles at the local level and analyze slurries properties such as particle percolation and agglomeration phenomena as function of particle sizes and composition.[287,288]

Furthermore, stochastic models, based on Monte Carlo approaches in two spatial dimensions have been applied to simulate slurries and the effect of solvent evaporation on the resulting coating structural properties, characterized by the particle's location and bi-dimensional porosity.[289]

Coarse-Grained Molecular Dynamics (CGMD) models have been recently proposed that allow the 3D simulation of slurries and of the effect of the solvent evaporation on the structural properties of the resulting coating.[285,286,290,291] Such models address the scales disparity problem through a “bead” type representation of sets of atoms and molecules: for instance, an active material particle is represented as a unique particle instead without atomic-scale resolution, but the model still allows the main physicochemical interactions with other slurry constituents. The CGMD modelling approach was recently upgraded to capture the effect of evaporation rate on carbon-binder migration and subsequent electrode heterogeneities.[292]

The electrode calendaring step has been also addressed using primarily Discrete Element Method models, well established for simulating powder mechanics and transferred to the battery field.[293–295] Similarly to an MD approach, these models rely on the resolution of Newton equations accounting by particles mechanical interaction. They allow analyzing the resulting particle assemblies as a function of the applied pressure and particle shape and size distribution, in terms of percolation and associated properties of interest for the optimal battery cell operation such as the associated electronic conductivity. Their sequential linking with 3D-resolved electrochemical/transport models allow assessing the impact of the manufacturing parameters, and the calendaring in particular, on the electrode performance.[294,296]

Lattice Boltzmann Method simulations have been used to address the step of electrolyte filling in two[297] and in three spatial dimensions.[298] These models allow investigating the effect of several experimentally controllable parameters, such as the compression ratio of the porous electrode, on wettability with respect to variations of parameters such as the electrolyte/electrode contact angle and the porosity.

Data-driven modeling efforts have also been reported at larger scales (machinery level). A multi-level simulation concept was recently presented,[299] which allows evaluating the impact of varying process parameters on battery properties (including performance) distributions.

ML modeling efforts aimed at accelerating the manufacturing process optimization have been reported recently. At the process level, ML works have been focusing on the prediction of formulation, solid content and slurry viscosity on electrode active material loading and porosity.[300] Other ML studies have been directed at the optimization of the calendaring parameters for optimal electrode conductivity and performance.[300] ML has been also used to predict three-dimensional electrolyte infiltration in porous battery electrodes with significantly lower computational costs (few seconds) than is required by physics-based simulations (several hours to few days).[301] Such approaches have been extended toward the accelerated optimization of the manufacturing process of solid-state electrolytes for battery applications.[302]

At the machinery level a multi-level ML study was presented,[299] which allows evaluating the impact of varying process parameters on battery properties (including performance) distributions. Several works present workflow strategies for the use of data-driven approaches to acquire data along the manufacturing process, including technical building services and cell diagnostics, with the objective of having a better control of the whole manufacturing chain.[303,304] Within this context, data mining methods have been used to analyze the production process from data extracted from it to identify the key quality drivers of produced battery cells. Methods used include artificial neural networks, random forest and decision trees, giving different levels of accuracy. Some authors have been also focused on process induced uncertainties and their propagation throughout the process chain to enable identification of the most relevant process parameters to lower manufacturing discard.[305]

The most advanced computational modeling approach to optimize the manufacturing process would probably be based on the integrative coupling of high-throughput characterization with ML and physics-based computational modeling. Duquesnoy et al. recently demonstrated this idea for the electrode calendaring process, paving the way toward multi-fidelity digital twins of the entire electrode manufacturing process.[306]

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5.5. Conclusion and Outlook

All in all, modern battery manufacturing processes should emphasize in pursuing the following goals:

- Accelerate the development of new cell designs in terms of performance, efficiency, and sustainability. Enhancing Multiphysics models, incorporating the critical steps of the formation and ageing steps, and alleviating the computational burden by implementation of ML algorithms or reduced order methods would facilitate to build more accurate and efficient tools. Even coupling these models with advanced optimization algorithms within AI frameworks would accelerate the inverse design of electrodes and cells.
- Closing the loop between cell design and cell manufacturing development, where the cell performance and ageing aspects should be considered prior to the manufacturing phase, through a holistic digital twin covering the whole battery cell manufacturing chain, from materials to cell usage conditions.
- Accelerate the optimization of existing and future manufacturing processes in terms of cell chemistry, manufacturing costs, and sustainability/environmental impact by building computationally efficient and accurate digital twins of the manufacturing process.
- Accelerate the development of additive manufacturing for the rapid near-industry production of battery cell prototypes and for the production of 3D structures of the electrodes.

6. State of the Art: Recyclability

6.1. Background Recycling

The increasing electrification in today's society raises the demand for battery recycling processes that are not only efficient and cost-effective, but also sustainable. In this way, the resource requirements and thus the ecological footprint of battery technologies could be reduced while increasing their success. Hence, optimization and automation of recycling processes is needed to meet the forecasted need. Despite this underlying demand, at present only a few battery recycling companies in Europe are operating on an industrial scale. At the same time, there is a trend toward new pilot scale companies in battery recycling and the application of hydrometallurgical processes. As current and future challenges regarding battery recycling have already been outlined in the BATTERY 2030+ Roadmap,[4] this chapter focuses on currently applied recycling processes and pathways for Li-Ion batteries (LIBs).

6.2. Regulation in Europe

The rising demand in Europe led to the implementation of a fit-for-purpose Batteries Directive (Directive 2006/66/EC),[307] which has already been outlined in the BATTERY 2030+ Roadmap.[4] Due to various shortcomings in the implementation and design of the Directive, identified in a Commission evaluation in 2019, a proposal for a new legislative act was published in 2020.[307] Currently under discussion is to increase the current collection and recycling rate for portable batteries from 45% to 65% in 2025 and 70% in 2030. All other batteries (e.g., from electric vehicles or stationary applications) must be fully collected and recycled while achieving high recovery rates. In addition, the proposed regulation establishes a framework for the re-use (second life) of batteries from electric vehicles.[308]

6.3. Recycling Processes

The recovery of the precious metals Co, Li, and Ni is a primary effort when recycling spent LIBs, beside Al from housing and Cu from current collectors.[309,310] A complete recovery of LIBs requires the application of different methods, making industrial LIB recyclers often use a combination of different technologies.[311] At present, recycling processes to recover critical materials such as Co, Li, or Ni can be classified into pyrometallurgical and hydrometallurgical methods.[309,311,312] Alternatively, the battery components are recovered as a whole in direct recycling processes. Several pre-treatment processes to deactivate the battery and separate the battery to its individual components precede this.[311] **Figure 7** shows an overview of the currently existing recycling processes.

6.3.1. Collection and Transportation

Since LIBs are classified as hazardous materials, strict safety measures (special transport containers, warning signs, packaging, etc.) against short circuits or leakage of the electrolyte must be observed during collection as well as transport. These high safety requirements related to waste logistics result in significant costs for battery recycling.[314] Since a variety of batteries exists, differing in size, weight, capacity and cell chemistry, collection and transportation is also subject to increased complexity. Depending on the battery, the requirements for packaging (strong outer packaging, protection against short circuit, etc.) and shipping (road, rail, sea freight, airfreight, etc.) differ.[315] Currently, both battery manufacturers and producers incorporating a battery into their products are responsible for the management of spent LIBs that they distribute, especially for financing collection and recycling systems.[316]

6.3.2. Pre-Treatment

Due to the complexity of the LIB structure, pretreatment processes are required aiming for maximized recovery rates, safe disposal of harmful components and safe treatment of LIBs. Thereby, safety aspects related to chemical, electrical, fire and explosion hazards have to be considered.[309,311,312,317] Especially for the downstream processes, pretreatment is seen as a key step.[312,314] The pretreatment processes described in the following originate from the mining industry and are therefore already mature and suitable for the recycling of spent LIBs. The processes often occur in combination.[309]

Discharging: A discharging process prior dismantling reduces the electrochemical energy content of the spent battery

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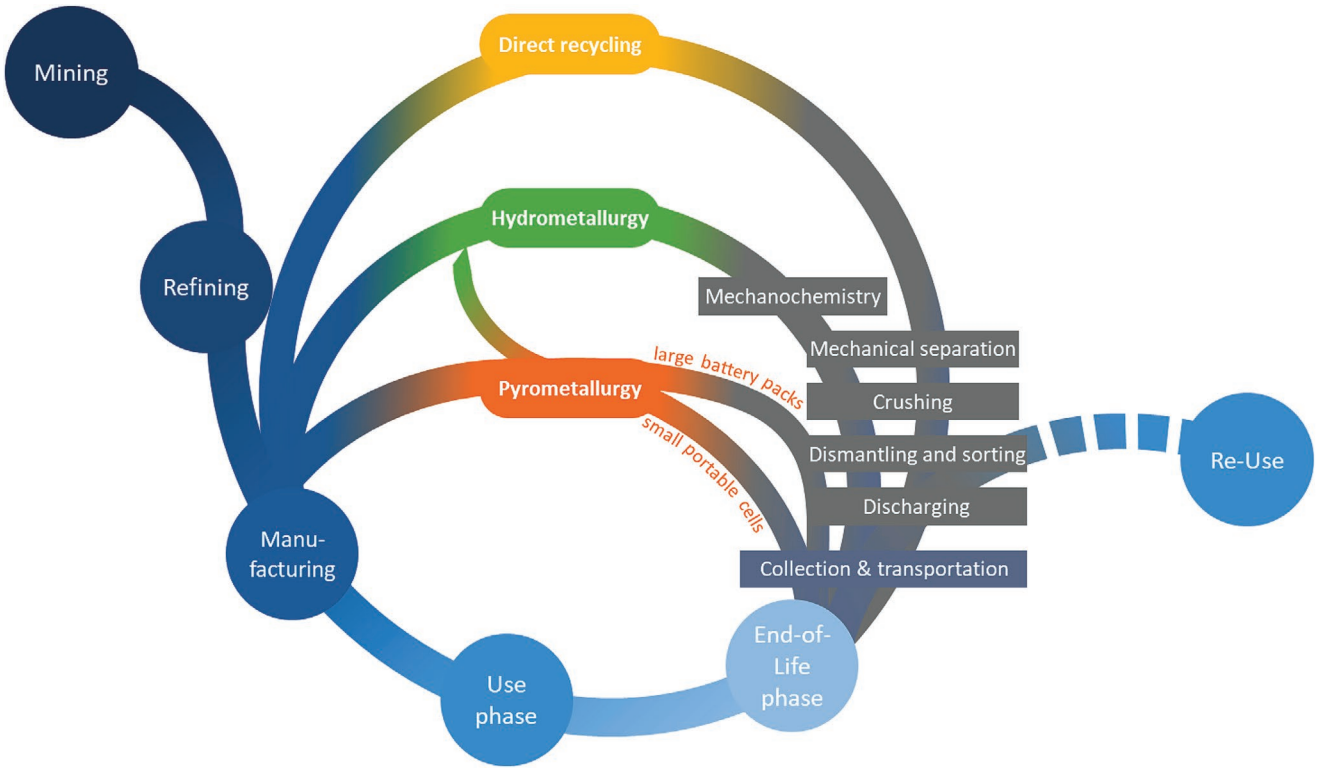

Figure 7. Overview of state-of-the-art recycling processes based on refs. [4] and [313].

and thus prevents self-ignition or short circuits.[314,316] Several discharging approaches are applied:

External Circuit: Ohmic discharge by a load carrying circuit is the most common and a viable method for large battery cells with high capacities.[314,318,319] Additionally, costs savings could be realized, if the electricity from the discharge can be recovered.[318]

Salt Brines: Discharging in salt brines, mainly NaCl, is a common method for low-voltage batteries. Due to the high electrolysis rate and the strong gas evolution, this method is unsuitable for high-voltage batteries.[309,314,319,320]

Thermal Treatment: Thermal treatment (i.e., pyrolysis or calcination) removes and decomposes combustible electrolyte components by thermo-chemically degrading the organic compounds (e.g., separator, binder, and plastic case) which deactivates the battery. The processes are carried out in rotary furnaces, vacuum induction furnaces or blast furnaces.[311,314] The resulting decomposition products are separated in air and smoke cleaning systems.[71] To prevent a redox reaction of the metals, but enable an efficient decomposition of the organic compounds, an optimum temperature range between 500–600°C is stated.[310,312,319]

Dismantling and Sorting: Spent LIBs are delivered to recyclers either as complete batteries, battery packs sourced from, that is, electric vehicles, or as a bulk mixture of small size LIBs. Battery packs comprise large assemblies of battery modules and include many individual periphery components, such as housing, battery management system (BMS) or cooling parts. Therefore, they are first dismantled manually into modules or even cells.[309,312,314,318] This process is fraught with major safety risks due to the high voltage of the batteries (more than 450 V).[312] Thus, in industrial processes the aim is to use human-robot-cooperation, not only from a safety point of view, but also from an economic vantage point.[309,314,319,321] Concepts for a fully automated disassembly by industrial robots are controversially discussed as battery types are wide-ranging, complex and developing rapidly.[312,314,321,322] After battery pack and module dismantling, the LIBs are sorted by size, chemistry, and format in order to determine process routes for further treatment.[309,312,314] The batteries are further dismantled, manually with knives and saws, into individual cells, which in turn are further disassembled.[309,310,320] In the case of direct recycling, anodes, cathodes and separator are then separated and dried for 24 h at 60 °C in an oven.[310] It should be noted that these sorting and dismantling processes are only performed in hydrometallurgical, or direct recycling, and not in pyrometallurgical recycling. The subsequent separation of current collectors and active materials, which are adhered by a binder (normally PVDF or PTFE), poses a major challenge.[309,310]

Mechanical Treatment: Mechanical treatments allow the reduction of the material volume and the separation of the individual battery materials.[309,311,319] Several processes are available which can be used individually or in combination.

Crushing: In order to liberate the anode and cathode material, the LIBs must be crushed and ground using impact crushing, hammer crushing or a coarse roll crusher[309,320] which is typically done in a two-stage process.[314] First, the feed is pre-shredded and then the components themselves are liberated.[314] Thus, the powdered electrode materials fall out[311] and properties of the electrode components as well as particle size and distribution are changed.[309] If not done before, the spent LIBs can be deactivated during the comminution process.

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In that case, the comminution should be performed in a brine solution (wet crushing) or an inert gas atmosphere, so that safety precautions are maintained.[309,319]

Size Separation/Sieving: Sieving separates crushed battery component based on their different particle sizes. Through several sieving processes with varying size fractions, the materials can be further separated. A general distinction between the coarse fraction, including large particles such as plastic, electrode foil, separator, or steel casing, and the fine fraction, including smaller particles such as cathode and anode powders, can be made.[309,311,319,320]

Density Separation: The variation in density of the LIB components (e.g., copper electrode foil has a higher density than the aluminum foil) enables their separation by using drag forces and gravity.[312,320,323] For this purpose, a zig-zag separator, shaking table or vibrating screen with high air flow followed by a cyclone separator can be used.[320,323]

Magnetic Separation: An external magnetic field separates the components based on their ferro-, para-, and diamagnetic properties[311] and is mainly used to remove Fe-containing components.[312,320,323]

Mechanochemical Treatment: A chemical reaction initiated by mechanical energy is called a mechanochemical reaction. High-energy milling, such as planetary, ball or roller mills, is used to reduce the particle size of the battery parts and to achieve an increase in the specific surface area and a break-up of the structure. Breaking up the crystal structure of the cathode materials enables a simplified leaching process at room temperature. In addition, the reactivity with other materials is increased so that soluble compounds can be formed. Mechanochemistry is seen as an environmentally friendly recycling method for LIBs due to the low use of solvents.[309,319]

6.3.3. Pyrometallurgical Treatment

In pyrometallurgical treatment, battery components are smelted in a large furnace at high temperatures (about 1000 °C) to obtain a metal alloy consisting of the metals Cu, Ni, Co, and sometimes Fe, which is purified and separated by hydrometallurgy afterward.[309,311,318,319,323] Li and depending on the battery composition also Mn and Ti are usually not recovered as metals, but oxidize and form the slag, which can be used as an aggregate.[311,318,319,323] The electrolyte, binders, plastics, and carbon materials contained in the anode burn in an exothermic reaction, supplying energy to the process.[311,318,319,323] Despite sustainability concerns (such as the high energy requirements as well as the generation of toxic gases which, however, are filtered and cleaned), pyrometallurgical processes are one of the most commonly used recycling technologies for the valuable metals found in LIBs.[309,318,319,323] as the process underlies minor pretreatment and is robust (in respect of mixed and misdirected chemistries) and productive.[309,314]

6.3.4. Hydrometallurgical Treatment

In hydrometallurgical recycling, the valuable cathode materials are dissolved in acids and the individual metals are separated by solvent extraction.[314,319,323] Inorganic acids, such as H_2SO_4[309,310,318,319] HCl,[309,310,319] HNO_3[309,310,319] or H_3PO_4[310,319] are used to dissolve the metal components in a leaching process. Recently there are researches on environmentally friendlier organic acids and bio-organisms as leaching agents.[309,319] Subsequently, the metals are concentrated and purified by chemical precipitation, ion exchange or solvent extraction.[309,310,318] Additional purification processes such as extraction electrolysis or electrowinning are used if the resulting metal salts do not meet the quality requirements.[314] Compared to pyrometallurgical processes, hydrometallurgy is seen as more sustainable due to the higher recycling efficiency, reduced energy consumption and lower amount of emissions.[309,319,323] However, the hydrometallurgical technology involves complex processing steps,[309] necessitating a prior pretreatment process and a minimal amount of external material content.[319,323] Besides, there is a high consumption rate of chemical reagents in hydrometallurgical recycling processes.[310]

6.3.5. Direct Recycling

Instead of decomposing the active materials into substituent elements, direct recycling aims to recover anode and cathode materials as a whole for direct reuse in LIB production.[311,312,318,324] This approach is used particularly for battery scrap, which can be considered an important recycling source, from battery production facilities.[325] Current approaches are stated to be mechanical, electrochemical, cathode-to-cathode and cathode-healing technologies.[324] Direct recycling is seen as economically advantageous since complex purification processes and active material synthesis are avoided.[311,312,318] Thus, there are no strong acids used, which has a positive effect on the process environmental impact.[311] It is also claimed that direct recycling enables the recovery of all battery components.[318] Nevertheless, no standardized methods are currently established, due to the remaining challenges to recondition the materials at a suitable purity and quality level for battery reuse.[311] Besides, the recycling efficiency is highly depending on the health of the spent LIB.[318]

6.4. Recycling Activities in Europe

With regard to the rising amount of spent LIBs and the increasing need of efficient recycling technologies, the number of recycling facilities seems to be comparatively low. Currently there are 16 companies either already operating or in planning, as displayed in **Table 1**. Roughly, half of them are located in Germany. Only 6 out of 16 companies are operating on an industrial scale with an announced capacity of more than 1000 tons per year. Thereby the highest capacity, 10 000 tons per year, is provided by Redux Recycling GmbH (Germany), followed by Umicore (Belgium), with a capacity of 7000 tons per year. Even though Hydrovolt (Norway) states a capacity of more than 8000 tons per year, it is currently not in operation. In addition, VW Salzgitter (Germany) and BASF (Germany) each are currently constructing battery recycling facilities. Counting five companies, the combination of mechanical and

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Table 1. Li-ion battery recycling companies in Europe adapted and updated from refs. [311] and [326].

Company	Country	Scale	Process applied	Announced capacity tons/a	Source
Accurec	Germany	Industrial	Thermal + mechanical + pyrometallurgical + hydrometallurgical	3000	[311,326]
AEA Technology Batteries	United Kingdom		Dissolved in April, 2017		
AkkuSer Oy	Finland	Pilot	Mechanical	700	[311,326,327]
BASF[a)]	Germany	Pilot	Hydrometallurgical	N/A	[328]
Batrec Industrie AG	Switzerland	Pilot	Thermal + pyometallurgical	200	[329]
Duesenfeld GmbH	Germany	Pilot	Mechanical + hydrometallurgical	N/A	[311,326]
Euro Dieuze Industrie	France	Industrial	Mechanical + hydrometallurgical	6000 car batteries/a	[311,330]
Erlos	Germany	Pilot	Mechanical + direct	700	[311,326]
Hydrovolt[a)]	Norway	Industrial	N/A	More than 8000	[331]
Nickelhütte Aue GmbH	Germany	Industrial	Thermal + pyrometallurgical + hydrometallurgical	7000	[311,326]
Redux Recycling GmbH	Germany	Industrial	Thermal + mechanical	10 000	[311,326]
TES (formally Recupyl)	France	Pilot	Mechanical + hydrometallurgical	N/A	[311,326]
Primobius GmbH	Germany	Pilot	Mechanical + hydrometallurgical	1000	[332]
Promesa GmbH & Co. KG	Germany	Industrial	Mechanical	3200	[326,333]
SNAM	France	Pilot	Mechanical + pyrometallurgical + hydrometallurgical	300	[311,326]
Umicore	Belgium	Industrial	Pyrometallurgical + hydrometallurgical	7000	[311,326]
Ute Vilomara	Spain		Only spent LIB collection, no recycling		
VW Salzgitter[a)]	Germany	Pilot	Mechanical (+ hydrometallurgical[b)])	1500	[334]

N/A: insufficient available information; [a)]In planning; [b)]Performed by partner.

hydrometallurgical processes represents the most frequently used recycling method, followed by four companies applying a mixture of thermal and/or mechanical, pyrometallurgical, as well as hydrometallurgical processes. One company claims the use of hydrometallurgy only, two companies disclosed the use of mechanical treatment only and one facility proclaimed a combination of thermal and mechanical methods. Only one facility is applying thermal and pyrometallurgical treatment without hydrometallurgy and only one other company claims to use direct recycling processes, but only for cathode material. Overall, mechanical and hydrometallurgical processes seem to be the dominating approach for recycling facilities in Europe followed by a mixture of thermal, mechanical, pyrometallurgical, and hydrometallurgical processes.

6.5. Challenges and Outlook

A detailed analysis on challenges and potentials of LIB recycling can be found in the study of Harper et al.[318] providing a comparative overview of the advantages and disadvantages of the different processes, as well as in the SWOT analysis provided by Mohr et al.[311] Overall, the robustness and simplicity of pyrometallurgical recycling are offset by the high-energy usage and the loss of valuable materials.[309,311,314,318] Hydrometallurgy can provide high recycling rates with high purity, but uses potentially toxic solvents and complex processes.[309,311,318,319,323] Direct recycling enables the recovery of a wide range of battery components with low waste generation, but the technology is still very immature.[311,318] In addition, there is a trade-off situation as discussed by Weil et al.[335] cost-effective and efficient recycling versus quality and amount of recovered materials possibly hindering the realization of an ideal recycling depth.[335] A detailed elaboration of the challenges posed to LIB recycling, including their temporal dimension (short, medium, and long term), can be found in the BATTERY 2030+ Roadmap.[4]

7. Conclusion

After several hundred years of development, battery technology has become a key factor for large parts of modern industry.

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New and above all—large—applications that are fed by electrochemical storage systems are being considered. In order to keep pace with the accelerated introduction of battery electric vehicles, stationary storage systems and new mobile devices, it is necessary to establish new approaches for research and development in the battery sector. Not only is the number increasing, but so is the demand for better performance of storage devices. These should have more energy and performance, and be manufactured on a sustainable material basis. They should also be safer and more cost-effective and should already consider end-of-life aspects and recycling in the design. Therefore, it is necessary to accelerate the further development of new and improved battery chemistries and cells.

This can be achieved through an integrated, circular process that could replace the previously established Edisonian approach, in which each step of the discovery chain is sequentially dependent on the successful completion of the previous step(s). The European BATTERY2030 + initiative addresses these challenges with a chemically neutral approach that aims to reinvent the way batteries are invented. The approach is supported by five different research areas that are interconnected and in which the respective state of the art was collected and presented in this article.

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Conflict of Interest

The authors declare no conflict of interest.

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