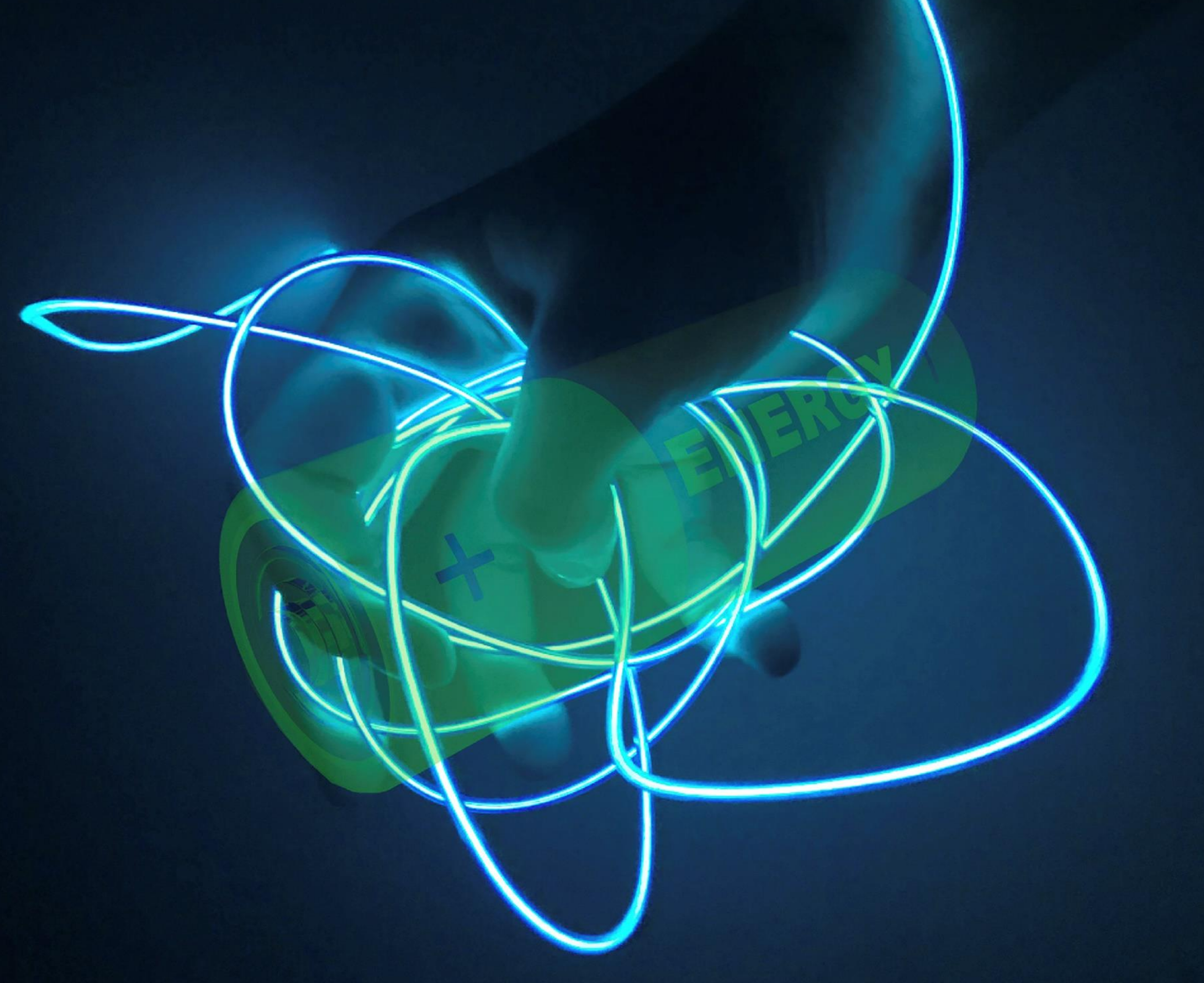
BATTERY 2030+



INVENTING THE SUSTAINABLE BATTERIES OF THE FUTURE

Research Needs and Future Actions

Updated August 2023

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PREFACE

Battery 2030+ is a large-scale cross-sectoral European research initiative bringing together the most important stakeholders in the field of battery R&D. The initiative fosters concrete actions to support the European Green Deal reaching a climate neutral society with a long-term vision of cutting-edge research reaching far beyond 2030.

In February 2020, the Battery 2030+ initiative published their first roadmap.[1] Two years later, in February 2022, the roadmap received its first update.[2] Since then, through its projects BIG-MAP, Bat4ever, Hidden, Instabat, Sensibat, Spartacus and the coordination and support action (CSA2), Battery 2030+ started to generate results following the visions and goals formulated in the roadmap. Due to the rapid pace of battery research in general and the most recent progress in the field, an update has been considered necessary.

This third version of the roadmap follows the main tracks from the earlier two versions while including updates on most recent developments in battery research, development and commercialization. It outlines the ambition to radically transform the way we discover, develop, design and manufacture battery materials, components, and cells for use in real applications. It remains our aim to make a collective European research effort towards ultra-high-performance, durable, safe, sustainable, and affordable batteries, and to support the urgent need for establishing European battery cell manufacturing. As the projects Bat4ever, Hidden, Instabat, Sensibat and Spartacus are reaching the end soon this roadmap also includes some pre-final results from each project. In May to September 2023 six new projects are joining the Battery 2030+ initiative, namely Healingbat, Opera, Opincharge, Phoenix, Salamander and Ultrabat. In 2024 projects related to both manufacturability and recyclability are joining.

In the process of formulating this roadmap, the stakeholders within the entire Battery 2030+ initiative have been engaged, comprising academia, RTOs and industry from 24 countries in Europe (including countries associated with the EU).

We are grateful to all the research and industry stakeholders who have actively taken part in shaping and improving this roadmap through their concrete and useful suggestions now incorporated into this document. Our roadmap will continue to be a living document that will be updated periodically as the research needs change and the battery field progresses.

August 2023

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1 Executive summary

Climate change is the biggest challenge our world faces today. Europe is committed to achieving a climate-neutral society by 2050, as stated in the European Green Deal[3] and reinforced with the green deal obligation “fit for 55 by 2050” regulation by the European commission.[4] This is also stated in the act REPowerEU.[5] The transition towards a climate-neutral Europe requires fundamental changes in the way we generate and use energy. If batteries can be made simultaneously more sustainable, safe, ultra-high performing, and affordable, they will be true enablers, “accelerating the shift towards sustainable and smart mobility; supplying clean, affordable and secure energy; and mobilising industry for a clean and circular economy” – all of which are important elements of the UN Sustainable Development Goals.[6]

In other words, batteries are a key technology for battling carbon dioxide emissions from the transport, power, and industry sectors. However, to reach our sustainability goals, batteries must exhibit ultra-high performance beyond their capabilities today. Ultra-high performance includes energy and power performance approaching theoretical limits, outstanding lifetime and reliability, and enhanced safety and environmental sustainability. Furthermore, to be commercially successful, these batteries must support scalability that enables cost-effective large-scale production.

Battery 2030+, is the large-scale, long-term European research initiative ***with the vision of inventing the sustainable batteries of the future, to enable Europe to reach the goals envisaged in the European Green Deal***. Battery 2030+ is at the heart of a green and connected society.

On the basis of our first roadmap in 2020, Battery 2030+ has started to create a vibrant battery research and development (R&D) community in Europe, focusing on long-term research that will continuously feed new knowledge and technologies throughout the value chain, resulting in new products and innovations. In addition, the initiative will attract talent from across Europe and contribute to ensuring access to competences needed for ongoing societal transformation.

The Battery 2030+ aims are:

- to invent ultra-high-performance batteries that are safe, affordable, and sustainable, with a long lifetime
- to provide new tools and breakthrough technologies to the European battery industry throughout the value chain
- to enable long-term European leadership in both existing markets (e.g., transport and stationary) and future emerging sectors (e.g., robotics, aerospace, medical devices, and Internet of things)

With this third version of the roadmap, Battery 2030+ has refined the originally expressed research directions, following actual developments[7–31], progress in the international research community as well as in the currently running ramp-up projects under the LC-BAT call within Horizon 2020 and the calls dedicated to Battery 2030+ within Horizon Europe. The chemistry-enabling approach of Battery 2030+ will allow Europe to reach or even surpass its ambitious battery performance targets set in the European Strategic Energy Technology Plan (SET Plan)[32], meet the "sustainability requirements for Batteries in the EU"[33] and foster innovation throughout the battery value chain. Battery 2030+ suggests three overarching themes encompassing six research areas needed to invent the sustainable batteries of the future. The three themes are: I) Accelerated discovery of battery interfaces and materials; II) Integration of smart functionalities; and III) Cross-cutting areas, all to enable the new chemistries of the future.

Theme I. Accelerated discovery of battery interfaces and materials is essential to secure new sustainable materials with high energy and/or power performance that exhibit high stability towards unwanted degradation reactions. Special attention must be paid to the complex reactions taking place at the many material interfaces within batteries.

Utilising the possibilities of artificial intelligence (AI), Battery 2030+ advocates the development of the Battery Interface Genome (BIG) – Materials Acceleration Platform (MAP) initiative to drastically accelerate the development of novel battery materials. A central aspect will be the development of a shared European data infrastructure capable of performing autonomous acquisition, handling, and use of data from all domains of the battery development cycle. Novel AI-based tools and physical models will utilise large amounts of acquired data, with a strong emphasis on battery materials, interfaces, and "interphases". Data will be generated for battery processes spanning multiple time and length scales using a wide range of complementary approaches, including computer simulations, autonomous high-throughput material synthesis and characterisation, in operando experiments and device-level testing. Novel AI-based tools and physics-aware models will utilise the data to "learn" the interplay between battery materials and interfaces, providing the foundation to improve future battery materials, interfaces, and cells.

Theme II. Integration of smart functionalities will enhance the lifetime and safety of batteries. Battery 2030+ suggests two different and complementary schemes to address these key challenges: the development of sensors probing chemical and electrochemical reactions directly at the battery cell level, and the use of self-healing functionalities to restore lost functionality within an operational battery cell.

New types of embedded sensors will allow the continuous monitoring of battery health and safety status. Sensor technologies and approaches that can be made suitable for monitoring reactions within a battery cell – for example, optical fibres, plasmonics, acoustics and electrochemical sensors – will realise more reliable battery systems. Such increased complexity inherently impacts manufacturability and recyclability, which must be considered early in the development cycle.

Self-healing batteries will utilise passive and active components in different parts of the battery cell that can be triggered by external stimuli or act continuously to prevent, retard, or reverse degradation and hazardous reactions within battery cells. Inspiration for this can be found in the area of drug delivery, underlining the need to work across research disciplines. When equipped with sensors, the battery cell could autonomously release the self-healing agents needed to control unwanted reactions and degradation phenomena, dramatically enhancing quality, reliability, lifetime, and safety.

New cost-effective sensors with high sensitivity and accuracy offer the possibility of "smart batteries". Battery 2030+ is targeting the integration of these new sensing technologies into the battery management system (BMS), to give a real-time active connection to the self-healing functions and a safer battery with a longer lifetime.

Theme III. Cross-cutting areas such as manufacturability and recyclability need to be addressed early in the discovery process. Can the new materials be upscaled in a sustainable way? Can we recycle the new cell concepts suggested in Theme II? Manufacturability is addressed from the perspective of the fourth industrial revolution, Industry 4.0.[34] Digital twins will be developed utilising the power of modelling and of AI to deliver solutions supported by advanced sensors, data infrastructure and communication protocols to replace classical trial-and-error approaches for manufacturing in speeding up this process. New recycling concepts, such as reconditioning active materials and electrodes, are central in this respect (see Figure 1).

Battery 2030+ is the large-scale collaborative multi-disciplinary research initiative for batteries that is necessary for Europe to stay at the forefront of global research. This initiative will allow European research institutions to supply new innovative knowledge and technology at the industrial level, and support battery cell development, production, recycling, and reuse. Over the coming decade, the strong Battery 2030+ research network will advance battery technologies far beyond the current state of the art.

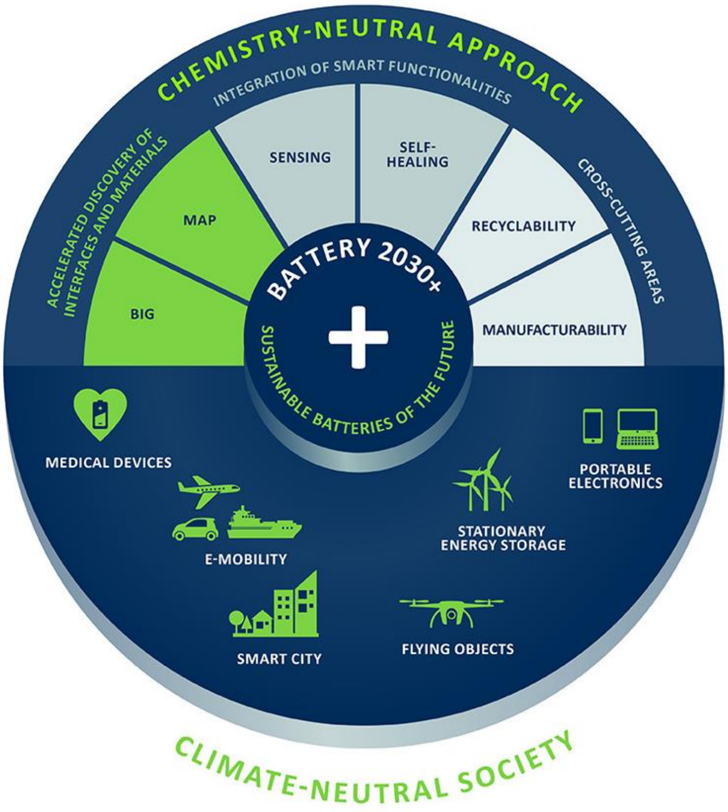


Figure 1. Battery 2030+: a holistic approach.

This roadmap is a living document and new research areas are to be expected as the Battery 2030+ initiative evolves with time.

2 Challenges

"Batteries are among the key technologies enabling a climate-neutral Europe by 2050"

Climate change, environmental pollution, habitat loss, and decreasing biodiversity have major impacts on our lives, economy, and society: We are facing global challenges that require coordinated actions. The EU-27's total carbon footprint in 2019 was equal to 6.7 tons of CO_2 per person, according to Eurostat.[35] By 2030, the EU wants to reduce its greenhouse gas emissions by 55% or more compared with 1990 levels, aiming at zero net emissions by 2050.[4] This goal has been formulated as part of the European Green Deal[3] launched in December 2019. The mission is to transform the EU's economy for a sustainable future, to make Europe the first climate-neutral continent by 2050 and to live up to the United Nations' Agenda 2030 and Sustainable Development Goals.[6]

In the initial roadmap for the European Green Deal, key policies, objectives and actions are formulated to reach the overall target. All EU actions and policies are to contribute to the objectives. The Battery 2030+ roadmap presented in this document supports this vision.

Rechargeable batteries with a very high round-trip efficiency are a key technology enabling energy storage for a vast number of applications, which is also expressed in the European Green Deal. Batteries can: accelerate the shift towards sustainable and smart mobility; help supply clean, affordable, and secure energy and mobilise industry for a cleaner, circular economy including full life cycle assessment (LCA).

Unsurprisingly, battery demand is rising dramatically.[36] All international institutions forecasting the future lithium-based battery market predict rapid growth over the next ten years. Europe alone will need an annual cell production capacity of at least 200 GWh in the next five years increasing steadily towards the TWh range for European companies (see Figure 2).

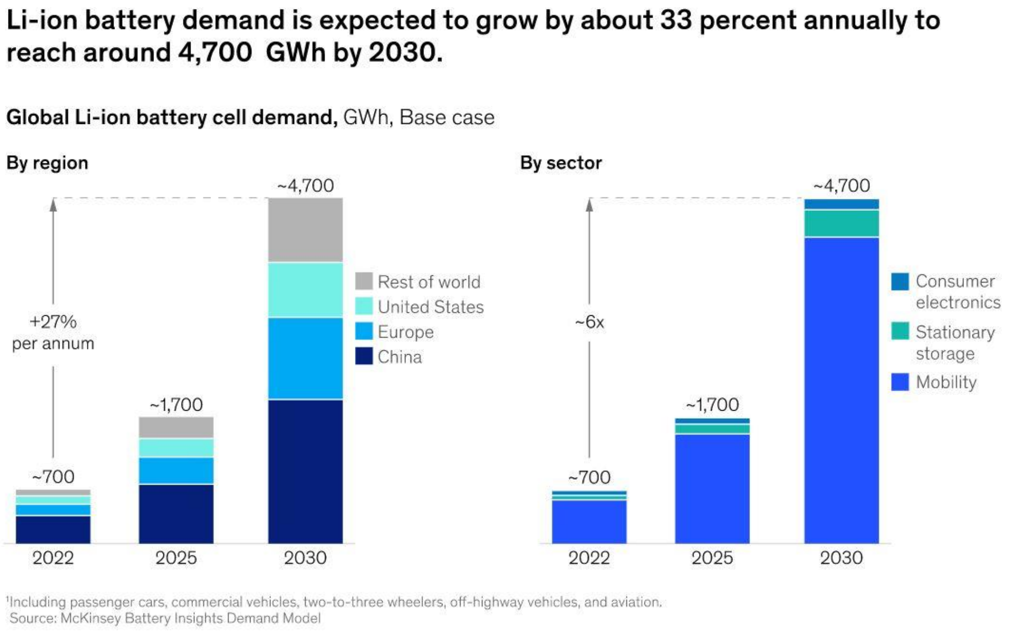


Figure 2. Expected growth in global battery demand by region (left) and sector (right).[37]

The market for high-energy-density rechargeable batteries is currently dominated by the lithium-ion (Li-ion) battery (LIB), which performs well in most applications. However, current generation LIBs are approaching their performance limits. Without major breakthroughs, battery performance and production will not keep up with the developments necessary to build the climate-neutral society.

While LIBs will continue to play a major role in the energy storage landscape, disruptive ideas are required that can enable the creation of the sustainable batteries of the future and lay the foundation for European competitiveness during the transition to a more electricity-based society.

Consequently, there is a need to create a dynamic ecosystem that dares to include long-term, transformational research starting at fundamental technology readiness levels (TRLs) that can rapidly feed new knowledge and concepts across all TRLs as well as into commercial products. To develop the necessary breakthrough technologies, immense multi-disciplinary and cross-sectorial research efforts are needed. Europe has the potential to take the lead thanks to both thriving research and innovation (R&I) communities covering the full range of involved disciplines and well-established innovation clusters with industry. However, to realise the vision of inventing the batteries of the future in Europe, we must join forces in a coordinated, collaborative approach that unites industry, researchers, policymakers, and the public in pursuing those goals.

In this context, European Commission Vice-President Maroš Šefčovič launched the European Battery Alliance (EBA) in October 2017[38] to support the battery industry in Europe throughout the value chain. Since the EBA launch, a European Strategic Action Plan on Batteries was published in March 2018, setting the direction for the development of a competitive battery industry in Europe.[39] The European Commission then set forth a state of play for the main actions to be implemented in the framework of the Strategic Action Plan, with Battery 2030+ being one initiative mentioned in the annex.[40]

One action in the Strategic Action Plan[39] calls for preparing an ambitious, large-scale, and long-term research programme on batteries as a complement to the more short- and medium-term actions of the EBA. The Battery 2030+ initiative is up to the task and hereby presents its vision for transformative battery research in the upcoming decade and beyond.

3 Vision and aims of Battery 2030+

Battery 2030+ is the large-scale, long-term European research initiative ***with the vision of inventing the sustainable batteries of the future, to enable Europe to reach the goals of a climate-neutral society***

For this vision to become a reality, Europe needs to re-emerge as a global leader in the field of batteries by accelerating the development of underlying strategic technologies and, in parallel, building a European battery cell manufacturing industry based on clean energy and circular economy approaches. Europe has the potential to take the lead by combining its strengths to ensure that we create a more coordinated and truly collaborative approach that unites industry, researchers, policy makers and the public in reaching these goals.

Battery 2030+ thus brings together the most important stakeholders in the field of battery R&D to work on concrete actions that support the implementation of the European Green Deal, the UN Sustainable Development Goals, as well as the European Strategic Action Plan on Batteries[39] and the SET Plan.[32]

The Battery 2030+ aims are:

- to invent ultra-high-performance batteries that are safe, affordable, and sustainable, with a long lifetime
- to provide new tools and breakthrough technologies to the European battery industry throughout the value chain
- to enable long-term European leadership in both existing markets (e.g., transport and stationary storage) and future emerging sectors (e.g., robotics, aerospace, medical devices, and Internet of things, etc.)

Based on a Europe-wide consultation process, the Battery 2030+ roadmap presents the actions needed to deliver on the overall objectives and address the key challenges in inventing the sustainable, safe, high-performance batteries of the future. Battery 2030+ suggests long-term research directions based on a chemistry-neutral approach focusing on the three main themes and six research areas outlined below.

4 Battery 2030+: A chemistry-neutral approach

Battery 2030+ follows a chemistry-neutral approach to facilitate the invention of the batteries of the future. Its goal is not to develop a specific battery chemistry, but to create a **generic toolbox for transforming the way we develop and design batteries**. Thanks to its chemistry-neutral approach, Battery 2030+ has an impact not only on current lithium-based battery chemistries, but also on all other types of batteries, including redox flow batteries and on still unknown future battery chemistries (see Figure 3). Battery 2030+ addresses key challenges such as achieving ultra-high battery performances, enhancing the lifetime and safety of battery cells and systems, and ensuring a circular economy approach (including the LCA approach) for the sustainable batteries of the future.

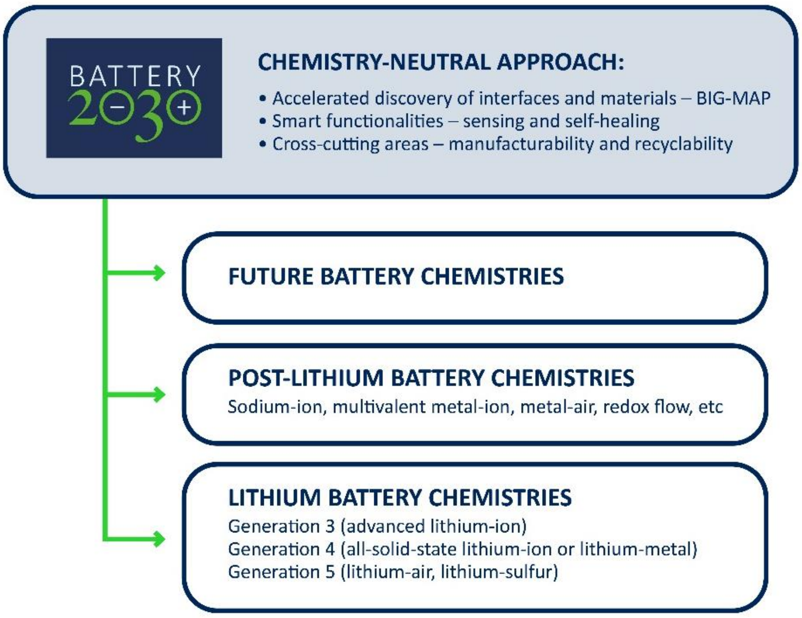


Figure 3. The Battery 2030+ chemistry-neutral approach will have an impact on both current state-of-the-art and future, as yet unknown battery technologies.

Battery 2030+ will join forces to focus on three overarching themes encompassing six research areas to address the key challenges in inventing the sustainable batteries of the future. These themes are summarized in the following and will be explained in detail in Section 7.

4.1 Theme I: Accelerated discovery of battery interfaces and materials

Creating autonomous, "self-driving" laboratories for the accelerated discovery and optimisation of battery materials, interfaces and cells

At the core of inventing the batteries of the future lies the discovery of high-performance materials and components that enable the creation of batteries with higher energy and power. Battery 2030+ advocates the development of a battery **Materials Acceleration Platform (MAP)**[12,41] to reinvent the way we perform battery materials research today. This will be achieved by creating autonomous, "self-driving" laboratories for the accelerated discovery and optimization of battery materials, interfaces and cells. This can be done by combining powerful approaches from high-throughput automated synthesis and characterisation, computational simulations of materials and interface, autonomous data analysis and data mining, as well as Artificial Intelligence (AI) and Machine Learning (ML).

Interfaces in batteries are arguably the least understood aspect of the battery, even though most of the critical battery reactions occur there, such as charge transfer reactions, dendrite formation, solid electrolyte interphase

(SEI) formation, and cathode–electrolyte interface (CEI) formation. Building on MAP, Battery 2030+ proposes to develop the **Batteries Interface Genome (BIG)**[9] that will establish a new basis for understanding the interfacial processes that govern the operation and functioning of every battery. The accelerated design of battery materials requires the detailed understanding, e.g., from characterisation, simulation and embedded sensors and tailoring of the mechanisms governing interface formation and evolution. This involves studying the mechanisms of ion transport through interfaces and, even more challenging, visualising the role of the electron in the interfacial reactions. These processes determine whether the ultra-high-performance batteries developed will be safe to operate and exhibit the long lifetimes that are necessary.

A central aspect will be the development of a shared European Battery Data space and data infrastructure capable of performing the autonomous acquisition, handling, and analysis of data from all domains of the battery discovery and development cycle. Novel AI-based tools, physical and hybrid physical-ML models will utilise the large amounts of data gathered, with a strong emphasis on battery materials and interfaces. The data generated across different length and time scales, using a wide range of complementary approaches, including numerical simulation, autonomous high-throughput material synthesis and characterisation, in-operando experiments, and device-level testing, will all contribute to new material and battery cell development.

Integrating these two research areas, BIG and MAP (**BIG–MAP**) will transform the way we understand and discover new battery materials and interfaces. Theme I will deliver a transformative increase in the pace of new discoveries for engineering and developing safer, longer-lived, and sustainable ultra-high-performance batteries. In a further step the sensor data of the smart batteries of the future can also be integrated into the BIG-MAP approach. This gives a link to theme II.

4.2 Theme II: Integration of smart functionalities

Increasing safety, reliability, and cycle life of batteries
by introducing smart sensing and self-healing functionalities

Even the best battery will eventually fail, which is why methods must be developed that increase safety, reliability, and cycle life of batteries by introducing smart sensing and self-healing functionalities. Degenerative processes within a battery cannot be suppressed completely, and external factors such as extreme temperatures, mechanical stress, excessive power during operation, or simply ageing will, given time, act detrimentally on battery performance. From the perspectives of sustainability, economic efficiency, and reliability, new ways need to be found to increase safety and lifetime particularly in critical applications.

The Battery 2030+ vision is to incorporate smart **sensing** and **self-healing** functionalities into battery cells with the goals of increasing battery reliability, enhancing lifetime, improving safety, lowering the cost per kWh stored, and, finally, significantly reducing the environmental footprint.

Non-invasive sensing technologies offering both spatial and time resolution will be developed to monitor key battery cell parameters during operation and to determine defective areas or components within the cells that need to be repaired by activating/adding self-healing functions. In the battery of the future, sensors will make it possible to follow chemical and electrochemical reactions "in vivo" directly inside a battery cell during real-world operation. New sensor technologies will emerge that can diagnose the early stages of battery failure, thermal runaway, and unwanted side reactions leading to early battery ageing.

Self-healing functionalities will become an important property of future batteries in applications that require them with high reliability, high quality, and long lifetimes. Combining sensing and self-healing functionalities will result in batteries with a predictable lifetime and documented State of Health (SoH), state of safety, and usage history. Smart functionalities will enable better acceptance of used cells in primary and secondary applications.

With its two research areas, Theme II will address the need for safe and long-lived batteries.

4.3 Theme III: Cross-cutting areas

Making manufacturability and recyclability integral parts of battery R&D at an early stage

The battery of the future will be designed based on virtual representation taking into account sustainability and circular economy concepts including life cycle assessment (LCA).[42] As a consequence, considerations regarding manufacturability and recyclability are integral parts of battery R&D and must be considered at an early stage. Materials sourcing, processing, manufacturing and assembly processes must be tailored to accommodate new chemistries and follow innovative approaches to allow for efficient remanufacturing and re-use requirements.

The manufacturability and **recyclability** of batteries are thus key cross-cutting areas that will develop through close collaboration between those addressing themes I and II. From the outset, new knowledge and ideas about how to manufacture and recycle batteries will inform the materials discovery and development processes.

The manufacturing of current and future battery technologies is addressed in this roadmap from the standpoint of the fourth industrial revolution, i.e., Industry 4.0[34] and digitalisation. Developing digital tools for predicting the impact of manufacturing parameters on the characteristics and performance of the final cell is a highly valuable approach that reduces the reliance on costly and time-consuming trial and error methods. Thus, developing effective digital tools for manufacturing predictions will require accurate and validated models, with efficient and reliable parametrization methods, access to in-line manufacturing parameters and expertise in simulation and data analytics techniques. As technology advances, these tools will become increasingly sophisticated and accessible, empowering cell manufacturers and designers to make informed decisions, optimize processes, and ultimately improve cell outcomes.

The new materials and cell architectures envisioned in Battery 2030+ call for new recycling concepts, such as reconditioning or reusing active materials and electrodes. To pave the way for such a shift, material suppliers, cell and battery manufacturers, main application actors, and recyclers will be directly coupled to accommodate the constraints of recycling when developing new batteries. The discovery of new materials using BIG–MAP will integrate parameters such as recyclability, critical raw materials, and toxicity into the algorithms.

With these two research areas, Theme III will ensure that all research approaches will consider the feasibility of scaling up new materials and battery cells as well as the possibility of recycling and reusing battery components at low cost and using climate-neutral approaches.

4.4 Battery 2030+: A holistic approach

4.4.1 The six research areas of Battery 2030+

BIG, MAP, Sensing, Self-healing, Manufacturability, and Recyclability are the six research areas that Battery 2030+ advocates as having major impacts on inventing the battery of the future. All these areas are interlinked, contributing new tools that will transform the way Europe discovers and develops batteries. Across these research areas, the **safety** and **sustainability** of newly developed battery technologies will be central guiding principles. The progress in all identified research areas will be essential for inventing batteries with properties that are tailor-made for their specific applications (see Figure 4).

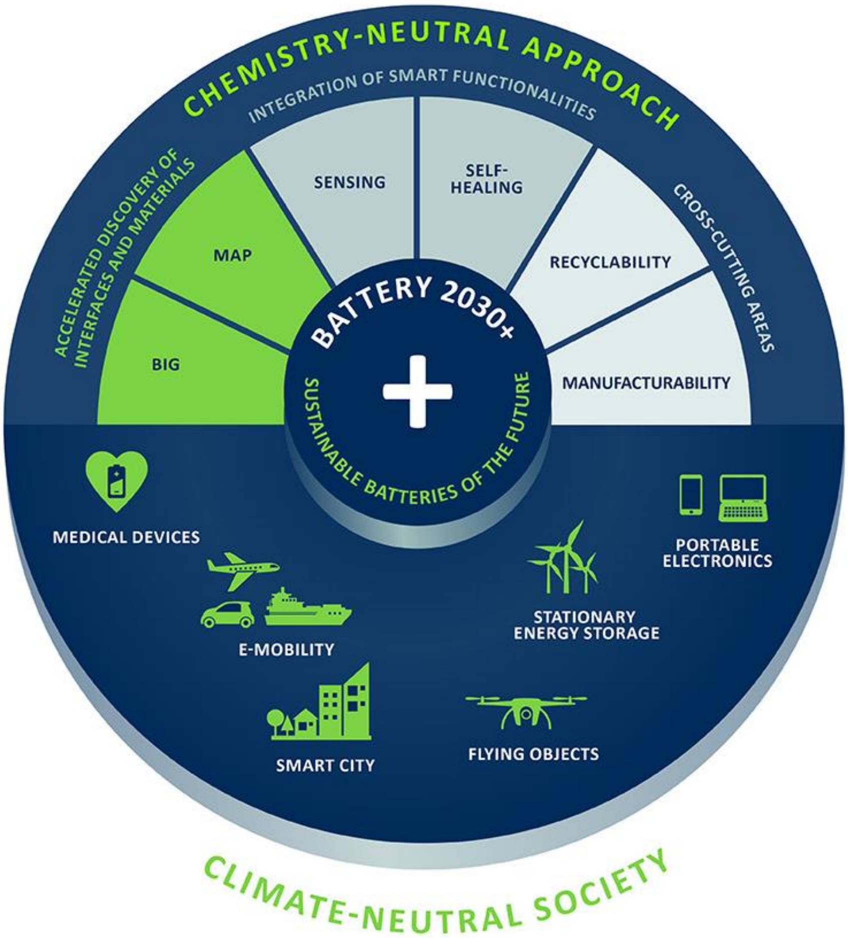


Figure 4. The Battery 2030+ vision is to invent the sustainable batteries of the future through a chemistry-neutral approach that will deliver ultra-high-performance batteries optimised for their intended applications, such as electro-mobility, stationary storage, medical devices, and robotics. Battery 2030+ focuses on three main themes and six research areas that are strongly linked, all contributing new tools for accelerating battery discovery and development.

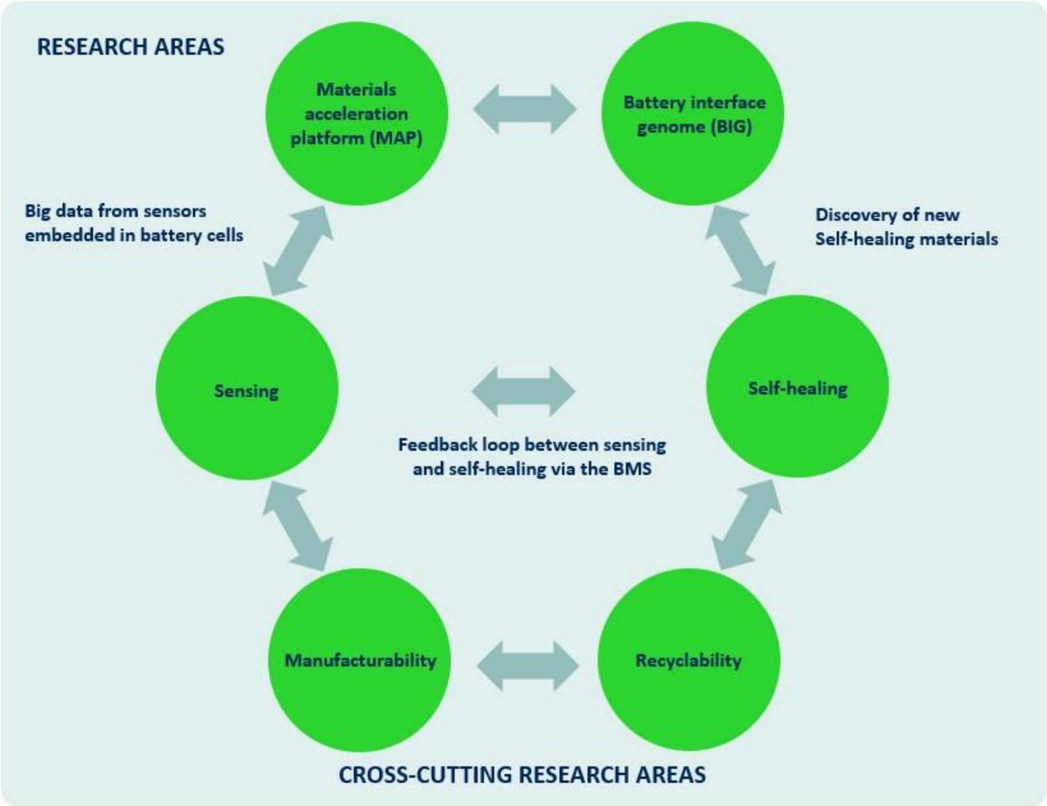


Figure 5. Interactions between the different Battery 2030+ research areas.

Some of the links between research areas are summarised in Figure 5, such as:

- The Materials Acceleration Platform (MAP) and the Battery Interface Genome (BIG) will be powerful tools for discovering new materials and engineering battery interfaces, and in particular will be used to discover or optimise self-healing materials and chemicals.
- Sensors integrated at the battery cell level will provide a huge amount of data for the research community, data that will be systematically exploited by feeding the AI used in MAP.
- Sensing and self-healing functionalities will be strongly connected via the battery management system (BMS), which will trigger self-healing based on information from the sensors.
- Finally, the development performed in the cross-cutting research areas (i.e., manufacturability and recyclability) will ensure that manufacturing and recycling processes become more efficient and sustainable based on current technologies. Building on this, the next step will be to develop new and more advanced manufacturing processes that will enable the production of new materials, technical interfaces, sensors and self-healing functions emerging from the other research areas.

For each research area, short-, medium-, and long-term goals have been identified and are presented in Table 1. With the goal of a closed-loop between the research areas resulting from points of contact and synergies between them, specific goals and more details on their cross-links will be presented in section 8.

Table 1. Short-, medium-, and long-term goals for BIG–MAP, Sensing, Self-healing, Manufacturability, and Recyclability.

Research areas	Short term (3 years)	Medium term (6 years)	Long term (10 years)
BIG-MAP	Put in place a pan-European interoperable data infrastructure and user interface for battery materials and interfaces.	Fully implementing BIG in MAP to integrate computational modelling, autonomous synthesis and characterisation of materials.	Demonstrate the integration of manufacturability and recyclability parameters into the materials discovery process.
	Establishing integrated experimental and computational workflows.	Integrate data from embedded sensors into the discovery and prediction process.	Integrate battery cell assembly and device-level testing into BIG-MAP.
	Demonstrating BIG-based hybrid physics- and data-driven models of battery materials.	Develop and apply predictive hybrid models for the spatio–temporal evolution of battery interfaces/interphases to perform inverse materials design.	Implement and validate digital twin for ultra-high-throughput testing on the cell level.
	Deploy autonomous modules and apps for on-the-fly analysis of data characterisation and testing using AI and simulations.	Demonstrating transferability of the BIG-MAP approach to novel battery chemistries and interfaces.	Establish and demonstrate full autonomy and chemistry neutrality in the BIG MAP.
	Developing multi-modal high-throughput/high-fidelity interface characterisation approaches.	Integrating novel experimental and computational techniques targeting the time and length scales of electron localization, mobility, and transfer reactions.	Demonstrate a 5–10-fold improvement in the materials discovery cycle and interface performance.
Sensing	Miniaturise and integrate the identified (electro)chemically stable sensing technologies with multifunctions at the cell level and in real battery modules, in a cost-effective way compatible with industrial manufacturing processes.	Development of virtual sensors to limiting the number of physical sensors to a minimum	Master sensor communication with an advanced BMS relying on new AI protocols by wireless means to achieve a fully operational smart battery pack.
		Deliver proof of concept of higher quality, reliability, and lifetime on the cell and module levels.	
	Deploy sensors capable of detecting various relevant phenomena (e.g., interface dynamics, electrolyte degradation, dendritic growth, metals dissolution, and materials structure change).	Adaptation of the reliability of the sensor integration	
		Address challenges on integration, measurement and compatibility of sensors related to new cell technologies (e.g., all solid-state batteries)	
Self-healing	Continue developing the research community that includes a wide range of R&D disciplines to develop self-healing functionalities for batteries.	Integration of self-healing functionalities into battery components. Biomimetic membranes developed as a new functionality.	Upscaling of the manufacturing of self-healing batteries is needed in long term, including a cost-benefit estimation.
	Developing autonomous and non-autonomous (on demand) self-healing functionalities for specific battery chemistries, targeting loss of capacity and loss of power.	Feedback loops between cell sensing, BMS, and/or AI modules.	Different cell design concepts and novel designs, including e.g. bi-polar systems, are needed. Manufacturing lines to be adapted on the designs needs.
Manufacturability	Proof-of-concept (POC) of a digital twin of cell design based on accurate multi-physics multi-scale models and AI data-driven models for LIBs.	Initial POC of a digital twin of cell manufacturing process for LIBs at pilot line level by integrating data-driven aspects (data acquisition, sensorization, communication and interoperability) into the developed models.	Full POC of a manufacturing digital twin for LIBs by integrating the cell design and the manufacturing process sub-loops.
	Improvements towards new greener and more sustainable manufacturing processes for LIBs (3D printing, dry processing) are foreseen.	Developing a methodology that will be adapted to the manufacture process for new battery technologies (SSBs, SIBs, etc.).	POC of a digital twin of novel cell manufacturing routes with closed-loop recycling of optimized LIBs.
	Up-scaling of process models along the LIB cell manufacturing to machine models for optimal designs through pilot validation.	Development of advanced in-line sensors for implementing in manufacturing plants	The new concepts in cell manufacturing are transferred to the industry and academia.
	Improve methodology for scaling up process (from lab to pilot and further)		
	Accellerate and efficient parametrization methods		
Recyclability	Integrated design for sustainability and dismantling.	Demonstrating automated cell disassembly into individual components.	A full system for direct recycling is developed and qualified.
	Demonstration of new technologies for battery packs/modules sorting and re-use/re-purposing.	Sorting and recovery technologies for powders and components and their reconditioning to new active battery-grade materials demonstrated.	Combination of direct recycling with other secondary processes in order to identify optimised solutions aiming at this targets (combination of direct recycling and secondary processes enabling to achieve 98 % recovery)
	Establishing a European system for data collection and analysis.	Significantly improve, relative to current processes, the recovery rate of critical raw materials	
	Developing automated disassembly of battery cells.	Testing of recovered materials in battery applications.	
	By design develop sustainable batteries: integrated design for optimising CRM content, lifetime, sorting, re-purpusing, dismantling, recycling	Develop prediction and modelling tools for the reuse of materials in secondary applications	
	Address how direct recycling can be handled in a chemsitry-neutral way (mix of technologies) by making recyclability an intergral part of battery R&D at an early stage	Integrate battery passport	

4.4.2 Ontologies and standards as tools for collaboration and innovation

For Battery 2030+ being able to achieve the ambitious goals laid out in this roadmap, research within the initiative – and beyond – must meet the highest standards in terms of data generation, data processing, data storage, data exchange and metadata treatment. It is therefore one of the goals of the initiative to help the battery research community develop powerful research data management (RDM) strategies and tools as well as consensus-based standards and guidelines for experimental and theoretical research on batteries.

Combined, RDM tools and standardisation will not only improve the general quality of research within Battery 2030+ and enable the FAIR[4] (Findable, Accessible, Interoperable and Reusable, Reproducible, Reliable, Relevant)[43] data principles. More importantly, collaboration will be possible on entirely new levels, allowing for novel, autonomous research approaches, accelerated materials discovery, and data-based research in a field that has thus far mostly adhered to classical trial and error research. Developing protocols and standards will play a key role in connecting the six different research areas of Battery 2030+, and it will also enable collaboration with partners outside of Battery 2030+.

The implementation of protocols and standards in battery research, characterisation, development, and production needs to proceed along the complete research and development chain, from materials synthesis at universities to cell production in pilot facilities, from basic theoretical research to electrochemical testing of full cells. It is envisioned to follow a step-by-step process that is specifically adapted to the needs of the different research areas where it is deployed, but that follows a proven scheme, and that includes both partners within Battery 2030+ as well as outside the initiative, such as Batteries Europe, relevant EU projects (e.g., LiPlanet), regulatory bodies, and formal standardization bodies (e.g., CEN and CENELEC on the European level). Such a step-by-step process starts with the identification of important R&D areas where standards and protocols will have the highest impact and ends with the publication of (authoritative) standard documents and guidelines.

A consistent ontology creates clear definitions of the vocabulary, data inputs and outputs, relations and processes in battery research and development. It is the basis of a harmonized approach for generating and processing data and for creating a common data sharing infrastructure.

According to EN 45020, standards are “documents, established by consensus and approved by a recognized body, that provide, for common and repeated use, rules, guidelines or characteristics for activities or their results, aimed at the achievement of the optimum degree of order in a given context”. In practice, standards can accelerate battery research and innovation in several ways. They can create “order” on various different levels ranging from general agreements on ontologies and data interfaces (metadata), over agreements on the order of process steps, e.g., in a coin cell assembly process, down to details about how certain individual measurands (key control characteristics or key performance indicators) should be determined by specifying measurement methods and protocols.

The development of ontologies and standards is driven by demand: Both the research community as well as industry must benefit from the activities and must therefore lead the way in identifying areas where standards could have a positive impact on collaboration and innovation activities.

The concrete role ontologies and standards will play in the different research areas will be addressed in the more detailed, research area-specific Sections 7.1 to 7.6 of this roadmap.

5 Impact of Battery 2030+

By following a coordinated, multidisciplinary, and harmonised, European approach, **Battery 2030+ will have major impacts on the battery technology ecosystem and beyond.**

5.1 Impact of a large-scale battery research initiative

Battery 2030+ aims to invent the sustainable batteries of the future. More specifically, it will lay the scientific and technological foundation and provide the necessary tools to enable the next generation of high-performance, safe, and sustainable batteries in Europe. Having these novel battery technologies at our disposal will have societal and environmental impacts on many levels. It will increase energy security, reduce the environmental footprint in many application areas, and help forge a climate-neutral society while creating new markets and jobs.

The collaborative approach of Battery 2030+ creates strong **synergies** for Europe. While open scientific competition is certainly integral to any research that strives for new discoveries, an integrated large-scale approach will put our limited R&D resources to their best use and accelerate new innovations.

A large-scale initiative is needed not only to gather appropriate resources but also to attract the **talent and competences** necessary to achieve the scientific-technical goals and to support European industry with a skilled workforce. Educational and outreach programmes will enrich the European battery community, make Europe a world-leading and dynamic repository of battery knowledge, and help create and maintain the necessary critical mass of motivated researchers who will strive to realise our common vision.

This perspective on a sustainable build-up of knowledge and competences to meet current and future challenges is also strongly reflected in the research visions and plans of Battery 2030+: to meet the need to create a dynamic ecosystem that dares to include long-term, transformational research starting at fundamental technology readiness levels (TRLs) that can rapidly feed new knowledge and concepts across all TRLs as well as into commercial products.

A consolidated and **coordinated exploitation plan** will bring the new fundamental concepts and ideas of Europe's battery community to the market more efficiently. This will be possible by interacting with and supporting other European initiatives, industry stakeholders, and networks that either are part of or associated with Battery 2030+, or that will be engaged early on.

An overview of the current R&I landscape in Europe is given in Figure 6, illustrating how Battery 2030+ is positioned among other important European initiatives. Batteries Europe, the new European Technology and Innovation Platform on Batteries (ETIP) pursues a timely implementation of low-carbon energy technologies to answer research requirements across the entire battery value chain to accelerate the establishment of a globally competitive European battery industry. While Batteries Europe focusses on the short to medium term, Battery 2030+ aims at long term innovation by reinventing the way to invent future batteries. On the way there, collaboration and information exchange with Batteries Europe will be one major part towards a sustainable and competitive value chain in Europe. In that respect, Batteries Europe has already published six roadmap papers in 2021 on “New and emerging technologies”, “Raw materials and recycling”, “Advanced materials”, “Cell design and manufacturing”, “Mobile applications of batteries” and on “Stationary applications for batteries”.[44–49] Along with their document on “Development of reporting methodologies”[50] they supplement the European Battery R&I landscape and the vision of Battery 2030+ by important strategic focus areas.[51]

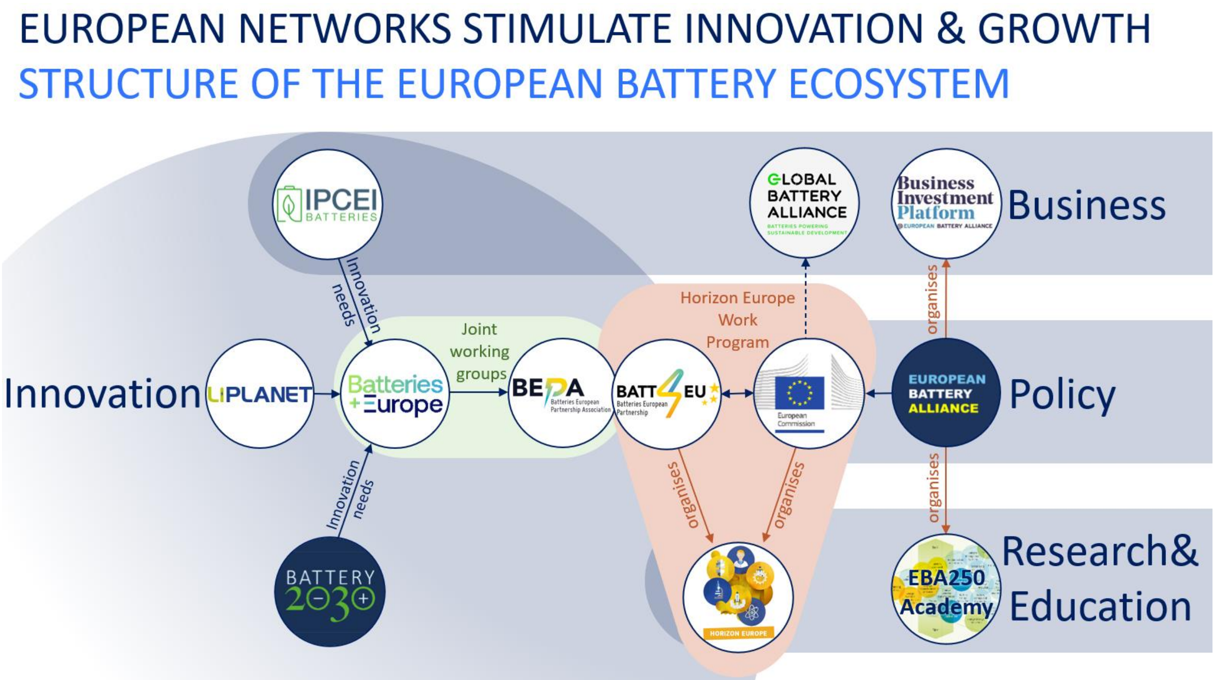


Figure 6. Structure of the European battery ecosystem. Graphic adapted from VDI/VDE-IT.

5.2 Impact along the battery value chain

The Battery 2030+ community will actively address the impact of scaling on energy density, i.e., the reduction in weight- and volume-specific metrics when scaling from the materials level to the battery pack level. The Battery 2030+ themes will also address the unwanted chemical and electrochemical side reactions that reduce battery capacity with time.

Figure 7 schematically illustrates how the different components of a battery affect its overall performance. The active battery material can store a certain amount of energy per weight or volume (specific energy, energy density, 100 %). As the different components of a real battery are added – for example, binders, conductive fillers, and other additives within the electrodes; current collectors, separators, electrolyte, packaging, wiring, cooling, and battery controller – the energy content per weight and volume drops, as from the storage capacity point of view a considerable quantity of “dead mass” is added. Finally, the specific energy decreases during use towards the end of life, which is defined differently for different applications.

To obtain a high-performance battery, scientists and engineers need to pursue a concerted approach. While the engineers have already made progress in providing more space for the active material in the battery pack, thus enabling longer driving ranges, the materials need to be further developed and improved, as well. Only these combined efforts can eventually reach satisfactory performance of a battery. A validated approach is to start with materials having high specific energy, and to minimise losses along the manufacturing chain and during use. For novel and future battery chemistries, this is a challenge, as: (a) high-performance materials are still lacking; (b) digital tools to efficiently manufacture new cells, to gain improved process understanding, and to accelerate development while exploring new manufacturing routes have not been developed; and (c) performance degradation remains an issue. The themes and research areas of Battery 2030+ will address these issues as shown in Figure 7.

Along the complete battery value chain, the battery community will benefit from the development of ontologies, standards and protocols: If developed in close coordination with relevant European partners outside Battery 2030+, these will enable data-driven battery research, accelerated materials discovery and new ways of collaboration and pave the way to a connected battery research community. Standards can additionally play an important role in ensuring the sustainability goals of the initiative, as guidelines and standards are sorely needed for certification of green battery production processes.

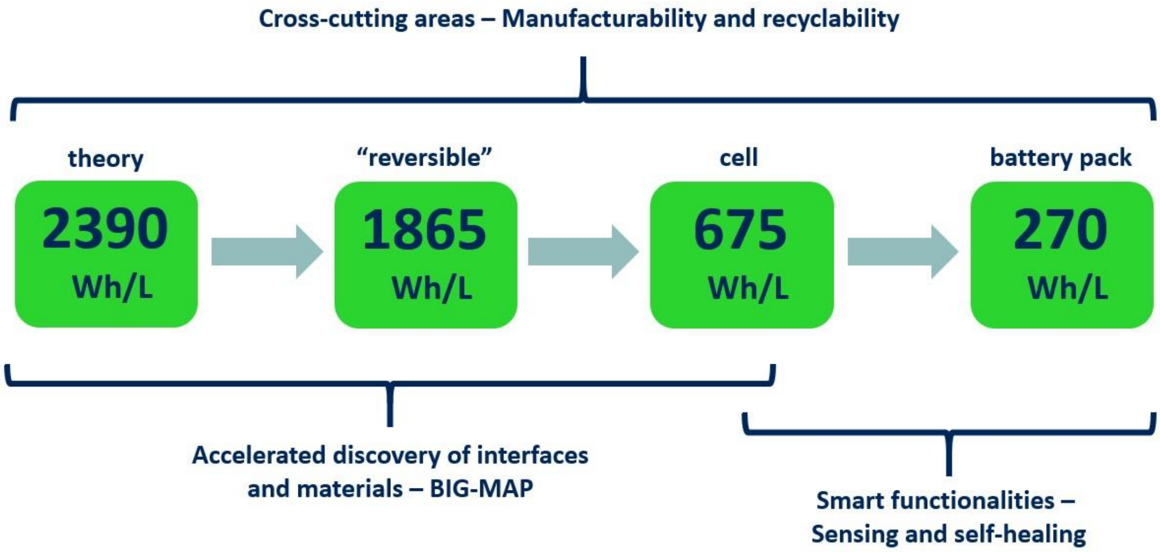


Figure 7. The decrease in total capacity as more inactive material is added when going from the material to the complete battery pack. The identified research areas will address these losses throughout the battery value chain. The additional capacity loss due to degradation. Representative example capacity based on graphite-SiO_x, NCA cylindrical cells adapted from M. J. Lacey presentation at the 3rd Battery 2030+ annual conference in Uppsala.

5.3 Impact on the European SET Plan targets for batteries

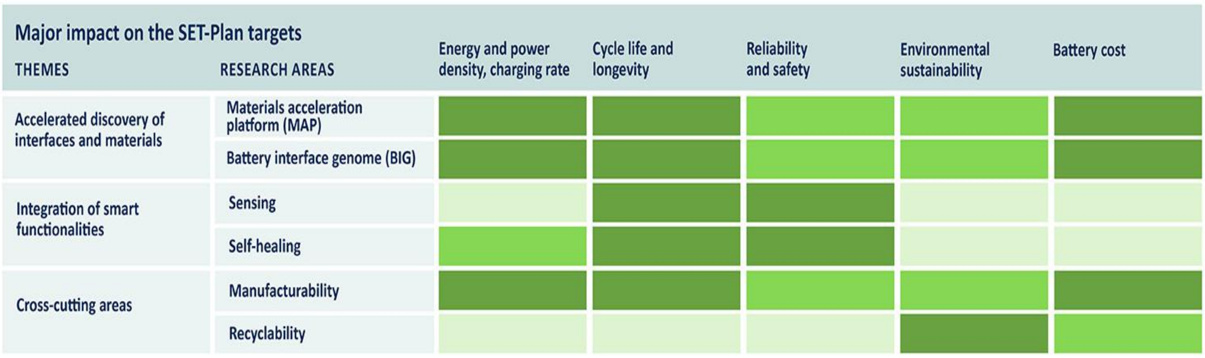
Battery 2030+ suggests actions pushing battery technologies far beyond the current state of the art. This will have an impact throughout the battery value chain by enabling and accelerating the attainment and surpassing of the SET Plan targets.

The integrated SET Plan Action 7[32] highlights the large impacts of batteries on European society “from education to economics, from knowledge to environment and from business to resource security”. The plan states that Europe has a strong R&I base in, for example, materials but that this sector is highly competitive and there is a need for “augmented R&I to keep up with the pace of battery development and uptake around the world”. The working group requested a challenge-based holistic approach asking: “What can we achieve together? Which challenges can we not solve alone?”

The SET Plan action 7 concentrates mainly on the transport sector, while the Battery 2030+ initiative also addresses the great need for efficient and sustainable batteries in other areas. Our approach with three themes and six research areas will have a positive impact on the development of batteries for a wide range of applications, including transport electrification, stationary storage enabling renewable energy use in the electricity grid, and new emerging possibilities and applications. The new knowledge generated will also be transferred to new educational curricula at various levels.

In Action 7 of the SET Plan, key performance indicators (KPIs) are continuously updated to guide European battery developments. The Battery 2030+ research areas will have an impact on all these KPIs and will ensure that Europe can reach (or even surpass) the SET Plan targets at an accelerated pace (see Table 2).

Table 2. The major impacts Battery 2030+ research areas will have on the SET Plan targets. Dark green = high impact, lighter green = medium to lower impact.



6 Current state of the art and Battery 2030+ in an international context

The global competition of the battery market is dominated by Asia and specifically China. Today China also dominates the research efforts, being the country with the largest volumes of published papers in peer-reviewed international journals. The research efforts are also strong in the US and Europe is not far behind the US.[52]

Research is conducted on all kinds of batteries at all TRLs and along the full battery value chain. The main focus today is on the generations of LIBs, but future battery chemistries are coming stronger and stronger.

All global efforts have made several countries and regions inspired to make their own strategic plans for batteries and energy storage. In these plans, that primarily deals with how to strengthen the market and companies, education and skill development as well as research are highlighted.

The battery roadmaps that exist are mainly focused on the timeline for new generations of battery chemistries: when they will be available on the market and what capacities they could reach. Many of these roadmaps also express the short, medium and long-term expectations. Various associations and countries have published roadmaps for batteries or strategies for energy storage, including batteries. Some recent roadmaps are from: ETIP,[44–49,53] EASE,[54] EMIRI[55], EUCAR,[56] implementation of the SET Plan Action 7,[32,40] JRC,[57–60] China,[61] Austria,[62] Finland,[63,64] France,[65] Germany,[66] Hungary,[67] Italy,[68] Lithuania,[69] Netherlands,[70] Norway,[71] Portugal,[72] Spain,[73] Sweden,[74] India,[75,76] Japan,[77,78] and the USA.[79] Many of these have been formulated since the last update of this roadmap.

This Battery 2030+ roadmap with the different approach described above has the goal to accelerate research to faster reach the long-term goals. This approach is globally quite unique, but now several national battery programs in Europe with a long-term research focus have adopted elements of this roadmap: France, the Netherlands and Sweden are some examples.

The state of the art of the research that Battery 2030+ will move ahead on is related to the current market and how the batteries of the future can be developed.

Today's rechargeable battery market is led by lead acid (49.9%) and lithium-ion batteries (LIBs) (45.7%).[80] Nickel-cadmium, nickel-metal hydride, and non-rechargeable chemistries also matter commercially. Redox flow batteries (RFBs) are being developed for stationary energy storage. Emerging battery technologies using novel chemistries require intensified research. Sodium-ion batteries and Li-metal systems are advancing, while sustainable materials-based batteries (e.g., potassium, magnesium, aluminium, calcium) need substantial R&D.

LIBs find use in electronics, EVs, energy storage, and grid backup due to superior energy density, efficiency, and reliability. Their emergence in the last decade is due to their superior energy density compared to lead-acid batteries (see Figure 8). Commercial LIBs came to market in the 1990s, with energy density doubling[81] and costs dropping by about 15-fold from 1995 to 2019.[82–84] Global efforts aim to enhance performance through improved materials, electrolytes, design, and production methods.

Na-ion batteries move from labs to applications like stationary storage and car starters, competing with lead-acid batteries. Enhancing electrolytes and electrodes holds potential for future Na-ion applications.

RFBs aim at large-scale storage with benefits like scalability and recyclability. Presently, vanadium and zinc-bromine RFBs dominate, but research seeks ubiquitous materials and non-corrosive electrolytes for cost-effective storage, supporting more independent renewable energy deployment in Europe.

In line with its chemically neutral approach, Battery 2030+ will be open to chemistry and materials developments and discoveries, regardless of the technology; this will facilitate Europe’s technological independence to develop a battery ecosystem. Technological sovereignty would facilitate the electrification of the economy, thus contributing to combat climate change in line with the Green Deal and Fit for 55 Package, among others.

The status of current commercial batteries and selected future chemistries is summarised in Figure 8, which depicts the **energy performance** characteristics of the major rechargeable battery types. The figure does not take power into account. More details of the state of the art can be found in several reference sources.[85–88]

A number of battery properties, including safety, cost, lifetime, energy, and power, need to be improved to produce the batteries of the future.

Safety and safety hazards are regulated in the Battery Directive 2006/66/EC in the upcoming Eco-design Directive for Batteries with an update concerning batteries and waste batteries in the amending regulations 2019/1020 and COM/2020/798. In its roadmap, the European Council for Automotive R&D EUCAR[56] set safety levels for battery cells and battery packs as guidelines for judging battery quality.

The **cost** of batteries is of course highly relevant. Today’s price for state-of-the-art LIB packs is roughly USD 150–120/kWh.[89] The expected cost will decrease to well below USD 100/kWh by 2024,[89] a cost level that all future batteries must reach to be competitive. In Battery 2030+, the cost of materials and battery cell production must be considered in order to deliver the right solutions for the future.

The **lifetime** of a LIBs is limited and must be at least doubled by 2030. Battery 2030+ focuses on the possibility of increasing the “first life cycle” of the battery, while battery “second life or second use” will be addressed through actions at lower TRLs.

Power is an important parameter. A high-power capacity is necessary, for example, to charge a vehicle rapidly. The limitation today is the transport of ions through interfaces within the battery cells, which means that new cell designs and materials need to be discovered.

We are now entering a phase in which the energy performance at cell level is levelling off for LIBs. Therefore, new concepts in LIBs aim to improve battery level performance (e.g., cell-to-chassis) by reducing the need for passive components in the battery architecture. However, it will nonetheless be difficult or even impossible to satisfy future requirements for electrochemical energy storage using only solutions based on currently commercialised technologies.

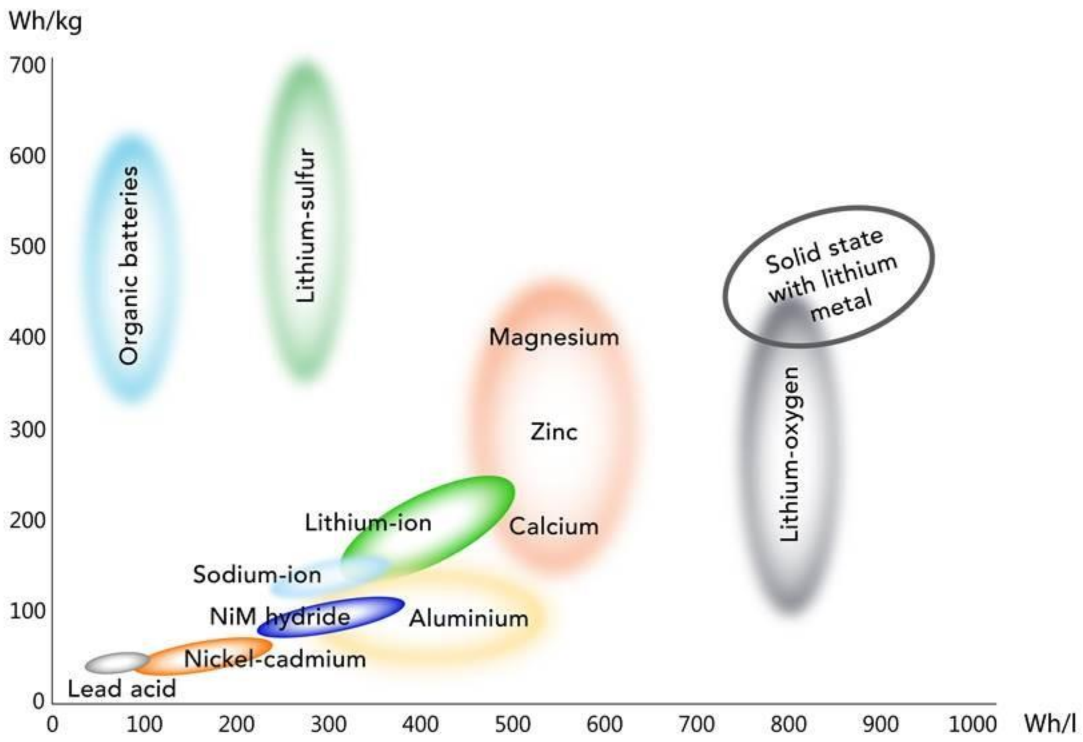


Figure 8. Current commercial batteries and targeted performance of future possible chemistries. The post lithium batteries chemistries are given as names indicating all kinds of metal-type batteries in respective category. There is a large uncertainty of their respective position in the graph. NiM hydride refers to nickel metal hydride.

The Battery 2030+ initiative intends to push the current state of the art for energy content by embracing the multiple possible future battery chemistries shown in Figure 8. Special attention is paid to future chemistries important for the transport industry as well as stationary storage and to realising targets set by various international roadmaps and by the EU SET Plan. Figure **9** compares the European goals (shown in green), based on the development of different generations of batteries, with those of China, Japan, and the USA.

Some international targets for automotive batteries expected for the coming years are shown in Figure 9.[90] The green line represents the different generations of LIBs and when they are expected on the market, according to the SET Plan. China, Japan, and Europe all have very similar expectations and almost overlapping targets, with the solid-state battery project to be on the market around 2030.

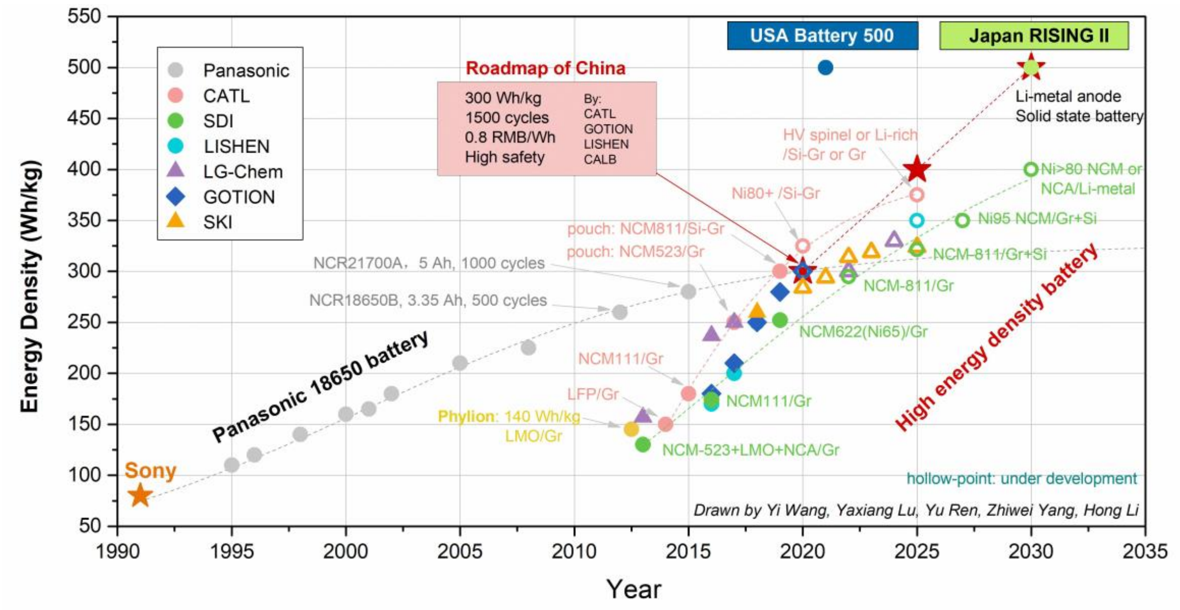
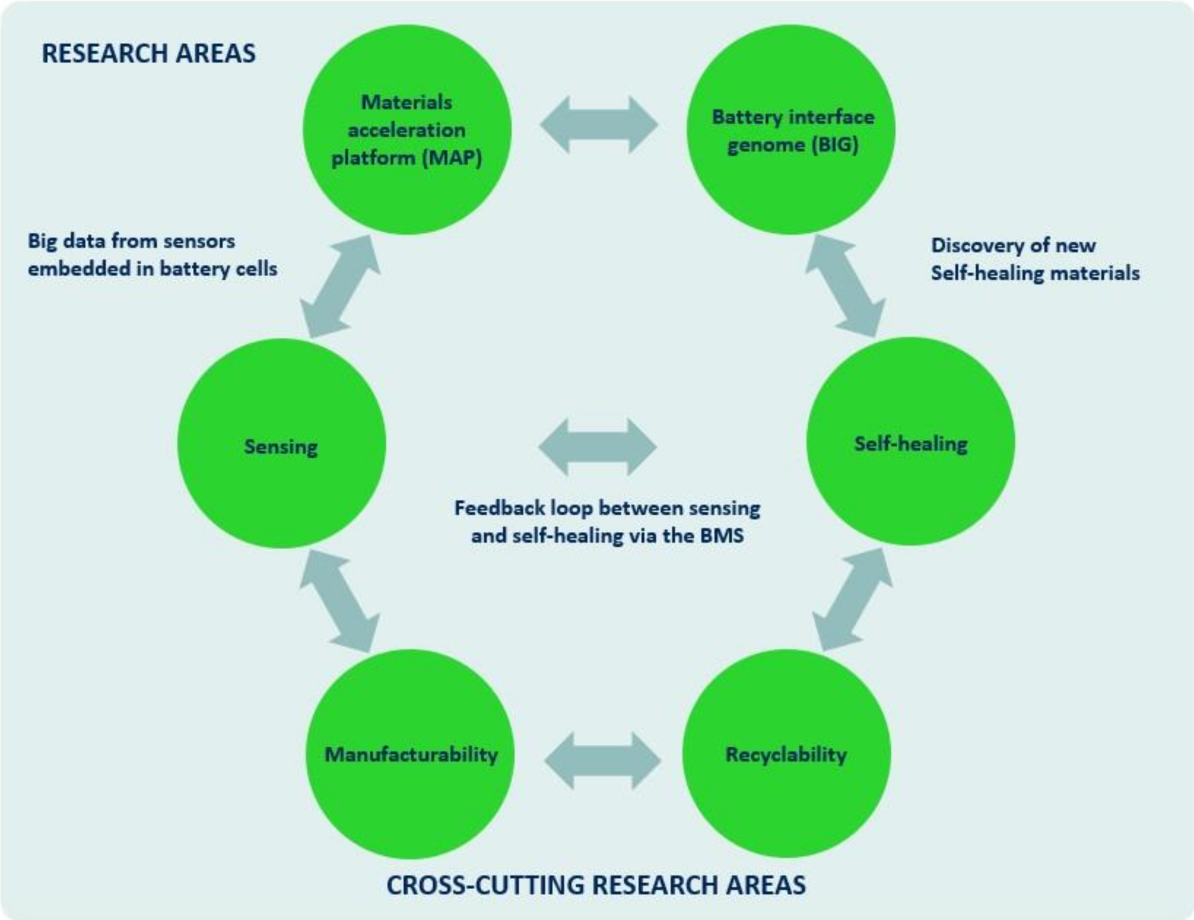


Figure 9. Comparison of the gravimetric performance of different batteries for automotive applications. The targets from the SET Plan coincide with the green line (different NCM-based generations of lithium-ion batteries). Japanese Rising II follows targets similar to those of the SET Plan, while China's targets (red stars) are slightly more ambitious up to 2030. The expectations for the lithium-metal solid-state battery are the same in all roadmaps. This figure was provided by Professor Hong Li of the Chinese Academy of Sciences.[90]

In comparison, Battery 2030+ sets forth challenge-driven research actions and identifies roadblocks to be addressed to reach the goals of the SET Plan. **Battery 2030+ therefore does not target a specific technology, but instead aims to invent the tools needed to radically transform the way we discover, develop, and design ultra-high-performance, durable, safe, sustainable, and affordable batteries.** In an international context, this is new and offers a unique touch to the European research eco-system. This statement is based on a bibliometric study, surveys of the actual larger programs existing globally and in Europe.[52] Through this approach, Battery 2030+ is intended to foster harmonised and coherent cooperation in Europe. As far as we can see, this approach differs from those expressed in the available published international roadmaps.

7 Research areas of Battery 2030+



The areas of research advocated by Battery 2030+ rely on these cross- and multidisciplinary approaches, with a strong wish to integrate other areas of research to enable cross-fertilisation. In this section, detailed descriptions of the research areas proposed in this roadmap are given. Each section describes the current status in the field, the challenges and expected progress in realising the vision, and the overall objectives of Battery 2030+.[7] Beyond the specific research areas, Battery 2030+ engages in defining new standards for research data management and for protocols to enable a higher level of interconnectivity between the projects and the whole battery community.[91] The European battery research and development landscape is well equipped to carry out the ideas proposed in this part of the roadmap. There are state-of-the-art high-throughput robotised material screening laboratories available in Europe as resources. Furthermore, Europe provides access to high-performance computing, the EuroHPC, and expertise within the European Materials Modelling Council. In addition, there are a number of synchrotrons and neutron facilities in Europe represented by the organisations League of European Accelerator-based Photon Sources (LEAPS) and League of Advanced Neutron Sources (LENS), which are resources with potential to enable the BIG–MAP initiative.

7.1 Materials Acceleration Platform (MAP)

Materials discovery, optimization, design, and development crosscut the entire clean energy research and utilisation portfolio. Advanced materials are at the foundation of European prosperity, competitiveness, social security, and quality of life. Innovations in clean energy conversion, storage, and use, particularly for emerging battery technologies, will shape the European high-tech market in the coming decades. Achieving, maintaining, and extending technological advantages is, therefore, both scientifically and politically imperative and demands enabling technologies to be spearheaded within the European Union. Relying on traditional trial-and-error–based discovery, research, and proliferation processes toward commercialization will lead to economic stagnation or decline. In Battery 2030+, we outline a new path for the accelerated discovery and rapid development of ultra-high-performance, sustainable, and intelligent batteries, which hinges on the development of faster and more energy- and cost-effective methods of battery discovery and manufacturing.[12]

This section outlines the opportunities, challenges, perspectives, and status of establishing a community-wide European battery **Materials Acceleration Platform** (MAP),[41] to be integrated with the **Battery Interface Genome** (BIG) described in the next chapter. The emerging BIG–MAP discovery framework and shared data infrastructure,[91] as developed in the www.big-map.eu project, has now been demonstrated to be both modular and versatile in order to accommodate all current and future battery chemistries, material compositions, structures, and interfaces. Drawing from initial inspiration from initiatives like the Materials Genome Initiative[92] and following the format of Mission Innovation: Clean Energy Materials (Innovation Challenge 6) MAP Roadmap,[41] and further conceptual iterations,[93] BIG-MAP is utilizing AI and machine learning to integrate and orchestrate data acquisition and utilisation from a number of complementary approaches and technologies, which are summarized in Figure 10 and discussed in the sections below.

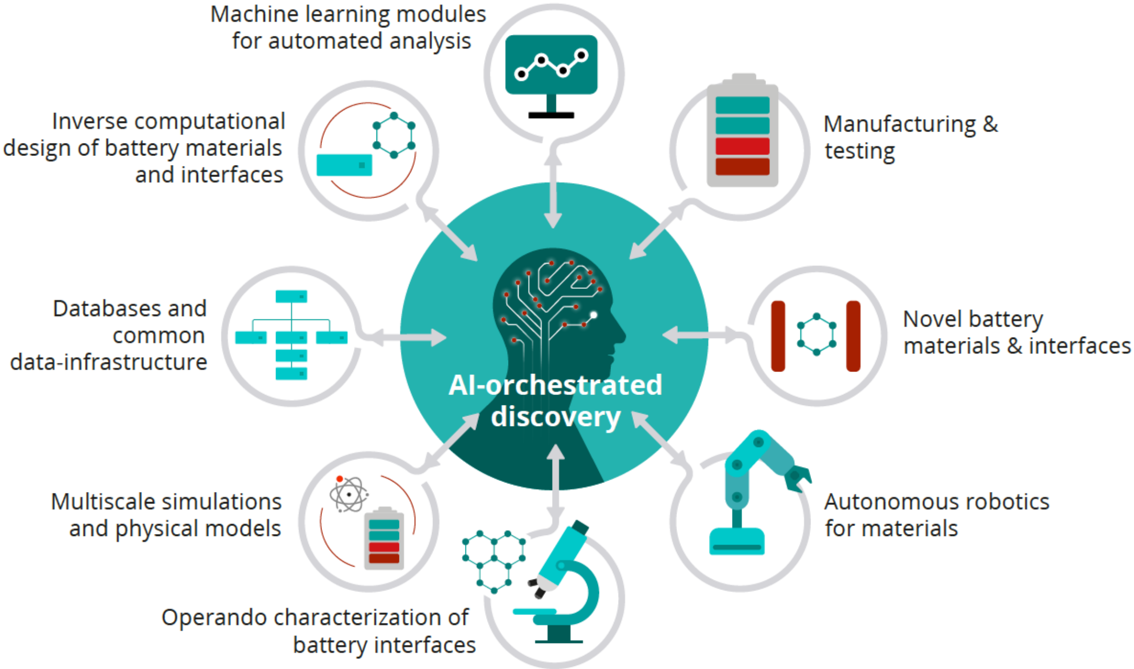


Figure 10. Key components of establishing the battery Materials Acceleration Platform BIG-MAP (www.big-map.eu).

Realizing the full potential of each of the core elements of the battery MAP framework still entails significant innovation challenges and the development of key enabling technologies. These technologies enable a completely new battery development strategy by facilitating the inverse design and tailoring of materials, interfaces, processes, and devices. Ultimately, coupling all MAP elements will enable AI-orchestrated and fully autonomous discovery of battery materials and resulting cells with unprecedented breakthroughs in development speed and performance as well as safety aspects.[17]

The successful integration of computational materials design, AI orchestration and machine learning, modular and autonomous synthesis, robotics, and advanced characterization will be the foundation for dramatically accelerating the traditional materials discovery process.[94] The creation of autonomous, "self-driving" laboratories[95] capable of designing and synthesising novel battery materials and of orchestrating and interpreting experiments on the fly, will create an effective closed-loop materials discovery process. Its development and practical implementation are presently pursued in the large-scale European project BIG-MAP,[96] which constitutes a quantum leap in accelerated materials design and discovery, which can be achieved only through the integration of all relevant European expertise. This has recently been demonstrated in the implementation of the decentralised and asynchronous MAP "FINALES",[97] which supports multiple tenants across different laboratories and countries.

7.1.1 Current status

Conventional research strategies for the development of novel battery materials have relied extensively on an Edisonian (i.e., trial and error) approach, in which each step of the discovery value chain is sequentially dependent upon the successful completion of the previous step(s). While several steps of the process have been automated and integrated in part, until now, only smaller steps have been taken toward full autonomy and closed-loop discovery.[14] Building upon the advances in the BIG-MAP project, there is now an integrated AI-accelerated combinatorial and high-throughput (HT) ecosystem. Examples include electrolyte formulation,[98] full battery cell building, end of life (EOL) characterization, rapid half-cell experiments, and hyperspectral mapping capabilities. Through the development of initiatives like FINALES, these systems are now being integrated with machine learning, atomic and multi-scale computational material design, and *operando* characterisation[99] techniques in a circular design loop. They can accelerate the discovery cycle of next-generation battery technologies, such as high-capacity Li-ion cathodes[100] and materials for secondary metal–air batteries.[101] However, additional layers of interoperability and further acceleration are needed to reach the highly ambitious goals of Battery 2030+. Especially, to react to the various battery prototype options and material design and synthesis routes; making these accessible for a wide range of HT optimisations requires a synergetic and wider material approach on the EU scale. Ideally, such a circular materials development process should integrate experimental and theoretical research in a closely coupled development platform that enables near-instantaneous cross-fertilisation of the results of complementary techniques, carefully defining performance, and characterisation proxies to measure the optimised battery structures and materials. In the following sections, we summarise the state of the art in key areas of MAP.

Interoperable data infrastructures and ontologized archives and databases are central requirements for the accelerated rational design of battery materials and interfaces, to ensure access to and the interoperability of high-quality FAIR data[43] and multi-sourced and multi-fidelity data from different scales and domains, such as experiments, testing, and modelling. Many ongoing efforts in North America, e.g., the Materials Genome Initiative, Europe, and beyond, aim to create extensive, flexible, and shareable databases and repositories[102,103] for experimental data. Additionally, computational infrastructures such as PRACE and EuroHPC, and platforms such as ASE[104]/MyQueue, SimStack,[105] AiiDA,[106] and Materials Cloud[107] facilitate workflows[16] for efficient and reliable high-throughput calculations[16], while only few examples like the OPTIMADE[108] REST API bridge computational and experimental data. Recent EU-led efforts generated data infrastructures specifically aimed at battery-related data capable of handling the types and quantities of heterogeneous multi-sourced data envisioned here. To fully exploit these data, extensive efforts, for example, by the European Materials Modelling Council (EMMC),[109] have been made to develop ontologies (e.g., EMMO), i.e., common knowledge-based representation systems, to ensure interoperability between multiple scales and different techniques and domains in the discovery process. A battery interface ontology BattINFO[13,110] is under development in the BIG-MAP project, which is now being integrated with other ontologies across different scales, e.g., manufacturing, and domains to facilitate the work of battery experts in different fields to convert real-life observations to a common digital representation. There are substantial efforts to establish standardised infrastructures that allow users to acquire, store, preserve, track, and share data in a curated, well-defined format that can be accessed from different platforms and for different purposes. A detailed and dynamic Data Management Plan (DMP) has been established to coordinate these efforts and ensure a linkage between data and tasks of the project.[91] To make it more operational, the DMP is being interconnected with BattINFO and, ultimately, should connect all the projects under the Battery 2030+ umbrella. With an outset in the structure and architecture of BIG-MAP data Archive (https://archive.big-map.eu) and the DMP-compliant data infrastructure, this option should be developed and opened for all projects in the Battery 2030+ community and alter possibly all EU battery projects, offering multiple layers of security and data-sharing, i.e., i) project-level, ii) Battery 2030+ restricted access, and iii) fully open source, e.g., in Materials Cloud (https://www.marialscould.org).

Multiscale modelling: Battery performance and lifetime are determined by many processes on vastly different time and length scales.[111] Simulating batteries across scales from molecules to battery packs and beyond requires insight from very different time and length scales, following the EMMC guidelines: (1) *electronic scale*, allowing the description of chemical reactions – electronic density functional theory (DFT) and *ab initio* molecular dynamics (AIMD); (2) *atomistic and mesoscopic scale* – molecular dynamics (MD) and kinetic Monte Carlo (KMC) simulations; and (3) *macroscopic scale* continuum simulations.[112] A single differentiable computational model of virtual materials design encompassing all these phenomena and scales is beyond the limits of current computing power and theory. However, advances in machine and deep learning models, and explainable AI (XAI) provides new possibilities for autonomous parameterization and advanced/hierarchical multi-scaling.[113,114] Fuelled by activities in BIG-MAP, significant efforts have been made to coherently combine traditional single-scale models into multi-scale workflows,[16] including the exploitation of AI and steps toward battery XAI. These methodologies are now being made accessible to the community in the online app store and GitHub repository, https://big-map.github.io/big-map-registry/, where the protocols are shared. An overview of the potential impact of these techniques is given in Bhowmik et al..[12] Multi-scale modelling techniques are currently being developed, for example, to optimise real and virtual electrode microstructures[115] and to study the effects of the fabrication process on cell performance[116] and electrode surface film growth.[117]

Experimental characterisation of materials and interfaces is important to map the chemical space across an extended range of time and length scales. Multiple datasets must be collected to fully characterise the battery cell behaviour, with different degrees of fidelity and reproducibility depending on the requests, from in-depth fundamental understanding to fast property screening. For instance, cutting-edge techniques available at large-scale facilities, such as synchrotron and neutron sources, play a key role in ensuring the sufficient acquisition of data describing battery materials and concomitant interfaces.[118] They provide information at unprecedented spatial and temporal resolution for a specific domain or process,[119,120] including in *operando* mode inside realistic cells. However, there is a clear need to go beyond the usual single-technique characterization schemes and obtain a more holistic vision to connect the pieces of knowledge gained individually by stand-alone experiments.[9] This relies on new infrastructures, such as the European multimodal platform which is the process of being constructed in BIG-MAP that will embed an array of ontologized tools capable of operating on request and producing multi-dimensional multi-parameter datasets. It calls for the ability to develop integrated and standardized multimodal workflows, including correlative analysis of multi-scale multi-technique data,[6] and to perform autonomous, on-the-fly analysis of the vast amounts of data generated at laboratory, synchrotron, and neutron facilities across Europe.[121] Correlative analysis and multimodal approaches have been introduced earlier in biology and materials science, but they are still in their infancy in the field of electrochemical storage.[122] In combination with big-data analytics using the advances in machine-learning methods, multiplexing the heterogeneous sets of time-resolved space-resolved data promises to expand both the quality and quantity of insightful observables, providing multifaceted descriptions of reaction mechanisms at relevant scales in relevant conditions at suitable locations. The state of the art of the most pertinent structural and spectroscopic characterisation techniques related to battery materials and interfaces is discussed in detail in Section 4.3.

Autonomous synthesis robotics,[11] which can be controlled and directed by a central AI, is a central element of closed-loop materials discovery. Highly automated, high-throughput syntheses are now becoming state-of-the-art for organic and pharmaceutical research,[123,124] and examples are also emerging in the development of solids and thin-film materials.[11,125] Automated high-throughput synthesis of polymer electrolytes can surely get wide inspiration from now-established approaches for HT synthesis of organic molecules. In contrast, automated HT synthesis of bulk inorganic materials is only in its early stages. For energy storage materials, robotic-assisted synthesis and automation have opened the field to the high-throughput screening (HTS) of functional electrolytes and active materials constituting anodes and cathodes. Still, introducing, e.g., Li-/Na-ion battery materials, where the structure and molecular vibration proxies such as on transition metal ions define the ion diffusion environment, can offer broader challenges than those known from pharmaceuticals. This requires a combined approach of method and HT-synthesis development for materials, which allows for fast making and screening, but also the opportunity via ML and AI-assisted algorithms to manoeuvre large data sets. Synthesis routes of inorganic battery materials that have been established in textbooks and industry can be applied but are not necessarily the best choices for fast screening and require a revision and evaluation towards the state of tech to check for best matches.

Combined with computational approaches such as data mining and the correlation of structure–property relationships with the performance of battery active materials, automation has significantly impacted the discovery of novel and promising materials.[123] A key aspect is transforming from automation to autonomy in synthesis and characterization. Ideally, through the combined high throughput experimentation and computational approaches, one can significantly shorten the time spans from 10+ years per material in the battery field to gain a faster and more rapid integration.

Experimental and computational high-throughput screening. Extensive libraries of compounds (e.g., salts, solvents, active materials, additives) can now be efficiently screened via the use of automated and miniaturised assays, which enable to accelerate of the electrode and electrolyte formulations R&D activities and optimised integration of relevant battery materials.[11,16] Coupled with large-scale data analysis, acceleration of certain parts of the materials discovery process by up to one order of magnitude or more now can be achieved, while it still needs to be demonstrated for the full battery discovery process.[126,127] On the computational side, workflows have been developed to automate different steps of the calculations needed to screen for new compounds.[128] These workflows should be integrated with the high throughput experimentation with the computational feedback loops to drive adaptation and optimisation of the battery compounds in the high throughput experiments. Several examples of fully automated high-throughput experimentation (HTE) systems for electrolyte formulation, cell assembly, and selected relevant electrochemical measurements are now available.[129]

AI in materials discovery offers excellent prospects,[130] but the complexity and challenges of the autonomous discovery of novel battery materials and interfaces are at a much higher scale of complexity than can be handled by existing methods.[94] The availability of vast, curated datasets for training the models is a prerequisite for the successful application of AI/ML-based prediction techniques. Software packages such as ChemOS[131], Phoenics[132], Olympus[133], "Hierarchical Experimental Laboratory Automation and Orchestration" (HELAO),[134,135] FINALES[97] and "Modular and Autonomous Data Analysis Platform" (MADAP)[136] have been used in prototyping applications to demonstrate key components of an autonomous, self-driving laboratory, which has not yet been achieved for battery applications. Also, various fast high throughput screening adaptive methods exist for battery materials that can be combined according to the computational efforts.

7.1.2 Challenges

Availability of FAIR[43] and curated data: The development of predictive models to design future battery chemistries requires thorough validation on the basis of curated datasets with data of diverse quality (fidelity). In particular, the validation of the complex models required for the inverse design[137] of battery materials and interfaces requires the integration of high-fidelity data[43] covering complementary aspects of the material, interfacial and device characteristics. Currently, such datasets are sparse and cover only a fraction of the required data space; in particular, ontologies must be developed to make the data discoverable. The BIG-MAP Archive is one step closer to enabling this. However, it requires that the battery community increases its engagement in sharing curated data.

To accelerate development, a consolidated strategy to overcome current bottlenecks must be implemented to ensure the success of the Battery 2030+ initiative. Currently, the exploitability of existing data and databases remains very low, partly because of the vast size of the design space, and partly because system requirements impose constraints on materials that go beyond the optimisation of individual performance indicators. A central aspect is the uncertainty quantification and fidelity assessment of individual experimental and computational techniques as well as of generative deep learning, which pose a key challenge. Here, the central aspect is "knowing when you don't know" and knowing when additional data and training are needed (see Figure 11).[138]

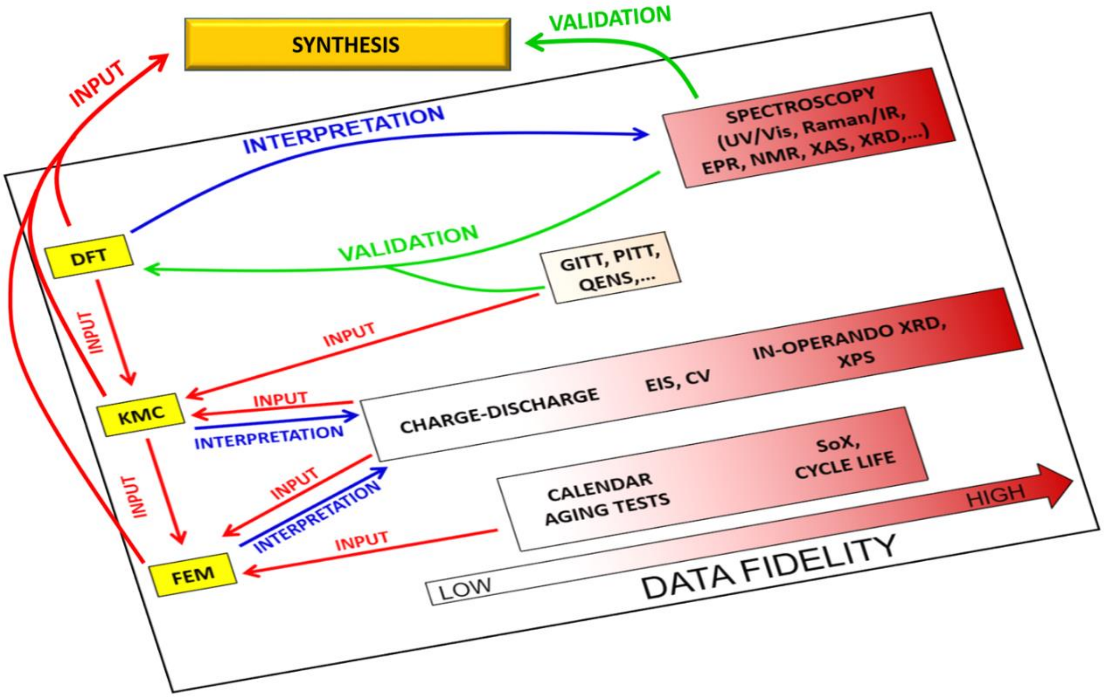


Figure 11. Illustration of the data flow between representative experimental and theoretical methods for studying battery interfaces. The fidelity of each method is generally proportional to its cost, but the fidelity–cost relationship can be optimised by acquiring data only when the given method/data is most valuable (adapted from[138]).

While machine learning could potentially massively accelerate the screening and identification of, for example, the structure–property relationships of inorganic energy materials,[139] a key challenge in the discovery of battery materials and interfaces is the development of autonomous workflows[16] for extracting fundamental relations and knowledge from sparse datasets[140] spanning a multitude of experimental and computational time and length scales.

Challenges for closed-loop materials discovery: To ensure full integration of data from experiments and tests into MAP, automated protocols for data acquisition and analysis must be developed. Currently, there are few examples of automated robotics for solid-state synthesis[95,125] and automated approaches for characterising battery materials and cells are also underdeveloped, while complementary approaches and inspiration can be drawn from activities in related fields and consortia like the Canadian Acceleration Consortium (https://acceleration.utoronto.ca/) and the Danish Pioneer Center for Accelerating P2X Materials Discovery (CAPeX, www.capex-p2x.com). Synthesis methods that have worked well to optimise battery materials in the lab may not always be the first line for high throughput and require careful evaluation. Next to the automated HT synthesis, high-throughput characterizations should be developed to validate the synthesized samples. This includes developing methodologies for the automated HT measurements (implementing robots, but in some cases adapting the sample preparation and measurement protocols to gain efficiency). And then this requires developing tools for the automated analysis of the large amount of data generated. Two examples from the BIG-MAP project are the PRISMA and FullProfAPP, which enable automated phase analysis and Rietveld refinements of large sets of X-ray diffraction (XRD) data (large series of individual samples or operando XRD data). Such tools need to be developed for other characterizations techniques. Several machine-learning–based tools have recently been developed to help in the data interpretation of relevant characterisation techniques, for example, NMR and XAS.[141,142] We also see a wide perspective to add here Raman spectroscopy as it is uniquely fit to capture light-weight elements like Li+ and Na+ and small structural changes in the realm defining diffusivity and capacity in those materials. These tools will enable automated analysis, but a wider portfolio of techniques with high predictability is needed to support a fully autonomous materials discovery platform. Higher-throughput characterization could also be developed at large-scale facilities, as the advent of fourth-generation synchrotron and X-ray free electron lasers open perspectives towards serial screening of materials and devices, also permitted by the massive progress towards fast online data reduction and processing. The high brilliance and high coherence of nano/micro beams also enable them to observe the structure and dynamics of matter at unprecedented levels, promising discoveries at the particle and local interfaces scale and key high-precision information beyond the usual bulk averaged, or electrode averaged measures.

An important bottleneck in closed-loop discovery is the lack of robust and predictive models of key aspects of battery materials and concomitant interfaces. A key challenge in this regard is the urgent need to increase the predictive ability of material synthesizability by modelling (i.e., identifying suitable equilibrium and out-of-equilibrium computable descriptors that effectively control synthetic networks). This pertains both to physics/simulation-based and data-driven materials discovery strategies. Only the full integration of physics/simulation-based and data-driven models generated through the exploitation of AI technology and recent network science developments[143] with automated synthesis and characterisation technologies will enable the envisioned breakthroughs required for the implementation of fully autonomous materials discovery.[138]

Another aspect of closing the loop towards an accelerated materials discovery by automated data analysis is the broad implementation of ontologies and standards within BIG-MAP itself as well as across all research areas which create input data for BIG-MAP. Short, medium and long-term goals on the way towards the accelerated research by use of ontologies and standardization were identified and are presented in Table 5.

In the short term, the development of an ontology defining a unified terminology and categories, properties, and relations for R&D data throughout Battery 2030+ is of high priority, with the BattINFO ontology and the BIG-MAP electronic lab notebook already in place.[144,145] The adoption of a unified ontology will be enabled and facilitated by implementing Electronic Lab Notebooks (ELNs). Ontologies and standards must eventually be made available by the scientific community. By their broad application, data will be made entirely FAIR.[43] For BIG-MAP, one primary goal to be reached is well-defined and standardised interfaces, enabling full reproducibility and interoperability. At this point, the Electronic Lab Notebook (ELN) could represent one step to the Platform as a Service (PaaS) and Lab as a Service (LaaS).

7.1.3 Advances needed to meet challenges

European strongholds in the battery community have always been at the forefront of the development of future battery technologies. This has resulted in a leading position regarding active materials development, the design of new liquid or solid electrolytes, development beyond LIB chemistries, as well as new experimental and computational tools to understand complex redox reactions at the heart of these electrochemical systems, to name but a few relevant areas. World-leading initiatives already exist at both the multinational level, for example, ALISTORE-ERI, and the national level with, for instance, the French network for electrochemical energy storage and conversion devices (RS2E), the Battery Pilot Hub in Grenoble, the Faraday Institution in the UK, and CELEST (Center for Electrochemical Energy Storage Ulm & Karlsruhe), MEET (Münster Electrochemical Energy Technology), POLiS (Post Lithium Storage Cluster of Excellence), e-conversion consortia in Germany, demonstrating that partnerships can be created beyond individual laboratories. The European research community is ready to support a truly European research effort dedicated to advancing our knowledge of battery materials by the creation of a European battery materials acceleration platform, combining the complementary strengths of each partner with the strongly collaborative existing environment.

Autonomous synthesis and battery-cell characteristic test robotics: The complex nature of the material synthesis and electrochemical characterisation of battery cells and testing are among the major bottlenecks slowing the development of new battery materials and altering performances and stabilities for the batteries.[9,11] To explore larger classes of materials in the context of specific characteristics and optimisations of their structure-property relationships, it is essential to advance the development of high-throughput synthesis robotics that address both electrolyte formulations and electrode active materials, as well as combinations thereof, both for the characterisation of the materials as such and in the context of functional cells. This requires various approaches for synthesis methods starting from either precursor pre-screening and solution development up to solidified material/layer compounds of materials and cell-level characterisation of performances.

High-throughput/high-fidelity characterisation: Even though an increasing number of approaches to the high-throughput testing of battery materials is reported in the literature,[146–148] many electrochemical tests do not work on short time scales; in particular, cycling experiments can take days to months or even years.[149] To exploit the opportunities afforded by the vast number of samples, an automated high-throughput infrastructure for the in situ and in *operando* characterisation of battery materials and resulting cells have to be established, including the development of versatile multimodal cells, standardised galvanostatic cycling protocols, and sample transfer methods. This infrastructure must address the issues of width and depth and should include filtration by identified hit/lead candidates. The combination of physics-guided data-driven modelling and data generation is required to enable the high-throughput testing of batteries, and they are incorporated active materials in the future, thus developing a battery materials platform for the accelerated discovery of new materials and interfaces.

A cross-sectoral data infrastructure: Accelerated materials innovation relies on the appropriate and shared representation of both data and the physical and chemical insights obtained from them.[124,150] This poses a substantial challenge to the international research community, which needs to join forces in establishing, populating, and maintaining a shared materials data infrastructure as well as corresponding data interfaces and standards.

Establishing a common data infrastructure will help ensure the interoperability and integration of experimental data and modelling in a closed-loop materials discovery process across institutions in real time. Realising such an infrastructure will make the data generated by individual groups and consortia instantly available to the community at large and drastically shorten R&I cycles. MAP will pioneer such an infrastructure based on a decentralised access model in which data, simulation protocols, and AI-based discovery tools and components from different sources can be used *via* qualified access protocols.

Scale bridging and integrated workflows:[9,16] The root of the multi-scale challenge is that it is not known how best to couple models and correlative data analysis at different scales in an effective and robust way. Essentially all effects observed at the macroscopic (e.g., cell) level are rooted in phenomena at the atomistic level, which generally are of quantum nature. The large gain in accessible time-scales and size of larger-scale models generally entails the sacrifice of detail and resolution. While there has been substantial progress in specific relevant use cases in the development of multiscale methods, releasing the full potential of multi-scale experimental characterization and modelling and inverse design requires a continued coherent effort by the modelling & experimental communities. We need integrated efforts to link scales beyond state of the art to address open challenges, such as SoX, manufacturability, and sustainability of batteries.[137] This can be achieved only by establishing interoperable workflows, which can communicate across various workflow engines, simulation codes, and experiments. Machine learning techniques and other physics-guided, data-driven models can be used to identify the most important parameters, features, and fingerprints[151] and also to help bridge the scales where there is no clear overlap of the models. They will also guide the design of experiments and analysis of multiple sets of data acquired across an extended range of time and length scales beyond standard single-shot stand-alone experiments. Surrogate models can be employed where no physical models are available. We see great potential to codify the workflows to the degree that they can be used outside the group which has developed them via accessible App-stores. In addition to purely computational workflows, workflows that integrate on-the-fly and/or highly-specific cutting-edge experiments (and vice versa) hold a huge potential to accelerate materials discovery. The integrated technology to realise such workflows is just on the verge of becoming accessible. MAP will exploit European computational infrastructures, such as those offered by PRACE and EuroHPC JU facilities like LUMI as well as the results of prior and ongoing EU and national funding efforts, for example, former and ongoing centres of excellence in HPC applications such as NOMAD and MaX. MAP will also exploit European experimental platforms such as the European Battery Hub providing new access modes to synchrotron and neutron facilities and a collaborative framework for AI-aided standardized multimodal characterization. While presently, most simulation and experimental efforts are directed towards understanding battery function, with an increasing emphasis on design, additional efforts are needed to develop models to address the full battery cycle life.

AI exploitation: AI-based generative models,[152] i.e., probabilistic models of observed data on the spatio–temporal evolution of battery materials and interfaces, can significantly contribute to the goals of MAP, and developing hybrid physics and data-driven models will be an essential part of MAP. Currently, there exist significant gaps in the spectrum of battery models, which preclude the development of comprehensive and accurate representations. Although AI-based techniques can potentially address these gaps, they often lack awareness of physical laws, leading to potential violations. The key to overcoming this dilemma is the development of hybrid models, which incorporate both AI-based predictions and the constraints imposed by the laws of physics. By combining the strengths of AI and physical models, we can create a synergy wherein AI is employed to adapt and enhance physical models or where the laws of physics appropriately bind the prediction space of AI-based models. However, these models must be trained on large, curated datasets from advanced multi-scale computational modelling, materials databases, the literature,[153] and *operando* characterisation. These data must span all aspects of battery materials from synthesis to cell-level testing.[138]

Unification[16,154] of protocols: MAP will offer a unique opportunity to leverage the size of this effort in the interest of standardising data and workflow methodologies from the entire battery value chain, by exploiting semantic access protocols enabled by EMMC and EMMO and by tapping private groups, with the goal of connecting academia and industry, materials modelling and engineering.[155] The development of an Open Battery Innovation Platform is needed to facilitate the sharing of infrastructures and data between partners and the integration of modelling into industrial processes to close the gap between in silico materials design, battery cell manufacturing, and their end use in everyday devices.

The inverse design of battery materials and interfaces effectively inverts the traditional discovery process by allowing the desired performance goals to define the composition and structure of the battery materials and/or interfaces that best meet the targets without a priori defining the starting materials. Presently battery interfaces emerge largely by experimental trial-and-error and design guided by intuition as a result of long processes (on the molecular time-scales). Computational or hybrid computational-experimental efforts to inverse design battery interfaces on the basis of the constituents of the system hold great promise to revolutionize battery performance and life-time. Interface-specific performance metrics at different time and length scales should be defined to gain a reasonable degree of control over how the interface evolves over battery lifetime.

7.1.4 Forward vision

Autonomous BIG–MAP: Our future vision is to develop a versatile and chemistry-neutral framework capable of achieving a 5–10-fold increase in the rate of discovery of novel battery materials and interfaces. The backbone of this vision is the Battery Interface Genome–Materials Acceleration Platform (BIG–MAP), which will ultimately enable the inverse design of ultra-high-performance battery materials and interfaces/interphases, and be capable of integrating cross-cutting aspects such as sensing (Section 7.3), self-healing (Section 7.4), manufacturability (Section 7.5), and recyclability (Section 7.6) directly into the discovery process.

The full BIG–MAP will rely heavily on the direct integration of the insights developed in BIG (Section 7.2) and the novel concepts developed in the area of sensors and self-healing, which will be discussed in Sections 7.3 and 7.4.

***In the short term*:** Develop a shared and interoperable data infrastructure for battery materials and interfaces, linking FAIR,[43] ontologised data, and metadata from all domains of the battery discovery and development cycle. Use autonomous workflows to identify and pass features/parameters between different time and length scales. Develop hybrid physics and uncertainty-aware data-driven models of materials and interfaces. These developments will be supplemented by establishing the necessary ontologies, data standards, and protocols.

***In the medium term*:** Implement BIG in the MAP platform (BIG–MAP), capable of integrating generative computational modelling, autonomous synthesis robotics, and materials characterisation. Successfully demonstrate the inverse design of battery materials and computational workflows supported by AI to model battery interfaces. Directly integrate data from embedded sensors in the discovery and prediction process, for example, to orchestrate the pre-emptive launch of the developed self-healing additives. Demonstrate transferability of the BIG–MAP approach to novel battery chemistries and interfaces, e.g., multivalent chemistries and flow battery materials.

***In the long term*:** Establish and demonstrate full autonomy and chemistry neutrality in BIG–MAP. Integrate battery cell assembly and device-level testing. Include manufacturability and recyclability in the materials discovery process. Demonstrate 5–10-fold acceleration in the materials discovery cycle through integrated experimental-computational workflows and development and use foundation models for the battery domain to explore the unknown unknowns. Implement and validate digital twins of ultra-high-throughput testing on the cell level and bridge to digital twins for processing and manufacturing.

7.2 Battery Interface Genome (BIG)

Experience has shown that when developing new battery chemistries or introducing new functionalities into existing battery technology, interfaces hold the key to exploiting the full potential of the electrode and electrolyte materials toward developing ultra-high-performance, sustainable, and smart batteries. The European battery R&D landscape consists of a multitude of leading research institutions, laboratories, and industries, many of which pursue complementary approaches to tackle this challenge in isolated efforts. With the Battery Interphase Genome (BIG), we seek to bring together expertise from across sectoral competences, industries, and end users to install BIG for accelerating the development and exploitation of radically new battery technologies. BIG and MAP will be deeply intertwined, where the MAP builds the physical and virtual platforms to support the understanding and insights generated in BIG.

Existing research methodology relies largely on incremental advances at the local scale, which are not pertinent for tackling the ambitious challenges within the timeline outlined in this roadmap. MAP will provide the infrastructural backbone to accelerate the application of our findings. At the same time, BIG will develop the necessary understanding and models for predicting and controlling the formation and dynamics of the crucial interfaces and interphases that limit battery performance.[156] In this respect, we must take into account studies of ion transport mechanisms through interfaces and, even more challenging, visualise the role of electrons in these interfacial reactions. Furthermore, as it remains an open question what the winning battery technologies will be for large-scale grid storage, mobility, etc., BIG will be highly adaptive to different chemistries, materials, and designs, starting from beyond state-of-the-art Li-ion technology, where substantial data and insights are available for training the models, to emerging technologies like Na-ion and all-solid-state, and radically new chemistries.

Batteries comprise not only an interface between the electrode and the electrolyte but a number of other important interfaces, for example, between the current collector and the electrode and between the active material and the additives, such as conductive carbon and/or binder and buried interfaces. Important also are interfaces between several types of active materials in composites and/or complex nanostructures with a hierarchy of active particles. Realising this, any globally leading approach to mastering and inversely designing battery interfaces must combine the characterisation of these interfaces in time as well as in space (i.e., spatio–temporal characterisation) with hybrid physical- and uncertainty-aware data-driven models.[157,158] Thereby integrating dynamic events at multiple scales, e.g., across the atomic and micrometer scales. In this respect, we must consider studies of ion transport mechanisms through interfaces and, even more challenging, visualise the role of the electron in these interfacial reactions. When mastered, interfacial reactivity helps to extend the thermodynamic and kinetic stability of organic electrolytes used in batteries; when it is not controlled, however, continuous parasitic reactions may occur, limiting the cycle life of batteries. The complexity of such interphases arises from multiple reactions and processes spanning a wide range of time and length scales that define their formation, structure, and, ultimately, their functionality in the battery. Their structural properties depend in a highly complex and elusive manner on the specific characteristics of the composition of the electrolyte, the structures of the electrode materials, and the external conditions. Understanding, controlling, and designing the function of interfaces and interphases[9] is, therefore, key for the development of ultra-performing, smart, and sustainable batteries.

The Battery Interface Genome – BIG – can be related to the concept of descriptors in catalyst design,[159] in which the binding energy of important reaction intermediates scales with that of the descriptor, and the identification and quantification of the descriptor value enables an accelerated and accurate prediction of the rate of the total reaction. Identifying the multiple dynamic descriptors (or genes) coding for the spatio–temporal evolution of battery interfaces and interphases is a prerequisite for the inverse design process and simply cannot be done using existing methodologies. This requires improving the capabilities of multi-scale modelling, machine- and deep learning, and systematic multi-technique characterisation of battery interfaces, including *operando* characterisation, to generate/collect comprehensive sets of high-fidelity data[160,161] that will feed a common AI-orchestrated data infrastructure in MAP. BIG aims at establishing the fundamental "genomic" knowledge of battery interfaces and interphases through time, space, and chemistries. The BIG will be chemistry neutral, starting from state-of-the-art Li-ion technology, where substantial data and insights are available for training the models, to emerging technologies like Na-ion and all-solid-state and radically new chemistries.

7.2.1 Current status

Battery interfaces and interphases – where the energy storage in batteries is facilitated but also where many degradation phenomena are initiated—have always been both a blessing and a major limitation in battery development. For instance, the growth of the so-called solid electrolyte interphase (SEI) on the anode as well as cathode electrolyte interphase (CEI) on the cathode has both a significant impact on the one galvanostatic cycling stability as well as safety of LIBs. Thus, when mastered, interfacial reactivity helps to extend the thermodynamic and kinetic stability of organic electrolytes used in batteries; when it is not controlled, however, continuous parasitic reactions may occur, limiting the cycle life of batteries. Understanding, controlling, and designing the function of interfaces and interphases is, therefore, key for the development of ultra-performing, smart, and sustainable batteries.

In comparison with the bulk dimensions of the electrode and electrolyte (~µm), the interface (or interphase) is several orders of magnitude smaller (~nm), and interfacial reactions are easily masked by their surroundings. Experimental and computational techniques must therefore be highly surface sensitive with exceptionally high resolution to probe such buried interfaces. Nevertheless, the experimental characterisation of battery interfaces has been an enduring challenge. Indeed, very few, if any, techniques can provide a full description of the events happening at the electrode-electrolyte interface.

Experimental and computational techniques have the challenge of being surface and interphase sensitive. Thus, no singular technique is currently capable of providing a comprehensive description of events happening at the many types of buried interfaces. This opens significant opportunities to support experiments with high-fidelity computational models, in parallel to the development of characterisation techniques capable of probing the chemical and morphological properties of interphases, intensive research efforts have been devoted to developing chemical and engineering approaches to control the dynamics of the interphases upon galvanostatic cycling. The most prominent approach is using electrolyte additives/co-solvents that react inside the cell during initial operation and coatings that can passivate the surface of electrode materials and thus prevent inevitable reactivity with the electrolyte. However, many years of Edisonian trial-and-error research have demonstrated the need to use several functional additive working in synergy to achieve an effective electrode-electrolyte interface. Accelerated development of such an interphase would greatly benefit from high-throughput techniques and AI-assisted rationalisation. We see perspective in defining such model fast screen interphase systems for stability and diffusion studies by using thin and thick film tech and other methods for fast alterations of the interphase microstructure and chemistry. In fast screen diagnostics, methods looking at the interphase based on spectroscopy are ideal for differentiating through frequency tailoring and possibly wavelength screening close and far from the interphase changes in chemistry and structures.

Physics- and uncertainty-aware data-driven methods: The complexity of electrochemical systems usually motivates the simplification of simulations such that they only qualitatively mimic the real situation in the battery or the experiment. A coupling of physics-aware data-driven methods would strongly enhance the quality of the determination of interface descriptors, features, and parameters by enriching the physical simulation with validated correlations between idealised physics/chemistry-based simulations and data on real materials. Interoperability and scale-coupling are also a challenge for experiments, requiring non-intrusive *operando* data acquisition on realistic cells working in representative conditions and subsequent AI-aided correlative analysis of large data sets.

A complete and closed mathematical description of the whole reaction mechanism is enormously challenging and unlikely comprehensible, since coupled ionic and electronic transfer reactions in an electrochemically relevant environment usually include coupled multistep reactions.[162,163] These multistep reactions are often either oversimplified or the reaction steps are modelled in too ideal environments.[164] In specific cases, it is possible to combine DFT methods with classical approaches to improve the description of surface reactions,[165] but generic approaches remain limited, and an efficient and systematic coupling is still lacking.

7.2.2 Challenges

Interfaces and Interphases: Despite decades of research, the details of interfacial reactions in the complex electrochemical environments in batteries (e.g., the composition and function of the SEI) remain mysteries. The structural properties depend in a highly complex and elusive manner on the relevant properties of the electrolyte components and resulting compositions, the structures of the electrode materials, and the external conditions. The complexity of such interphases arises from multiple reactions and processes spanning a wide range of time and length scales that define their formation, structure, and, ultimately, their functionality in the battery.

Intensive efforts were made in recent years to uncover the complexity of the interface dynamics and to control its reactivity and functionality, acquiring an enormous dataset whose depth remains largely under-exploited. Data must be collected, handled, and analysed in a systematic and automated/autonomous manner to be accessible to the central BIG–MAP AI orchestrating the accelerated discovery process. To ensure meaningful synergy between experiments, simulations, and AI-based models, simulations and models need to become more realistic and include experimental conditions. Similarly, the experimental conditions should be as reproducible and exact (i.e., ideal) to decouple effects and reactions.

Multi-scale modelling concepts: Key challenges in this regard include the development of new multi-scale modelling concepts (including physics-aware data-driven hybrid models to identify interphase descriptors) and the development of new characterization techniques, particularly under electrochemical conditions relevant to the application. Datasets enabling the training of such models are just becoming available now. Standardisation of experimental data, conditions, and observables as inputs to physical models to make the link between observables and descriptors.

A fundamental understanding is the first step in controlling the complex and dynamic processes at the interfaces in emerging high-performance battery technologies. This understanding relies on the availability and development of adequate experimental and computational tools capable of probing the evolution of the dynamic processes occurring at the battery interfaces and making them understandable to scientists. These tools should selectively provide information on the interface region, and special efforts must be made to couple complementary experimental, simulation-based, and AI-based modelling tools.[166] It could be envisioned that mature battery interface/interphase characterisation techniques could provide high-throughput experimental input about battery interfaces during operation. One of the key challenges in establishing BIG is to automate the acquisition, curation, and analysis of large datasets. These could feed the physics-aware data-driven hybrid models that will help better understand and predict interfacial properties and enable direct multi-scale bridging by developing integrated multimodal workflows for correlative characterization.

Combining physical and data-driven models: This will only be possible if datasets are acquired from reliable temporally and spatially resolved experiments, including data recorded under working conditions (i.e., *operando* measurements) and spanning the full range from optimised laboratory-based to large-scale research-facility–based measurements and high-throughput synthesis and laboratory testing. Combining physical and data-driven models run on curated community-wide datasets spanning multiple domains in the discovery process will enable us to establish the BIG[167,168] for interface/interphase development and dynamics. This has the potential to lay the foundation for the inverse design of battery interfaces/interphases[138], for example, using region-based active learning algorithms.[169]

Uncertainty quantification: Understanding and tracking different types of uncertainties in the experimental and simulation methods, as well as in the machine learning framework of, for example, unsupervised[167] and generative deep learning models,[170] is crucial for controlling and improving the fidelity of the predictive design of interfaces. Simultaneous utilisation of data from multiple domains, including data from an apparently failed experiment,[171] can accelerate the development of generative models that enable the accelerated discovery and inverse design of durable high-performance interfaces and interphases in future batteries.

7.2.3 Advances needed to meet challenges

Novel computational and experimental techniques and their combination: The development of new computational and experimental techniques targeting increased spatial resolution, time domains, and *operando* conditions is needed to generate new insights into the construction of ultra-high-performing battery systems. Realising this development is challenging for both theoretical and experimental science, and enhanced collaboration between disciplines is necessary to unlock the next generation of battery technologies. Experimental input is needed to identify realistic input parameters for the development of new computational models, and modelling results need to be validated against experimental results[172]. . Likewise, the interpretation of experimental results can be made with higher precision if theoretical models can be used in combination with experiments.

High-quality/high-fidelity data and insights are required to develop the battery interface genome, high-quality/high-fidelity data, and insights are required, which calls for developing superior *operando* experimental techniques for establishing atomic-level understanding on smaller scales and various time scales and dimensions. Moreover, on-the-fly acquisition and analysis should be targeted to provide instantaneous input for the materials acceleration platform developed in MAP. BIG, therefore, offers a unique opportunity to develop a common European platform and common European battery standards for data acquisition and transfer that could serve as worldwide standards.

In addition to the continuous improvement of optimised existing as well as development of new experimental techniques and methodologies targeting the scale of atoms and ions and also spatially-resolving heterogeneous distributions of atoms and ions from nano to meso and micro-scales, radically new ways of combining experimental, theoretical, and data-driven techniques will be necessary. For example, developing novel experimental and computational techniques targeting the time and length scales of electron localisation, mobility, transfer reactions, ion dynamics, and distributions. Advanced physics-based hybrid models and simulation techniques must be used for the interpretation of cutting-edge operando experiments. Efficient methods for using large datasets to determine the descriptors of multi-scale/multi-structure theories have to be developed. This should also include recent progress on graph and network theory applied to electrolyte interphase formation.[173] With these technical advances, new insights will follow, allowing us to control access to the fine-tuning of the battery interface and thus develop the next generation of ultra-high-performing batteries.

European data infrastructure: Currently, no shared infrastructure or large-scale database of battery-oriented interface properties is available comparable to, for example, existing structure databases for organic and inorganic materials.[174–176] Implementing such European data infrastructure, starting from the BIG-MAP Archive, would require further development and utilisation of characterisation techniques capable of providing a high-fidelity description of the interfaces and their dynamics. X-ray–based techniques, as well as neutron-based techniques, are examples of techniques that will be critical, specifically when combined, in order to gain information about battery interfaces. Furthermore, to accelerate our findings, systematic measurements in parallel with multi-technique information/data from the same materials/interfaces must be established, representing a game-changing approach differing from the current single-technique paradigm.[9] High-throughput experiments should be designed to allow the investigation of a large number of samples at great comparability and reproducibility alongside the provision of pertinent auxiliary data. This requires workflows that can generate and analyse large amounts of data in an automated/autonomous and correlated manner, representing a major advance toward defining a new methodology for acquiring data on battery interfaces.

Standardised testing protocols and interoperability: A key advance needed to establish BIG is the design of standardised testing protocols for battery materials and cells to allow extraction of critical information regarding battery interfaces (and bulk properties) by comparing cell performance with cell chemistry. Guidelines should be defined for that purpose, becoming the project's characterisation quality label. This checklist should be aligned and complement previously published ones by scientific journals or other recently published large-scale initiatives to ensure interoperability within the scientific community. BIG represents a unique opportunity to design a common European strategy in which experimental data on each new chemistry, successful or not, will feed into a common data infrastructure that will be broadly accessible, for example, by a central AI orchestrating the materials discovery. To meet the challenges of standardising experimental data and observables as input to physical models, implementing feedback processes may be considered pivotal. This will be achieved by creating a European database of battery-oriented material properties and a standardised classification of interfacial phenomena, as well as by defining common observables for physical modelling used to initiate paths and feedback loops for the multi-scale integration of datasets and modelling. Moreover, to support the standardisation of the testing protocols, platforms will be implemented and opened to European partners to certify the performance of batteries, helping better integrate academia and industry. Therefore, efforts towards standardisation should not be restricted to electrochemical testing or materials properties but should cover manufacturing of battery components and battery assembly. A stepping stone towards that goal is the definition of an ontology for active materials synthesis and manufacturing step. To create feedback processes with physical insights provided by multiscale modelling, physical models, and multimodal characterization, implementation of standards regarding *operando* measurements, modelling, and simulation is also necessary. Finally, protocols for data sharing, storage, and analysis must be implemented efficiently to ensure the efficient transfer of not only metadata for electrochemical testing and characterization data but also of analysed data using automated analysis tools.

AI-enhanced multi-scale/multi-feature approaches combining different computational and experimental tools will certainly be necessary to grasp the dynamics of the interface at different scales rather than a single physical property.[111] Through the use of AI-based techniques linking BIG and MAP, complex connections/features between scales that are imperceptible to humans will be recognised, and areas available for reliable predictions will be extended to new realms. However, modelling interphases and probing is complex due to the phenomenon's variety. Here, we envision the development of more accurate models that address more realistic interfaces, aging, and degradation, as well as complex design scenarios, requiring adequate mathematical frameworks to couple electronic, atomistic, and mesoscopic models with continuum models. Integrated experimental and computational workflows merging advanced multi-scale modelling, ML, and data analytics will master the complex coupling of relevant length and time scales, which are so relevant to batteries. Similarly, we envision the development of more coordinated and integrated experiments to accelerate correlative characterizations and real-time multiparameter materials mappings in well-defined and controlled conditions. The development of inverse modelling techniques that map the data back to model parameters will accordingly be pursued.

7.2.4 Forward vision

While the traditional paradigm of trial-and-error–based sequential materials optimisation starts from a known interface composition and structure and subsequently relies on human intuition to guide the optimisation to improve the performance, the forward vision is to enable inverse materials/interface design, in which one effectively inverts this process by allowing the desired performance goals to define the composition and structure that best fulfil these targets without a priori defining the starting composition or structure of the interface. To develop and implement suitable models for the inverse design of battery interfaces, it is necessary to incorporate the relevant physical understanding and the model capable of performing an inverse mapping from the desired properties to the original composition of the materials and external parameters/conditions. The generative deep-learning models described in Section 7.1 represent an efficient way to optimise the data flow and build the required bridges between different domains, helping solve the biggest challenges of battery interphases (Figure 12).

Inverse design strategy**:** This reliance on statistical correlations renders descriptors an ideal tool for data-driven AI methods. A promising route is the full integration of data-driven methods and physical-theory–based simulations, for example, inverse modelling with experimental datasets is used to reliably determine the interface descriptors of the detailed spatio–temporal evolution. Based on these, forward simulations give insight into the expected spatially resolved time evolution of the system. With the outlined approaches, this finite number of parameters/features can be extracted by combining many simpler experiments using modern mathematical inverse modelling techniques and extracting a continuous four-dimensional spatio–temporal field of physical variables that can then be reduced to determining a finite set of parameters.

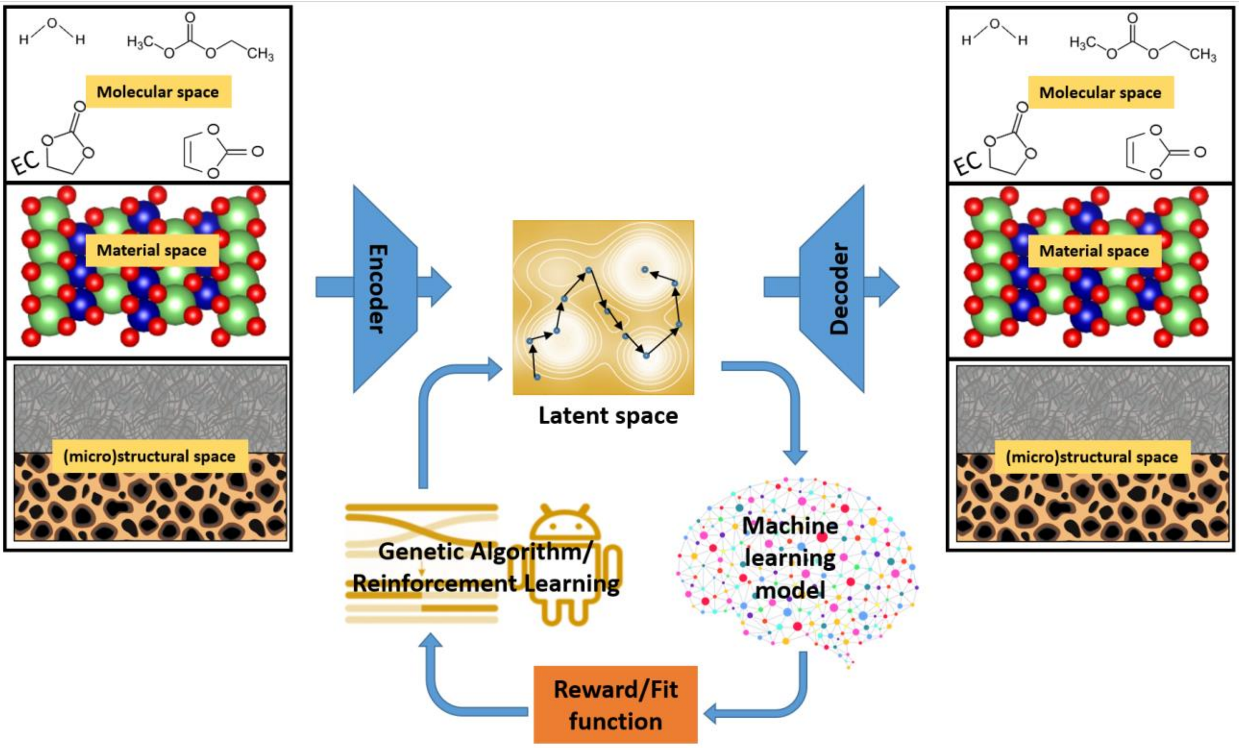


Figure 12. Generative model of interphase design. Variational auto encoder (VAE)-based encoding and decoding of chemical and structural information on a battery interphase into latent space, to enable generative battery interphase design through the use of, e.g., genetic algorithms or reinforcement-learning–based exploration[138]. Reprinted from Energy Storage Materials.[138]

By doing this, rather than the empirical development of battery chemistry and assembly, which has been the norm so far, we aim to develop inverse battery design driven by data input which will also benefit the investigation of both production and recycling processes. This will be done sequentially to achieve, within ten years, a fully autonomous and automated platform, integrating computational modelling, material synthesis and characterisation, battery cell assembly, and device-level testing (BIG–MAP). Finally, we envision the battery discovery platform and the battery itself as fully autonomous, utilising, for example, the sensors developed in Section 7.3 to send signals that can be understood by the central BIG–MAP AI to predict the spatio–temporal evolution of the interface. If the model predicts a potential failure at the interface, this will launch the release of self-healing additives, as developed in Section 7.4, to pre-emptively heal the interface and possibly increase the battery lifetime. Furthermore, the development of such an inverse design strategy will also benefit the investigation of both production (see Section 7.5) and recycling processes (see Section 7.6).

Full integration of **BIG–MAP** will occur stepwise according to the following combined timeline for Sections 7.1 and 7.2:

In the short term**:** Establish community-wide testing protocols and data standards for battery interfaces. Develop modules and apps for fast, automated analysis or even autonomous on-the-fly analysis of characterization and testing data using AI and simulations. Develop interoperable high-throughput and high-fidelity interface characterization approaches.

***In the medium term*:** Develop predictive hybrid physics- and uncertainty-aware data-driven models for the spatio–temporal evolution of battery interfaces. Demonstrate successful inverse design of battery materials and interphases. Integrate novel experimental and computational techniques targeting hard-to-abate time and length scales, e.g., of electrons and ion localisation, mobility, and transfer reactions.

***In the long term*:** Establish and demonstrate full autonomy and chemistry neutrality in the BIG–MAP platform. Demonstrate a 5–10-fold improvement in the interface performance. Demonstrate transferability of BIG to novel battery chemistries and interfaces.

7.3 Integration of smart functionalities: Sensing

Our increasing dependence on batteries calls for the accurate monitoring of battery functional status so as to increase their quality, reliability, and life (QRL).[17,18,177] In recent decades, numerous on-board electrochemical impedance spectroscopy (EIS) devices and sophisticated battery management systems (BMSs) have been developed for this purpose, but with limited success. Whatever battery technology is considered, its performance is governed by the nature and dynamics of the interfaces within the battery cell, which in turn rely on temperature-driven reactions with unpredictable kinetics. Although monitoring temperature is essential for enhancing battery cycle life and longevity, this is not directly measured today at the cell level in running electric vehicles (EV) or in development setups, where the fine-tuning of the later battery pack is developed.

Drastically enhancing battery cell QRL calls for better knowledge/monitoring of the physical parameters during cycling and an understanding of the science beyond the parasitic chemical processes taking place within the battery cells, i.e., fundamental science.

To challenge the existing limitations, we propose a disruptive approach of injecting smart embedded sensing technologies and functionalities into the battery cell, capable of performing spatial and time-resolved monitoring (see Figure 13), so that battery will no longer simply be a black box.[17]

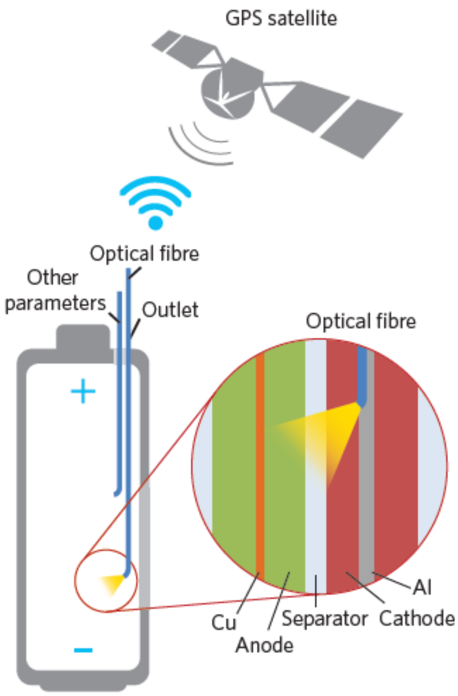


Figure 13. A future battery with an output analyser connected to sensor (optical fibres, wires, etc.) in addition to the classical positive and negative electrodes.

The long-term goal is that the 2030+ battery will no longer be simply a black box. This vision needs to be addressed hierarchically at both component and full system levels. Injecting smart functionalities into the battery cell can be done in several ways. It involves the possible integration and development of various sensing technologies to transmit information in and out of the cells. Sensors that can measure multiple parameters at various locations within a cell (i.e. spatially resolved monitoring) are especially important. Parameters such as temperature (T), pressure (P), strain (ε), electrolyte composition, electrode breathing (ΔV), and heat flow, measured with high sensitivity, would be valuable options.

The introduction of fluorescence or IR probes with optical read-out for the identification of chemical species is one option. This means that in addition to the classical + and – poles, there would also be an analytical output that can transmit and receive signals. To ensure the successful implementation of such embedded sensors in a practical battery cell, the adaptability of all the sensing technologies must be considered. The target is to probe the battery environment in terms of chemical reactivity and manufacturing constraints, together with adequate processing and transmission of sensing data. Lastly, and of paramount importance, is the need to identify state function estimators and to create the proper algorithms to wisely use the colossal amount of sensing data to develop intelligent responsive battery management systems. This needs to be done in collaboration with the BIG–MAP part of this roadmap.

In this section, we first review the current status of sensors and sensing activities within the battery field to identify the remaining scientific, technological, and systemic challenges. Strategies to alleviate them within the context of Battery 2030+ are discussed and highlighted prior to the presentation of our ten-year roadmap with specific milestones to bring these new concepts to fruition, up to the ultimate goal of creating highly reliable batteries with ultra-high performance and long life. The higher the capacity of a battery cell, the more important it will be to ensure safety and long life.

7.3.1 Current status

Over the years, many fundamental studies have examined different battery chemistries using sophisticated diagnostic tools such as X-ray diffraction, nuclear magnetic resonance (NMR), electron paramagnetic resonance (EPR), and transmission electron microscopy (TEM), which can ideally operate in situ and even in operando as the battery is cycled.[178] Although quite spectacular, these analytical techniques rely on specific equipment and cells and cannot be transferred to analysing commercial cells. In contrast, Li-distribution density and structural effects were recently imaged in 18,650 cells, but the imaging techniques used rely mainly on large-scale facilities with limited access.[179] Notable progress has been made over the years towards instrumental miniaturisation, so that bench-top X-ray diffraction units, scanning electron microscopes, and portable impedance (and even NMR) spectrometers exist, but we are still far from producing the test units needed to monitor batteries in their end applications. The need for a paradigm shift towards monitoring the battery's functional status in real time is still unmet.

Determining the state of charge (SoC) of batteries is a problematic issue nearly as old as the existence of batteries. This has resulted in a wide variety of ingenious monitoring approaches developed over the years, leading to numerous patents covering various sensing technologies (Figure 14). For decades, this sensing research was mainly devoted to Pb-acid technology, to make it more reliable and friendlier to customers. Throughout this period, great advances were made with the implementation of electrochemical impedance spectroscopy (EIS) as an elegant tool to evaluate the evolution of cell resistance upon cycling in Pb-acid batteries, enabling estimation of their state of health (SoH).[180] As such, portable EIS devices have been commercialised and used in the field of transportation, and as back-up units in telecommunications, to identify faulty batteries within a module. Such devices still exist but suffer from their poor reliability (<70%). Overall, SoC monitoring remains highly challenging, and there is currently no accurate solution. Estimation of SoC today relies on a combination of direct measurements such as EIS, resistance, current pulse measurements, coulomb counting, and open circuit voltage-based estimations.

As batteries become increasingly central to our daily lives, there are increasing demands for highly reliable and long-life batteries. This has revitalised battery-sensing activities with the emergence of novel approaches to passively monitoring the effects of temperature, pressure, strain, and ΔV of the SEI dynamic via diverse non-destructive approaches relying on the use of thermocouples, thermistors, pressure gauges, and acoustic probes.

However, most of this sensing activity relies on the use of sensors outside rather than inside the battery cells, limiting the knowledge to macroscopic properties but overlooking internal chemical/physical parameters of prime importance for monitoring battery lifetime. Implantable sensors are accordingly attracting increased interest, with optical sensing being predominant (Figure 14). Recent publications have reported the positive attributes of fibre Bragg grating (FBG) sensors and other sensors for: i) accurately monitoring T, P, and ε upon cycling, ii) imaging cell temperature, and iii) estimating battery SoC without interfering with cell performance. The time has come to move out of the concept mode and solve the remaining challenges if we ever want non-invasive battery sensing to become a reality. Industry needs comparable and traceable reference methods for the assessment of the state of the battery. This should be achieved by developing measurement methods and procedures for advancing the evaluation of SoC and SoH of batteries according to the best metrological protocols (see documentation by EURAMET, the European Association of National Metrology Institutes).[181]

7.3.2 Challenges

Numerous sensing technologies for battery modules and systems have been tried (see Figure 14) and it is outside the scope of this review to list them all; rather, our intent is to highlight the ones with the greatest chances of success at the battery cell level.

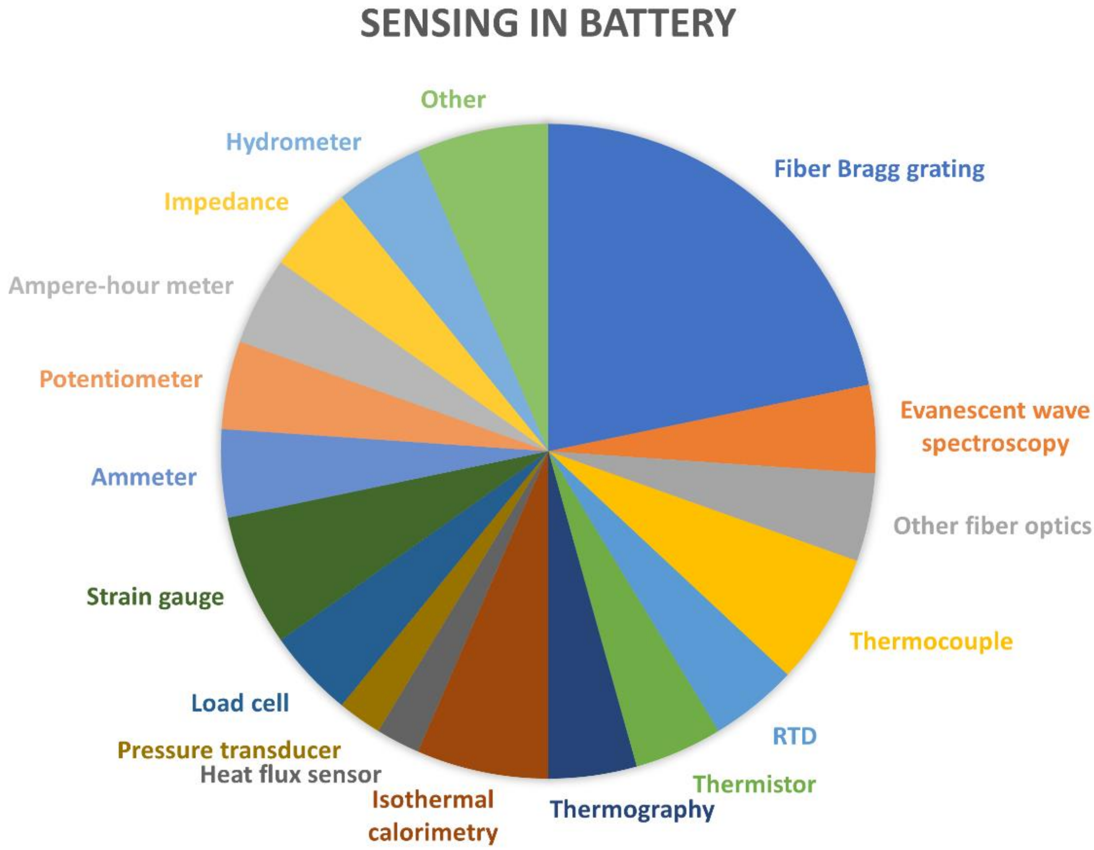


Figure 14. A glance at available sensing technologies for battery modules and systems. The numbers come from the relevant published articles found in the bibliography for each of the techniques in 2019.

Temperature sensors

Knowledge of surface temperature at one location of a battery cell has long been used to validate thermal battery management system (TBMS) models. Temperature sensors fall into four main classes: resistance temperature detectors (RTDs), thermally sensitive resistors (thermistors), thermocouples, and fibre Bragg grating (FBG) optical sensors. These differ in their accuracy and in the convenience with which they can be positioned within the cell. Thermistors, because of their thicknesses (1 mm), are positioned only on the top rather than at the surface of the cell, as opposed to (100 µm) RTDs.[182] Interestingly, longitudinal surface variation in cell temperature during operation has been mapped with an accuracy of ±1°C by screen printing thermal sensor arrays on the surface casing of 18,650 cylindrical cells. However, the scarcity of information regarding the inside of the cell limits the integrity of current TBMS models, calling into question their accuracy and predictive capabilities. Simplified attempts to alleviate this issue have consisted of implanting thermocouples within 18,650 and pouch cells, and the successful electrocardiogram of a 25 Ah battery was realised by embedding 12 thermocouples at specific locations within cells, and 12 additional ones at the same positions but on the surface of the cells.[183] This allowed temperature contours within the cell to be plotted, providing valuable information to validate thermo–electrochemical models. **Drawbacks of this approach reside in the positioning of the various thermocouples and in wiring them without affecting the tightness of the cell and its performance**. A more convenient way to assess temperature contours and identify hot spots within the cell uses infrared thermography, but this technique **suffers from poor spatial resolution together with limited temperature accuracy and susceptibility to background noise**.

Gauge sensors (ε, P)

Besides monitoring temperature, methods to sense intercalation strain and cell pressure are equally critical techniques for monitoring the SEI dynamics that affect the SoC and SoH of batteries. Early experiments have relied on the use of in situ strain gauge measurements to probe, for instance, the total volume change during the charging and discharging of Ni-Cd batteries. This work was extended to the study of commercial Li-ion $LiCoO_2$/C cells, and other cells, to measure the strain associated with phase transition as well as to quantify delays in the cell volume variation as a function of the cycling rate. Recently, using a strain sensor placed at the surface of the cell, Dahn et al. demonstrated that the irreversible volume expansion caused by SEI growth could be detected by in operando pressure measurements in addition to the establishment of a correlation between capacity retention and irreversible pressure increase.[184] The simplicity of such an approach, which relies solely on the use of external sensors, constitutes its advantage. However, placing **strain sensors at the cell surface falls short** in providing spatial information, which is critical for improving SoC and SoH batteries.

Electrochemical sensors

Electrochemical sensors are mainly used to sense battery chemical aspects such as SEI growth, redox shuttle species, and metal dissolution. Recently, Dahn's group has convincingly demonstrated the feasibility of using differential thermal analysis (DTA) as an elegant way to track substantial changes in electrolyte composition as a function of the state of life of the battery.[185] DTA of the entire pouch is envisioned as a non-destructive method to correlate the melting point of the electrolyte with the cell's state of health. **Therefore, it remains an ex situ technique with no chances of miniaturisation or of being used to track batteries in real applications**.

Typically, the electrochemical (voltammetric, amperometric) cell/system used in the laboratory can be viewed as electrochemical sensors for detecting various species, but an inherent drawback for use in battery sensing is miniaturisation issues. This is changing owing to recent advances in the field of biophysics/chemistry, so that electrochemical sensors are now extremely suitable for miniaturisation down to micro or even nano-dimensions using several mechanical, chemical, and electrochemical protocols to prevent environmental artefacts (e.g., convection). The combination of advanced electrochemical (pulse) techniques and unique suitability for electrode/sensor miniaturisation and electrode modification **provides an excellent basis for designing powerful new detection** microsystems that could be conveniently incorporated into batteries provided that remaining material aspects can be resolved.

A persistent challenge in electrochemical battery diagnostics is the development of effective and (electro)chemically stable and durable (quasi-)reference electrodes (RE) that can be used in voltammetric/amperometric and/or potentiometric detection regimes. REs have been of paramount importance in understanding various battery system chemistries at the lab scale, where a few tens of cycles are usually sufficient to unravel failure mechanisms and other limitations. They enable: (i) identification of the distinct contribution of each cell component to overall battery performance; (ii) the correct interpretation of current and voltage data with respect to the components; and (iii) study of the reaction mechanisms of individual electrodes. However, there are difficulties in: (i) having REs of well-selected chemical composition to ensure chemical inertness to the cell environment; and (ii) defining the proper RE geometry and location with respect to the other cell components, which depend on the cell configuration to prevent experimental artefacts. The use of REs for battery sensing **is therefore appealing. However, it must be realised that, as of today, reliable, user-friendly, chemically stable**, long-lasting, and artefact-free cell configurations do not exist. Solutions are waiting to be found.

Optical sensors

Fibre Bragg grating (FBG) sensors, which correlate the wavelength dependence of the emitted signal with local temperature, pressure, and strain, are by far the most studied type of optical sensor. Few research groups have shown how FBG sensors could be used to thermally map a battery pack.[186] Moreover, PARC (a Xerox company) has demonstrated the feasibility of obtaining high-performing Li-ion pouch cells for EV applications with embedded FBG sensors attached to the electrode while not observing major adverse effects of the embedded fibre on the cell life for at least 1000 cycles.[187] Based on the accuracy of the strain measured using FBG sensors, the SoC was estimated with less than 2.5% error under different temperature conditions and under dynamic cycling. As well, the authors could predict the cell capacity up to ten cycles ahead with approximately 2% error. However, a difficulty with FBG use is that is simply decouples pressure and temperature. Thus, the further addition of surface/ambient FBGs together with a thermal model enabled the operando monitoring of heat generated during the cell operation.[188] Furthermore, Rayleigh sensors, unlike FBGs, can provide axial resolution, in addition to being less expensive to manufacture.[189] Nevertheless, they require a more expensive interrogation system and greater calculation resources to analyze the large amount of data generated.

A solution to this decoupling issue has been provided by the arrival **of microstructured optical fibres (MOFs),** also known as photonic crystal fibres (PCFs).[190] Unlike FBG sensors, whose functioning relies on a change in refractive index between core and cladding to obtain total internal reflection of light, MOFs achieve total internal reflection by the manipulation of their waveguide structure, enlisting air holes within the fibre core whose patterning determines the specific properties of MOF sensors. Hence, with careful design of the air-hole pattern, MOFs offer a feasible way to measure temperature and pressure independently with a single fibre. However, MOF fabrication is still in its infancy.

Nano-plasmonic sensing (NPS), introduced to the field of batteries as recently as 2017, has the advantage of focusing, amplifying, and manipulating optical signals via electron oscillations known as surface plasmons (SPs). NPS technology relies on the shift in the wavelength of the plasmon resonance peak, due to a change in the refractive index (RI) of the surrounding medium nearest (<100 nm) the sensor surface. These sensors can then be used for the in operando monitoring of physicochemical phenomena occurring on the nanoscale, such as SEI growth, lithium intercalation/deintercalation, and local ion concentration variations.[191] However, making such sensors requires the deposit of a metallic plasmonic nanostructure on top of the fibre, whose physicochemical stability upon cycling in presence of electrolytes remains undetermined.

Acoustic sensing. Batteries are breathing objects that expand and contract upon cycling, with volume changes as great as 10%. This leads to important mechanical stress (i.e., cracking) inside the battery's materials that can generate acoustic signals. "Listening" to and analysing the elastic acoustic waves generated by battery materials during operation has long been defined as potentially interesting for the study of batteries. The acoustic emission (AE) technique is used to monitor numerous types of battery chemistries (e.g., Pb-acid and Ni-MH), and was more recently implemented in the study of LIBs during the formation stage. However, AE suffers from some important limitations relating to the minimum threshold stress required to generate acoustic waves and to the lack of spatial recognition as a sensing technique. In contrast, AE is very effective for: studying the formatting step of batteries; detecting operation conditions leading to excessive stress on the battery's materials; and detecting the early signs of abnormal behaviour that could lead to safety issues. Such limitations have been partially addressed by measuring the speed of ultrasonic acoustic waves, generated by piezoelectric transducers, propagating through the battery. Using this advance, researchers have exploited the physical properties of the transmitted acoustic signal (e.g., amplitude and frequency distribution) to estimate the SoC of LIBs.[192] **Nevertheless, a remaining limitation of the acoustic interrogation technique is the copious wiring** required to connect the acoustic transducers used for signal emission and reception.

Standardization

The integration use of sensing functionalities in battery cells and packs requires communication between sensors and battery management system (BMS). The establishment of standards for data generation, transmission and interpretation in conjunction with a smart BMS will help enable improved lifetime, safety and faster charging by means of increased process understanding and reproducibility. Standardized protocols will be in place for reporting both data and metadata.

On the next level and on the premise that a comparability of sensor results is guaranteed, the connection to the BMS can be realized: Sensor connectivity and data management with the BMS interface at the cell, module, and pack levels will be integrated while maintaining the compatibility with battery manufacturing processes. This therefore includes a standardized sensor integration process and connection to the BMS.

In the long term, a standardized communication with the BMS and the generation of standardized sensor data for the Battery 2030+ Electronic Lab Notebook is the goal.

Short, medium, and long-term goals on the way to standardization in Sensing and to reach a fully automated extraction of standardized data for communication with the BMS are listed in Table 5.

In summary, the field of battery sensing is moving beyond proof of concept and is becoming crucial to the design and monitoring of smarter batteries. However, for this to happen, we need to master the efficient sensor data processing and the communication between sensors and BMS systems. The communication interfaces must be viewed as an integral part of the sensor, and must be considered during the co-design of sensor and cell. Eventually, sensor information should provoke autonomous reactions of the BMS, based on proven cell and battery models and supported by AI and machine learning approaches. To realise the potential of this fascinating field, advances in both hard- and software are needed. This matter is discussed next, directly linking to the methods developed in the BIG–MAP part of Battery 2030+.

7.3.3 Advances needed to meet the challenges

Our proposed disruptive approach to meeting these challenges is to inject into the battery smart embedded sensing technologies and functionalities **capable of performing the spatially and temporally resolved monitoring of changes detrimental to battery life.** This long-term vision needs to be **addressed hierarchically on both the component and full system levels.**

Injecting smart functionalities into the battery will include the integration and development of various sensing technologies previously used in other research sectors, technologies that rely on optical, electric, thermal, acoustic, or even electrochemical concepts **to transmit information into/out of the cells.** Sensors that can measure with great accuracy multiple parameters such as strain, temperature, pressure, electrolyte concentration, and gas composition and can ultimately access SEI dynamics must be designed/developed. For successful implementation of the sensing tooling in a practical battery, sensors will have to be adapted to the targeted battery environment in terms of (electro-)chemical stability, size, and manufacturing constraints, including recyclability.

The manufacturing constraints also include the consideration of system design trade-offs. The identified sensors have different requirements in terms of signal generation as well as data acquisition and processing. Optical and acoustic sensors require signal generation and dedicated data acquisition electronics, which are ideally positioned directly on the battery cell to avoid wiring. Moreover, these types of sensors required data acquisition in the several kHz or even MHz range, which puts severe constraints on the data communication with the BMS when considering multiple data streams required to support a high spatial resolution. The system design trade-offs include the analysis of local versus central data pre-processing and hardware requirements for associated data transmission volumes together with the overall techno-economic optimization of all required electronic components.

Addressing manufacturing constraints is no doubt important, but an urgently missing gap to achieve this is the lack of expertise on the practical implementation of sensors into cells and electrode-electrolyte components. This expertise exists but outside the battery community. Let's recall for instance that either optical sensing relying on FBG's, LPG's are commonly used in civil engineering for health structure monitoring (bridges, buildings, etc.). For instance, the insertion of FBG's into composites is an inherent part of the processes used for assembling H_2 storage cylinders with the sensors being wisely wired to monitor cracks. So rather than to reinvent sensor integrations, it will be wiser to set-up open calls to attract the sensing industrial community. This is CRUCIAL for Battery 2030+ success in battery sensing. As of today, real opportunity of optical sensing multiplexing was demonstrated to access several metrics with a single fiber, hence minimizing the wiring.

For sensors, there are two successive steps: first the integration of existing sensors followed by the development and the integration of new specific sensors dedicated to the battery. In both cases, it is important to ensure the metrological traceability of these sensors with regards to primary references in order to ensure comparable measurements and hence more meaningful experiments (see documentation by EURAMET, the European Association of National Metrology Institutes).[181]

Owing to the harsh chemical nature of the battery environment, we need to develop sensors with innovative chemical coatings having extremely high chemical and thermal stability. Equally, the integration/injection of sensors in the battery will necessitate reducing their size to a few microns, so they can fit into the thickness of electrode separators and hence not affect cell performance. In terms of manufacturing, a pressing goal is to make sensors an integral part of the battery, not simply an addition. Different strategies can be applied; for example, as has been done for thermistors, printing processes for sensor fabrication would create new opportunities for the integration of sensors both outside and inside battery cells as well as on battery components for in situ measurements. Such new avenues will have to be explored in conjunction with Battery 2030+ manufacturing and recyclability activities. Moreover, an ultimate challenge is to develop adequate data transmission concepts to bypass the connectivity issues associated with implementing today’s sensors and that are adapted to the noisy electromagnetic environment of the battery. It must be realised that adding wires to the cell could make manufacturing so expensive that it would outweigh sensor benefits. Besides wireless communication, the use of power line communication that employs the existing current-carrying wires, could be an alternative. Another route towards less wiring could consist of the development of novel sensors capable of monitoring several parameters at once, for instance, coupling FBG, MOF, and NPS functions on a single sensor while not interfering with cell performance. Similarly, different Bragg gratings could be inscribed into the same fibre to allow for so-called multiplexed measurements. Distributed sensing as offered by MOFs could be a possible solution as well, if we master their design. Lastly, cells must be used to develop sensing concepts, anticipating that findings could be implemented in modules and battery packs.

To enable the commercial success of advanced sensor concepts, the economic benefits must be demonstrated. The addition of new sensors and required associated electronics comes with upfront investments, which are potential showstoppers for many of the very often price sensitive battery applications. On the other hand, the exploitation of these sensors can lead to significant performance enhancements, like a significantly extended lifetime or the ability to provide more accurate SoC or SoH estimates, which represents an economic advantage throughout the entire lifetime. It is important to identify, define and determine suitable economic performance indicators to evaluate a cost-versus-benefit analysis to motivate the uptake of the technology by the industry and end users.

To ensure societal impact, our approach must be systematic and include the tripartite connection among battery pack, BMS, and application. Sensing will provide a colossal amount of data, which is a blessing for AI. Wise incorporation of this data into the BMS is must also be considered. Obviously, this aspect will greatly benefit from the AI pillar of Battery 2030+, so that transversal efforts are being planned and will be highly encouraged in developing sophisticated BMS and TBMS systems based on the synergy between AI and sensing. Nevertheless, for this integration of data into the BMS as well as to the ensure an efficient link with the AI, standardization is needed. This includes the need for standards regarding data transmission between sensors and the AI, more specifically using standard data formats as well as the use of protocols for data sharing and battery cycling.

7.3.4 Forward vision

Within a ten-year horizon, the development of new sensors with high sensitivity, high accuracy, and low cost offers the possibility of access to a fully operational smart battery. The integration of this new technology at the pack level, with an efficient BMS having an active connection to the self-healing function, is the objective of the Battery 2030+ roadmap. Needless to say, realising this long-term vision of smart batteries includes several research facets with their own fundamental challenges and technological bottlenecks. Among the foreseen milestones are the following:

Achieved in the last three years: The three projects Instabat, Sensibat and Spartacus working on the topic sensing have achieved the first two of the short term goals from the last roadmap. They have applied **non-invasive multi-sensing** approaches relying on various sensing technologies and simple integration that is **transparent to the battery chemical environment** offering spatial and time resolution and they have integrated sensors into existing battery components (e.g., separator, current collector, and electrode composite). The third short term goal was achieved partly. The projects have deployed sensors capable of in vivo access some of the relevant phenomena (e.g., interface dynamics, electrolyte degradation, dendritic growth, metals dissolution, and materials structure change). But there is still work to do to better detect more of the relevant phenomena. Monitor the normal/abnormal evolution of key battery parameters during cell operation and develop proper data processing and transmission concepts to provide the sensing data to the BMS. On the side of sensors, development of specific sensors dedicated to capture the internal phenomenon on the battery cell, stable over a wide range of condition (temperature, pressure, chemical concentration, electric field) has been achieved.

In the short term**:** Define suitable KPI for the techno-economic evaluation of the commercial viability of such concepts. Increase the operational temperature window by >10% through on-the-fly sensing. Some of the medium term goals from the last roadmap are now, with the results of the last three years, in the more reachable future and are moved to new short term goals. Miniaturise and integrate the identified (electro)chemically stable and **multifunction** sensing technologies and associated data processing and transmission electronics at the cell level but also in real battery modules, in a cost-effective way compatible with **industrial manufacturing** processes.

***In the medium term*:** Demonstrate the technical feasibility but also economic viability for selected use cases. Integrate sensor connectivity and data management with the BMS interface at the cell, module, and pack levels while maintaining compatibility with battery manufacturing processes. Standardisation of the sensor integration process and connection to the BMS. Establish new self-adapting and predictive controlled algorithms exploiting sensing data for advanced BMS. Integrate sensing and self-healing in BIG–MAP. Demonstrate the reduction of electrode overvoltage in multivalent systems by >20 %. Increase the accessible voltage window by >10 % in Li-ion batteries. Development of virtual sensors to limiting the number of physical sensors to a minimum. Adaptation of the reliability of the sensor integration. Address challenges on integration, measurement and compatibility of sensors related to new cell technologies (e.g., all solid-state batteries).

***In the long term*:** Master all aspects, including economic trade-offs, of data processing and transmission between sensors and an advanced BMS, also relying on new data-driven AI approaches to achieve a fully operational smart battery pack. Couple sensing/monitoring advances with stimulus-activated local purpose-targeted repair mechanisms, such as self-healing, in future cell-design and chemistry generations to produce smart batteries relying on an integrated sensing–BMS–self-healing system. Fully automated fabrication of smart cell from the sensors to pack integration.

Some additional key challenges in the field of sensing that were identified in the last three years are:

- **Safety**: it is feasible to measure various parameters, such as temperature, voltage, current, and state of charge (SoC), with high sensitivity. But a medium term (or even short-term goal) is the usage of sensor data to forecast very quickly the state of safety. Sensors must be able to detect and monitor these critical conditions in real-time to ensure early warning and preventive measures. This was one of the objectives of the sensor projects, but it was not achieved completely, so far. It can be also linked to the already stabilised short term goal "Deploy sensors capable of detecting various relevant phenomena".

- **Size and Integration**: As lithium batteries are used in various applications, including portable electronics, electric vehicles, and energy storage systems, the size and integration of the sensors become crucial. Developing compact and lightweight sensors that can be seamlessly integrated into battery systems without significantly impacting their overall size or weight is a challenge. This can be also solved using virtual sensors. Beside of the sensors themselves this holds also for the driving electronics and - combined to that - the energy consumption of the sensing devices.

- **Scalability**: linked again to new technologies mentioned by Olivier. As lithium battery technology advances and new chemistries emerge, sensors must be adaptable and scalable to accommodate different battery chemistries, sizes, and configurations. This flexibility is vital to support the diverse requirements of various industries and applications. This calls for sensor designs that are easy adaptable on different type of battery geometries and battery types. Whenever feasible, industrial scaled process solutions should be used to produce the sensors or sensor arrays. In order to ensure the quality and reproducibility, manual processing and integration should be avoided

- **Data Management and Analysis**: The large amount of data generated by battery sensors needs to be effectively managed and analysed to extract meaningful insights. Developing robust data management systems and algorithms that can handle real-time monitoring, data storage, and predictive analysis is a challenge in the context of lithium battery sensors.

7.4 Integration of smart functionalities: Self-healing

The development of substantially improved rechargeable battery cells is a must in the transition towards clean energy and clean mobility.[193–200] Besides the absolute need to develop sustainable batteries, our increasing dependency on batteries calls for great efforts to ensure their quality, reliability, lifetime and safety (QRLS).[201,202] Detection of irreversible changes (sensing) is a first step towards better QRLS[18]. However, to really ensure better quality, higher reliability, prolonged lifetime and improved safety, the cell should be able to automatically sense damage and also to reinstate the virgin configuration together with its entire functionality.[89] That can be obtained either with preventive or curative actions. Preventive actions, part of Battery Interface Genome roadmap (see section 7.2), are addressing degradation processes occurring during the battery operation. Since battery cells are working in different conditions and for various applications, we need curative approaches to obtain higher QRLS. Latter can be obtained by implementation of self-healing functionalities in the battery cell components coupled with a sensing approach. Nature has developed various preventive and curative defence mechanisms as an important survival feature. So a burning question is raised: Can we try to mimic natural healing mechanisms to fabricate smart and long-life batteries?[203] Biological systems offer a great diversity of self-healing processes with different kinetics, such as stopped bleeding (minutes), skin wound healing (days), and repair of broken bones (months). Nevertheless, the desire to accelerate healing time has led to the emergence of a vast and multidisciplinary field in medical science called "regenerative engineering."[204] Implementing self-healing features into batteries by now is a flourishing research field following different examples from the field of material science. Importantly, self-healing approaches to battery systems should be developed for different parts of the battery cell and their functionalities should be synchronized with battery chemistry. Different nonactive components like separators, binders or current collectors can act as a self-healing battery part enabling an intrinsic approach while the same battery components could be a storage place for extrinsic self-healing functionalities.

As in the medical field, which heavily relies on the vectorisation of drugs for the treatment of diseases,[205,206] it will be essential to develop, within the battery, a tool for the on-demand administration of molecules that can solubilise a resistive deposit (e.g., the solid electrolyte interphase layer) or inject self-healing functionalities to restore a faulty electrode within the battery (Figure 15).[207–211] This constitutes another transformational change within the battery community, as nearly nothing has been done to address this topic.

Sensing and self-healing functionalities are intimately linked. Our ultimate vision of smart batteries integrates both these functions. Signals from the sensors will be sent to the BMS and analysed; if problems are determined, the BMS will send a signal to the actuator, triggering the stimulus of the self-healing process. This game-changing approach will maximise QRLS, user confidence, and safety.

This far-reaching goal is not only ambitious but also motivating. Since there is no coherent European research effort addressing battery self-healing (BSH), there is a need to create the relevant research community by linking different disciplines, knowledge types, and practices.

An intimate synergy among sensing/monitoring, BMS, and self-healing will ensure success (see Figure 15), enabling Europe to take worldwide leadership in BSH.

This section attempts to review the current status of self-healing activities within the field of batteries and to identify the associated challenges. The proposed strategies to alleviate these challenges will be presented, as well as the ten-year long-term roadmap.

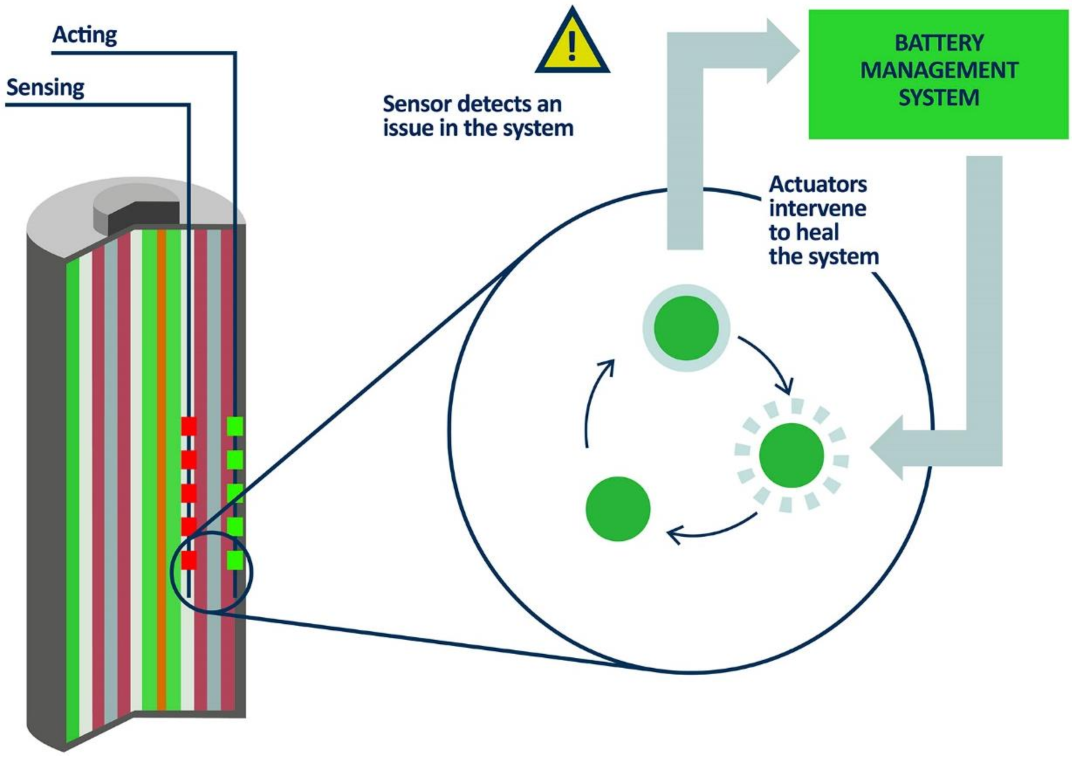


Figure 15. The synergy between sensing, BMS, and self-healing.

7.4.1 Current status

Self-healing mechanisms can be classified either as autonomous, when there is no need for any intentional healing stimulus, or as non-autonomous, when additional external stimulus (e.g., heat, light, and pH) is needed[212]. In both cases the components of the healing process need to be highly reactive to achieve fast and efficient reactions with solid surfaces. For this reason, very few self-healing approaches within the battery field have yet benefited from the general strategies and formalisms well established for human bodies. Copying nature's strategy, i.e., relying on the use of sacrificial weak bonds for self-repair, battery scientists have developed molecules – polymers – with intrinsic self-healing properties based on dynamic supramolecular assembly, such as hydrogen bonding, electrostatic crosslinking, and host–guest or Van der Waals interactions.[213,214]

A lot of different self-healing approaches exist nowadays caused by the huge variety of degradation mechanisms. An overview of the major degradation mechanisms in Li-ion batteries where self-healing can be of great importance is given in Figure 16. Intrinsic and practical problems are related to each degradation route but also give hidden opportunities for the development of unorthodox self-healing innovations in batteries. The interaction of the individual components is very complex and the ageing processes of a chemical, electrochemical and mechanical nature are closely linked.[18]

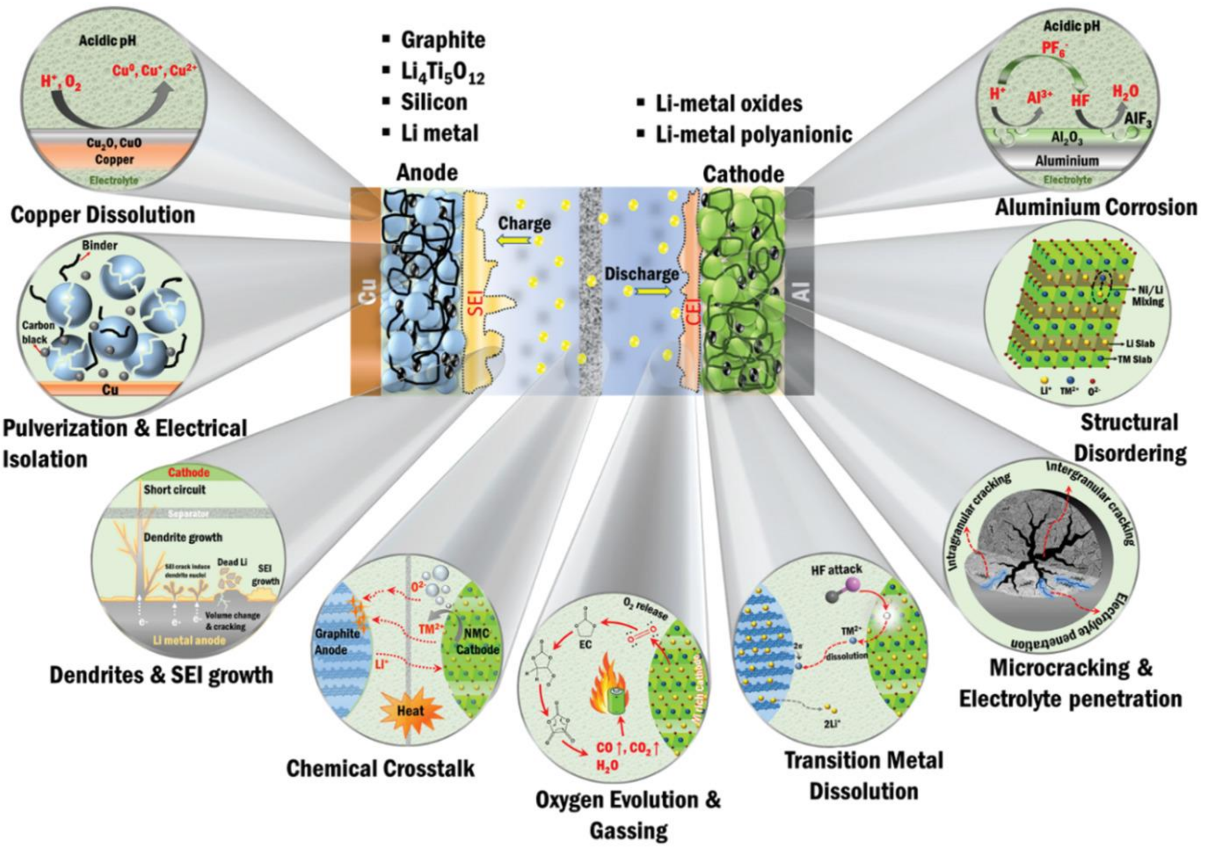


Figure 16. Overview of the major degradation mechanisms in Li-ion batteries.[18]

One self-healing strategy is the development of functionalised and flexible polymers that are chemically compatible with battery components, with reactive species produced in the material in response to damage. Another self-healing approach, so far barely applied in the battery community, uses microcapsules hosting healing species. These need to stay active upon their release, which is triggered by a stimulus[215]. Significant advances have been achieved in the field of thermo-switchable polymers with thermal self-protection integrated into the electrolytes and current collectors.[216–218] A plethora of self-assembling materials [219–222] and bio-inspired mechanisms pertaining to the field of supramolecular chemistry and biology have also been tested to exploit radically new smart functionalities for either intrinsic or extrinsic self-healing processes. Learning from nature, material scientists have developed different approaches such as self-healing fibre-reinforced polymer composites, self-healing coatings, self-healing cementitious materials, self-healing ceramics[18], self-healing organic dyes, self-healing concrete molecules, and many others.[223–225]

The use of self-healing electrolytes is yet another impressive strategy to improve the electrochemical performance and durability of both non-aqueous and aqueous batteries. In a proof of concept, the strategy was used to combat the polysulfide shuttling effect in lithium-sulphur (Li-S) batteries. A self-healing electrolyte system, based on the creation of a dynamic equilibrium between the dissolution and precipitation of lithium polysulfides at the sulphur/electrolyte interface, was successfully developed and enabled high specific capacity and high coulombic efficiency.[226]

Yet other self-healing strategies are developed to minimize formation of dendrites in Li-metal batteries. Among different solutions, Ding et al. used functional metal cation additives like Cs^+ and Rb^+ [227] which enable a sustainable self-healing electrostatic shield (SHES).

Moreover, and specific to batteries, the identified self-repairing chemical tools must be highly resistant to the harsh oxidizing/reducing chemical environment of the cell. This has slowed the introduction of self-healing approaches in the field of energy storage. However, this situation is rapidly changing, as shown by a few recent studies dealing with the incorporation of self-healing functionalities into batteries and super capacitors.[18,228,229]

In conclusion, the field of BSH is rapidly gaining momentum as a part of smart battery design as shown in Figure 17.

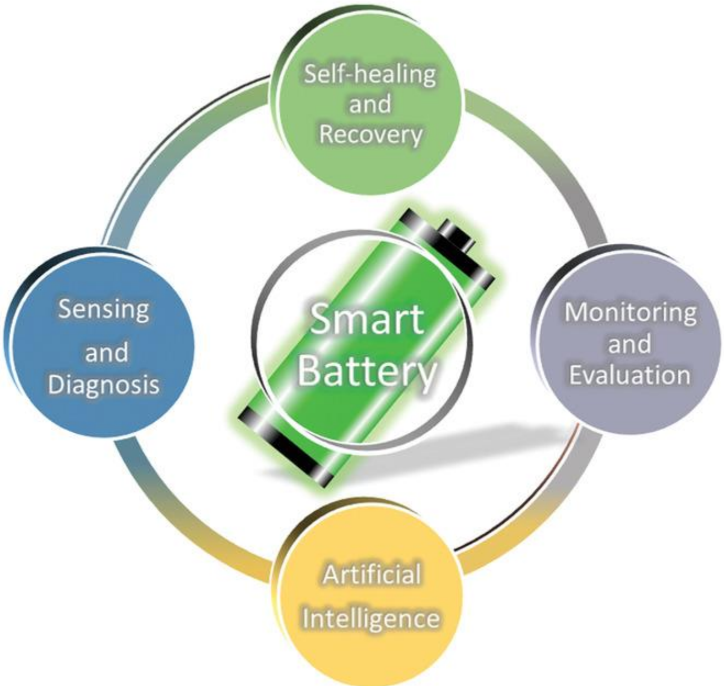


Figure 17. Schematic of self-healing mechanisms in battery material.[18]

7.4.2 Challenges

Self-healing activities within the field of batteries have mainly targeted the auto-repair of electrodes to restore conductivity, as well as functionalising membranes to regulate ion transport or minimise parasitic reactions. Some of these aspects are addressed in more detail below.

Restoration of electrode conductivity

The restoration of electrical properties after damage is of paramount importance in energy storage devices. Great hope is placed in the development of healing systems that use a conductive material that creates physical and electrical integrity between, for example, crack/fracture facets, coating shells, and electrodes/current collectors.

The first studies of the self-healing of conductivity used urea-formaldehyde microcapsules filled with carbon nanotubes (CNTs) dispersed in chlorobenzene or ethyl phenylacetate to provide both mechanical (solvent) and conductivity (CNT) healing. These microcapsules were tested by embedding them in layers of epoxy above and below a glass slide patterned with gold lines. Sample fracturing resulted in conductivity being lost as a crack formed in the gold line. The microcapsules burst when physically damaged, leading to the release of carbon nanotube suspension that restored conductivity within a few minutes (see Figure 18).[215,230]

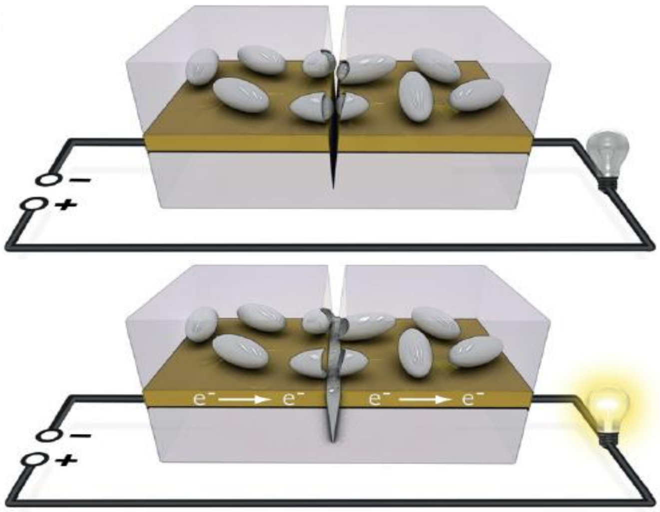


Figure 18. Testing self-healing of the gold line after damage.[215]

Other conductive chemical systems, such as carbon-black (CB) dispersions, were similarly encapsulated and tested.[231,232] These are very attractive since CB is already used as a conductive additive in graphite anodes. Such dispersions in combination with co-encapsulated poly-(3-hexylthiophene) (P3HT) were successfully used to restore conductivity in cracked silicon anodes. This increases the chances of developing a practical silicon anode for LIBs, which are prone to losing integrity because of their nearly 400% volume change during lithiation. Inherent drawbacks of this elegant approach are its irreversibility and the amount of required electrochemically dead microcapsules, penalizing the cell energy density.

Further discussion of Si anodes is merited. Wang's early work reported a polymer coating consisting of a randomly branched hydrogen-bonding polymer (see Figure 19) that exhibited high stretch ability and sustained the mechanical self-healing repeatability that helped the Si anode withstand large volume expansion after many cycles.[214,233,234] An extension of this concept by the same group has led to the design of electrodes with a 3D spatial distribution of the same self-healing polymer into Si anodes to ensure better adhesion, giving high cycling stability.[235] Besides hydrogen-bonded polymers, self-healing binders based on several other supramolecular interactions have also been employed for Si anodes[236–240] and sulphur cathodes.[241] Long-term testing is sorely needed to fully evaluate the practicality of these approaches.

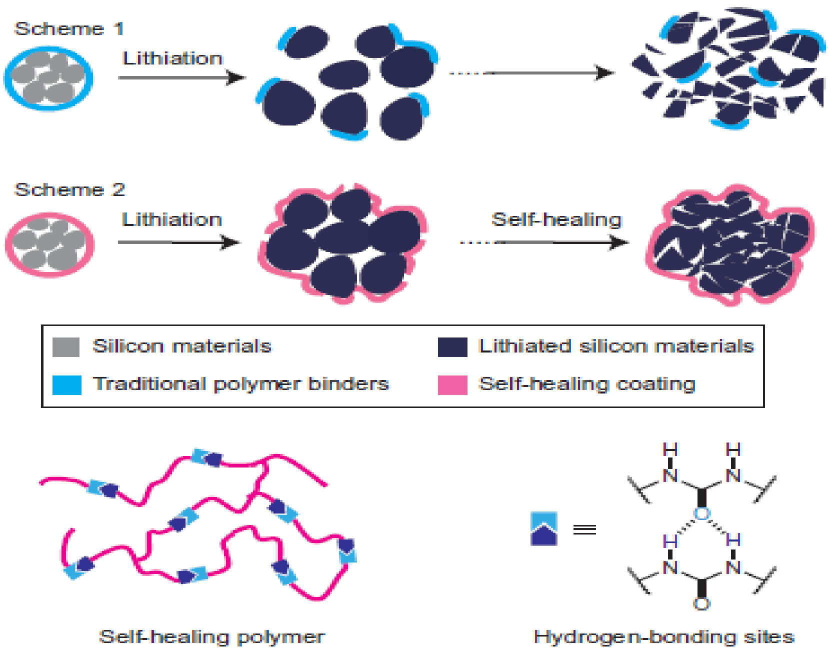


Figure 19. Design and structure of a self-healing silicon electrode.[230]

Another auto-repair concept developed by Deshpande et al.[242] relies on the use of liquid metal (LM) anodes, that is, a metallic alloy (Li_2Ga) having a low melting point so that the reversible liquid–solid–liquid transition of the metallic alloy can be triggered during lithiation/delithiation cycles. Thus, micro-cracks that form within the electrode can be healed during the Li-driven liquid–metal transformation. This approach was subsequently implemented in other Li-alloying negative electrodes as well as in other chemistries. For instance, self-healing Ga-Sn electrodes[243] were shown to have excellent cycling performance (>4000 cycles) and a sustained capacity of 775 mAh g^{-1} at a rate of 200 mA g^{-1}. Self-healing alloys (Na-Sn) were also implemented by Mao et al.[244] to improve Na-ion batteries.

Apart from batteries, an electrically and mechanically self-healing supercapacitor has been demonstrated. Its conductive electrode was fabricated by spreading a TiO2-functionalised single-walled carbon nanotube (SWCNT) film onto a self-healing polymer substrate consisting of a supramolecular network of H-bond donors and acceptors. The CNT contacts broken after damage were repaired by the lateral movement of the underlying self-healing polymer, thereby restoring the electrode configuration and electrical conductivity.[245] Specific capacitances of 140 F g^{-1} could be achieved with the feasibility of 92% recovery after several breaking/self-healing cycles. Interestingly, the self-healing insulator polymer widely used in these studies is based on the one reported by Cordier in 2008,[221] prepared by the supramolecular cross-linking of fatty dimer acids with urea. This polymer has often been the material of choice, as it functions without the need of any external stimulus while recovering repeatedly from several hundred percent of extensibility.

Supramolecular interactions frequently involve H bonding. This is not ideal for the design of self-healing binders for non-aqueous battery systems due to parasitic reactions involving hydroxyl groups. This constraint is no longer present in Li-based aqueous batteries. This was exploited by Zhao et al., who demonstrated a new family of all-solid-state, flexible, and self-healing aqueous LIBs using aligned CNT sheets loaded with $LiMn_2O_4$ and $LiTi_2(PO_4)_3$ nanoparticles on a self-healing polymer substrate[246]. The assembled aqueous LIB, once cut, could be healed in a few seconds by simply bringing the two parts back into contact. Similarly, a new-generation self-healing zinc-iodine flow battery (ZIFB), which consists of a porous membrane that can absorb I_3^-, was reported by Xie et al.;[247] from the working group of Prof. Li. By overcharging the cell, the I_3^- contained in the membrane oxidizes the zinc dendrite so that the battery self-recovers.

Designing self-healing electrolytes

The use of self-healing electrolytes is yet another impressive strategy to improve the electrochemical performance and durability of both non-aqueous and aqueous batteries. In a proof of concept, the strategy was used to combat the polysulfide shuttling effect in lithium-sulphur (Li-S) batteries. A self-healing electrolyte system, based on the creation of a dynamic equilibrium between the dissolution and precipitation of lithium polysulfides at the sulphur/electrolyte interface, was successfully developed with a sustained capacity of 1450 mAh g^{-1} and high coulombic efficiency.[248] To further improve the efficiency of Li-S batteries, Peng et al.[249], designed self-healing electrolytes (SHEs) preloaded with polysulfides and containing auto-repairing agents so as to mimic fibrinolysis, a biological process occurring within blood vessels. Through this process, the additive agent solubilises solid Li_2S, enabling its subsequent participation in electrochemical cycles. Li-S batteries with an optimised capacity could thereby be cycled more than 2000 times.

The self-healing functionalities can also be added to the ionic liquids. However, this requires chemical treatment to open up the organic rings resulting in polymerizable ionic liquids (PILs).[250] After that, self-healing functionality could be integrated in the polymer chains. The approach undoubtedly paves the way for the polymer and flexible battery technology. Thanks to the self-healing behaviour, PILs can found themselves in the wearable electronics.[251] On the other hand, PILs suffer from poor ionic conductivity, high electrode-electrolyte interfacial resistance. Therefore, the prototype cells are operable at elevated temperatures or via prewetting with electrolytes.[252,253] Furthermore, battery performance is highly dependent on the type of separator material used which concerns the sustainability.[254]

Lastly, dealing with aqueous zinc-ion batteries (ZIBs), Huang et al. designed, via the facile freeze/thaw fabrication of poly(vinyl) alcohol/zinc trifluoromethane sulfonate ($(PVA/Zn(CF_3SO_3)_2$), a hydrogel electrolyte that can autonomously self-heal by hydrogen bonding without any external stimulus[255]. By incorporating the cathode, separator, and anode into a hydrogel electrolyte matrix during the freezing/thawing process of converting the liquid to hydrogel, they demonstrated the assembly of ZIBs that display full electrochemical performance recovery even after several cutting/healing cycles. This approach offers broad prospects for fabricating various self-healing batteries for use as sustainable energy storage devices in wearable electronics.

Other self-healing strategies

Self-healing tools, consisting of a thin TiO_2@Si yolk–shell structure with self-healing artificial SEI + natural SEI, were also designed by Jin et al.[256]. When the TiO_2@Si yolk–shell structure became cracked, internal electrolyte was expelled due to the volume expansion of silicon during lithiation. This ensured contact between the silicon core and the TiO_2 shell covered with the artificial SEI. As a result, fresh natural SEI formed on the surfaces of both the silicon and the TiO_2 shell to connect and repair the cracks. With such a trick, coulombic efficiency exceeding 99.9% and excellent cycling stability were demonstrated.

Dendrite growth has long been a problem preventing the development of non-aqueous Li metal batteries, and stands out as a technological block to the development of today's solid-state Li batteries. Interestingly, Li et al. from the working group of Prof. Koratkar succeeded in achieving substantial self-healing of the dendrites by using a high plating and stripping current (~9 mAh cm^{-2}).[257] With a high current, they could trigger extensive surface migration of Li that smoothed the lithium metal surface, ensuring the homogeneous current distribution needed to prevent dendrite growth. Using repeated doses of high-current-density healing led to lithium-sulphur batteries containing 0.1M $LiNO_3$ that cycled with high coulombic efficiency.

This brief literature review highlights that the battery community is becoming aware of the benefits that self-healing could bring to the field in terms of performance and reliability. Although this field is still in its infancy, the reviewed studies have established a basis for new research trends while stimulating novel and exciting research activities leading towards BSH. Most of the reported auto-repair demonstrations are fundamentally elegant and appealing but far from practical. Such a fundamental–applied gap must be closed, and this poses numerous challenges calling for innovative research and technological development.

7.4.3 Advances needed to meet the challenges

Redox reactions occurring during battery operation are frequently accompanied by additional reactions at the thermodynamically unfavourable interface that release degradation products (i.e., dissolved transition metals or organic species from electrolyte degradation). These released metals or organic species can pass through the membrane and deposit on the anode surface or trigger the shuttling self-discharge mechanism. Therefore, it would be advantageous to functionalise the separator by anchoring to its surface chelating agents that could capture dissolved transition metal ions before they are reduced on the anode surface. Another option would be to graft proteins on the membrane to regulate the migration of parasitic organic species.

Functionalised membrane

The use of separators for grafting/anchoring to trap molecules inside their porous channels is attractive for several reasons. 1) The dissolved TM ions are transported due to diffusion and migration through the separator, rendering them available for capture by the anchored trapping molecules. 2) The porosity of the separator facilitates a high specific surface area for the deposition of an optimised number of traps per volume. The high number of ion cavity sites will increase the probability of ion capture, increasing the number of ions that can be captured per unit of volume. 3) The trapped molecules anchored inside the porous separators are far enough from the sites of electrochemical reactions that they are protected from negative/positive potentials that might affect their stability. 4) The separator provides an ideal host on which to graft molecules, which can take up ions at room temperature. 5) Last, the separator can be specifically designed with self-healing properties, like those of electrodes.

Among candidates for the functionalisation of separators, cyclodextrins turn out to be very promising due to their high solubility, lipophilic inner cavities, hydrophilic outer surfaces, bioavailability, and specific recognition ability for small guest molecules/cations, enabling them to form inclusion complexes. Moreover, specific to such cyclodextrin trapping is its temperature dependence – hence, the feasibility of using temperature as a stimulus for the uptake or release of trapped species on demand. Another option, although less environmentally sustainable, is the use of crown ethers or calixarenes whose highly open structure allows the anchoring of a variety of chelating ligands capable of regulating ion transport without risk of blockade. Moreover, the procedure for grafting them is not too complex. Implementing such concepts for the design of smart separators would be new and exciting.

Polymer membranes

Polymer membranes are being considered as solid polymer electrolytes and are also under study as electrode redox active materials or components of hybrid solid-state electrolytes. Even metal-coated polymeric current collectors are offered commercially. Since polymers can be formed or cross-linked in situ, they can be used as mechanical healing agents within the battery cell, similarly to epoxy or cyanoacrylate (i.e., super glue) resins. Moreover, they can act as a template for inorganic capsule formation on a medium time scale. With the use of composite components, the use of polymers in batteries is virtually unlimited, allowing for the development of self-healing strategies for most components and interfaces based on self-healing polymers. Polymers accordingly constitute the cornerstone of Battery 2030+ self-healing strategies.

Supramolecular assembly may offer a unique basis in the short term for addressing daunting challenges such as preventing the rapid decomposition of organic electrolytes, or liberating conductive self-healing materials for repairing electrodes and interfaces. Hydrogen bonding is the technique of choice to realise these possibilities, and could be used for battery components that can accommodate protic organic compounds. Similarly, ionomers can be non-covalently assembled by forming metal complexes between chains incorporating ionic chelating groups. Reversible covalent bonding (S-S) can also be used in place of non-covalent interactions, but this requires further work. Lastly, the exploration of multiphasic solid polymer electrolyte systems could also allow the application of different self-healing strategies whenever a stimulus can induce the mixing of domains.

Bio-sourced membrane

Another challenge is mimicking biological membranes in terms of their barrier selectivity, to control the decomposition of electrolytes so as to improve battery ageing. A key milestone will be to monitor, inside the battery, electrolyte stability using a sensitive and selective sensor at the single-molecule scale using nanopore technology with electrical detection. For this to happen, one must design thin and porous controlled membranes using the chemistry of non-toxic and bio-sourced molecules/proteins (e.g., cyclodextrins) whose selectivity can be achieved by the use and optimisation of protein engineering.

Self-healing electrodes

The restoration of electrical properties after electrode damage is crucial in energy storage devices. As for membranes, sliding gels made of reversible bonds could be used to control the organisation of the surface and to optimise the efficiency of the battery device. The main advantage of sliding gels in addition to their supramolecular interactions is the pulley effect along the polymer chain to absorb stress, permitting the reorganisation of the chain architecture to return it to its initial properties. We can also use this gel as a reinforcing mechanical bandage, hence our eagerness to explore this path. Another option to explore is based on the building of composite electrodes containing microcapsules capable of releasing healing agents with the application of a stimulus, as is done in medicine with the vectorisation of encapsulated medicines. Designing microcapsules with a mineral or polymeric shell, hosting Li(Na)-based sacrificial salts or other compounds that are released upon shell breaking due to a stimulus, is also worth exploration

7.4.4 Forward vision

The complexity of different degradation processes requires a multilevel approach with vectorization of the extrinsic and intrinsic self-healing functionalities developed for specific battery chemistries. Many nonactive battery components like the separator or the binder are ideal places to store microcapsules filled with sacrificial salt or additives which can substitute lithium deficiency or dissolve a resistive passive film, etc. Such extrinsic self-healing functionalities should be sensitive to temperature, volume, or pressure change and appropriate stimulus (triggering act) should be used for their activation. The development and implementation of on-demand (extrinsic) self-healing calls for the productive coupling of the sensing and self-healing programmes within Battery 2030+. We hope that the use of stimuli for on-demand self-healing will open up a wide range of possibilities for realising in vivo surgical intervention in batteries.

The sustainability of the batteries can be improved with the introduction of bio-sourced materials, which should be developed together with self-healing functionalities. Three different groups of bio-sourced polymers – proteins, polysaccharides, and polyesters or their derivatives and blends with other polymers – offer enormous possibilities for modification of different battery components. For instance, natural polymers can be modified to support battery self-healing functionalities, including controlled transport of cations, maintenance of electrode integrity and the possibility to capture degradation products by using scavenger or chelating molecules. However, mimicking biological membranes in terms of barrier selectivity in order to improve battery ageing or monitoring the stability of the electrolyte via highly sensitive and selective sensors on the biomimetic separator pose tough challenges. The functionalization of macrocycle cages such as cyclodextrins or calixarene on PET membrane separators or similar supports could improve the capture of parasitic redox species.

Lastly, it is important to point out that batteries should truly benefit from self-healing functionalities to the maximum extent. From that perspective, self-healing kinetics will become an important focus. We must be bold and open-minded to tackle the aforementioned challenges while constantly keeping in mind battery constraints in terms of their desired cell performance, the chemical environment within the cell, and the manufacturing process.

Today, European research effort to explore BSH is in progress and a certain know-how was developed considering the emerging opportunities that could give Europe worldwide leadership. Although the research TRL is still low, interest and involvement from the European Industry is remarkable. Accordingly, in order to valorise the developments in the lab-scale, a solid IPR and commercialization pathway needs to be established. This is what the Battery 2030+ programme is targeting, by putting together an ambitious BSH roadmap that will lead to a game-changing approach to maximising battery QRL and serving as a driver reuniting a multidisciplinary community that shares the dream of developing long-lasting batteries with self-healing functionalities. A few milestones towards realising such ambitious vision are listed below.

Achieved in the last three years: A selection of two EU projects (HIDDEN and BAT4EVER) were a first important step in establishing the new research community of developing self-healing functionalities for batteries. Two projects are covering some of the proposed functionalities and the community should continue enlarging.

***In the short term*:** However, some functionalities like design of functional membranes that can regulate ion transport to minimize parasitic reactions have not been established. Due to the nature of the projects, work is covering selected examples, but in the future further exploration of implementation of bio-sourced polymers that can mimic processes from life science and use of selective nanopores should be explored together with more extrinsic self-healing functionalities. Autonomous and on demand based self-healing functionalities were started with certain battery chemistries in the model systems. Yet, long term cyclability expectations are not met. Very initial results are coming up from the ongoing projects. Thus, it’s important to note that a clear pathway for commercialization should soon be established.

***In the medium term*:** Demonstrate wisely engineered separators with capsules holding organic/inorganic healing agents with various functionalities that can be triggered to auto-repair by a magnetic, thermal, or electric stimulus while being electrochemically transparent. In this aspect, self-healing functionalities with external stimulus were already addressed in the Proof-of-Concept level. On-demand temperature control by BMS don’t have a maturity at this stage although activities are recently started. However, there is a lack of chemistry-neutral methodology. Determine the response time associated with stimulus-actuated self-healing actions to repair failures pertaining to electrode fracturing or SEI coarsening.

***In the long term*:** Novel cell design concepts shouldn’t be excluded in the long term perspective. In this sense, alternative designs such as bipolar and/or blade cell designs could open different horizons. Accordingly, manufacturing lines need to be adapted. Establish efficient feedback loops between cell sensing and BMS to appropriately trigger, by means of external stimulus, the self-healing functions already implanted in the cell. Novel design of batteries with materials having accelerated self-healing kinetics. Upscaling of self-healing batteries should be demonstrated, including a cost-benefit estimation.

7.5 Cross-cutting area: Manufacturability

Battery cell manufacturing is at a crossroads. There is a need to scale-up production to efficiently produce billions of battery cells within just a few years. This must be done in a way that is economically and environmentally sustainable, whilst also remaining flexible enough to adapt to improvements in cell design or materials chemistry. This is a challenging – but achievable – task. This perspective will focus on the **manufacturability of battery cells**, their components, and materials. Focus is placed on the synthesis of innovative/breakthrough materials and on the interfaces created inside the battery in the manufacturing process.

Novel methods of manufacturing battery cells[10] are addressed in this roadmap from the perspective of digitalisation, where the coupling of both digital twin of a cell and digital twin of cell manufacturing is seen as an enabler to accelerate and optimize the manufacturability of new cell architectures, new and novel discovered materials as well as integration of new self-healing, and sensorisation functionalities.[258] The power of computational modelling and of Artificial Intelligence (AI) in particular, will be exploited to deliver digital twins both for innovative, breakthrough cell geometries and for both current and advanced manufacturing routes, avoiding or substantially minimising classical trial-and-error approaches. Fully digital product and manufacturing analogues will allow the understanding and optimisation of parameters and of their impact on the final product. These virtual representations will be used to actuate the physical assets supporting greater control of battery manufacturing facilities and production lines.

In addition, to facilitate the connection between BIG-MAP and manufacturability, interfaces need to be created between the different research areas that allow for an efficient exchange of FAIR[43] data and metadata. The development of infrastructure as well as ontologies, protocols and standards will play a key role.

Eco-design criteria, including design to allow easy disassembly for the recycling of cell components or materials, will be facilitated at both the cell design and manufacturing levels. Here, jointly used protocols and standards can play an important role to assess and ensure sustainable processes.

7.5.1 Current status

Among the existing energy storage technologies, Lithium-ion batteries (LIBs) are the most versatile and have unmatched energy density technologies for myriad applications.[57,259–262] Because LIB raw material deposits are unevenly distributed and subject to price fluctuations, current needs in certain applications have put unprecedented pressure on the LIBs value chain. As a result, there is an enormous demand for alternative energy storage chemistries that can replace LIBs. Sodium-ion batteries (SIB) have come into prominence recently due to their promising cost, safety, sustainability and performance benefits compared to LIBs.[263] SIB can also be produced on the same manufacturing lines used for LIB. Safe batteries are also a necessity to accelerate the deployment of EVs. In this regard, solid-state batteries (SSBs) are increasingly seen as very promising next-generation battery systems. The lithium-sulphur battery (LSB) has long been a research hotspot due to its high theoretical specific capacity, low cost and non-toxicity. However, there are still some challenges that impede the practical application of the LSBs. Other commercial battery technologies exist as well (e.g., lead acid, redox flow, Na-S)[264], with new technologies yet under development,[265] but, for clarity and conciseness, we will generally cite LIBs as a reference. The reader is advised to keep in mind that these differences exist, and that current LIB design and manufacturing concepts do not necessarily represent the whole picture for other present or future battery technologies, though they may share some general principles regarding manufacturing challenges.

Cell design

Today, most cell designs are based on three main formats: cylindrical, pouch, and prismatic. In detail, these geometries are based on certain widely accepted sizes (e.g., 21700 and PHEV-2) or engineered according to the application. For given cell designs, iterative improvements (e.g., in stack pressure, number of passive components) ensure steadily increasing energy densities and quality. Despite advancements in cell designs, the fundamental comprehension of structures of electrode materials remains unsolved. Therefore, acquiring a profound understanding of the physical and electrochemical processes at the microscale, as emphasized in the BIG MAP chapter, becomes essential for rationalizing the strategy of microstructural development.

Recent trends include the development of large-format battery cells like 4680 cylindrical cells or blade-type prismatic cells. The driving force behind these designs includes aspects like improved heat management, better packing efficiency, and faster rate performance. But reduced manufacturing costs are also an important motivating factor. The manufacturing of larger cells offers several advantages, such as reduced overall cell count, decreased equipment requirements, and improved production efficiency. However, this approach also introduces greater potential for heterogeneous degradation mechanisms, which may lead to poorer performance. To address these challenges, it is believed that a comprehensive understanding of the multi-scale multiphysics phenomena occurring within the cells, facilitated by modelling tools, is crucial. Such understanding will pave the way for advancements in future cell designs, enabling the exploration of new configurations for each component of the cell for improved overall performance.

Models of battery cells and their components are becoming a cornerstone of the cell design and optimisation process. They play a vital role in understanding cell behaviour, performance characteristics, and identifying areas for improvement. Furthermore, digital twins of cells are emerging as a central element in cell and pack control systems. In this context, a digital twin refers to a virtual representation of an actual battery cell that is linked to the physical counterpart, enabling real-time monitoring and control.

Currently, physics-based models are widely used for optimizing battery electrode and cell performance. Significant efforts have been devoted to developing methods that enhance the microstructure of electrodes. These efforts aim to understand the impact of nanoscale to mesoscale inhomogeneities on the durability of LIBs.[266,267] One prevalent approach is the use of microstructure-resolved physics-based models, which reconstruct the electrode microstructure to examine the interplay between microstructure and electrochemical performance, shedding light on the influence of heterogeneity on electrochemical state variables such as charge transfer, electrolyte concentration, and electrochemical potential at both global and local scales.[268] Regarding cell design optimization, the well-known approach in physics-based models relies on a simplified continuum approach pioneered by Newman and colleagues. This approach is typically implemented on a pseudo-two-dimensional (p2D) simplified mesh.[269,270] Recent advancements have significantly improved computational power, enabling the extension of this approach to pseudo-four-dimensional (p4D) meshes.[271] The p4D approach allows designers to consider the effects of cell design features such as tabs and electrode overhangs, as well as heterogeneous aging phenomena throughout the entire cell.

The current focus is on establishing connections between the parameters of these models and the controllable parameters in the manufacturing process. This linkage is crucial in enabling practical optimization workflows to optimize the manufacturing process and achieve desired outcomes. Furthermore, 4D-resolved finite element method models using electrode microstructures arising from manufacturing simulations have been also reported linking the heterogeneities of lithiation/delithiation upon cycling as a function of the manufacturing parameters (e.g., formulation, calendering degree).[272–274] Although these models are not yet connected to real-time cell operation, they hold potential as key components for future developments of digital twins of cells. By integrating them into real-time monitoring and control systems, digital twins will provide valuable insights for optimizing cell performance and enhancing overall battery management.

The challenges associated with the parameter identification process for building accurate physics-based model, have led to a growing interest in purely data-driven or machine-learning (ML) methods.[275] These approaches focus on creating models directly from observations rather than relying on a deep understanding of the underlying physical principles. Data-driven methods have gained popularity due to their ability to capture complex relationships and patterns in the data without explicit knowledge of the underlying physics. ML methods have been predominantly applied to estimate State of Health (SoH), Remaining Useful Life (RUL), End of Life (EOL),[276] and other related parameters. Although there have been a few exceptions significant emphasis has been placed recently on early prediction of cell lifetime,[121] but the amount of data required for training ML models can be a significant challenge.

The overarching trend is to construct models that are both efficient and accurate. One approach is the utilization of hybrid models that combine physics-based and data-driven techniques. These models leverage the strengths of both approaches, incorporating the physical understanding while incorporating data-driven capabilities to capture complex behaviors. Another promising avenue is the use of Physics-Informed Neural Networks (PINNs). These models integrate physical laws and constraints into neural network architectures, allowing for the incorporation of prior knowledge and governing equations. PINNs have shown promising results in accurately predicting battery performance and safety, offering an efficient and accurate modeling solution. By employing hybrid models and PINNs,[277] researchers aim to strike a balance between accuracy and efficiency, enabling the development of robust models that can effectively capture the intricacies of battery cells and provide accurate predictions.

These advancements are expected to have a significant impact on various aspects, including lifetime prediction, performance optimization, and overall battery management.

By integrating data from sensors, simulations, and other sources, the digital twin will provide continuous and accurate information about the cell's performance and state. This real-time monitoring capability will allow for prompt detection of anomalies, early identification of potential issues, and proactive maintenance strategies. Additionally, the digital twin facilitates control and optimization of the cell's operation. By simulating different operating scenarios and applying advanced control algorithms, it becomes possible to explore optimal strategies for maximizing performance, efficiency, and durability.

Overall, digital twins of cells will empower manufacturers to closely manage the performance of battery cells, as they provide a comprehensive understanding of the cell's behavior, enabling informed decision-making, predictive maintenance, and enhanced control strategies.

Battery manufacturing

Battery manufacturing, especially for LIBs, has become a well-established process. The manufacturing routes for LIB cells[10] can be broadly categorized into three stages: electrode production, cell assembly, and cell finishing.[278] Electrodes are usually manufactured by organic-solvent-based roll-to-roll casting of the slurry onto a metallic current collector, followed by a drying and calendering step to compress them to the desired thickness.[279] Hereby coating and drying are the most cost-intensive processes.[280] In this regard, there is a growing interest in the aqueous processing in the scientific community but also exploring new processing methods.[281]

The emerging dry coating technique has garnered significant attention and demonstrated promising prospects for the battery industry as a new path towards sustainability.[282,283] Until now, several other representative methods have been employed to realize solvent-free concepts in battery electrode manufacturing. These methods include pulsed laser,[284] sputtering deposition[285,286] and extrusion. Nevertheless, while solvent-free concepts in battery electrode manufacturing show great promise, further studies and research are required before they can be effectively scaled up and applied in industrial settings.

During the cell assembly phase of battery manufacturing, critical steps such as stacking and electrolyte filling take place. These steps are considered time-consuming and economically significant due to their impact on the overall production process. After cell assembly, the cell enters the cell finishing phase, where formation step takes place. This step is held in controlled chambers and is cost-intensive, affected by factors such as variations in material quality, manufacturing parameters, and cell design, but necessary to improve battery performance and lifetime.[287]

The digital twin for cell manufacturing is a powerful tool that leverages data collection, processing, and integration with developed models to create a virtual replica of the battery production process and machines. It enables real-time decision-making to enhance product quality, production efficiency, and control strategies. The digital twin comprises four core elements:

a) **Data:** referring to large amounts of data from various sources within the manufacturing process. This includes data from sensors, equipment, and other relevant sources, collected real-time.

b) **Models:** developed based on the understanding of the underlying physics and operational aspects of the production system. They enable simulation, prediction, and optimization of various parameters, allowing manufacturers to make informed decisions.

c) **Infrastructure:** includes hardware resources, such as servers and storage systems, as well as software platforms and tools for data management, analytics, and visualization.

d) **Communication Protocols**: enable the exchange of data and information between the virtual twin and the real-world equipment and systems.

By integrating data, models, infrastructure, and communication protocols, the digital twin for cell manufacturing will provide manufacturers with a comprehensive tool for optimizing production efficiency, quality, and control strategies. It will also enable real-time monitoring, analysis, and decision-making, leading to improved productivity, reduced costs, and enhanced overall performance in battery cell manufacturing.[288]

As for cell manufacturing models, there is a significant amount of relevant work on physics-based and machine learning modelling the main steps of the LIB manufacturing process: Analytical models have been proposed to estimate the rheological properties of the slurries,[289] while bi-dimensional Monte Carlo (MC)[290–294] and Brownian Dynamics (BD)[295–298] have been proposed to understand the particle suspensions in the slurries. 3D-resolved Coarse Grained Molecular Dynamics have been reported to predict the influence of formulation, solid content and active material particle size distribution on the slurry microstructure.[299] Homogeneous Computational Fluid Dynamics (CFD)-based models[300] have been reported to simulate the drying process of electrode coatings to analyse the potential migration of the binder as the solvent evaporates. Discrete Element Method (DEM) models have been reported to understand the influence of calendering parameters on electrode microstructure evolution, either by resolving only the active material spatial distribution,[301] or by explicitly considering both active material and carbon-binder.[300] Electrolyte infiltration[302,303] has been also modelled using a 3D resolved Lattice Boltzmann Method (LBM).[304–308] 3D-resolved process models have been also reported.[309]

As for the optimisation loop, an approach for multiobjective optimization and manufacturing inverse design has been recently reported in the literature.[310] The cost-effectiveness of physics-based models has been addressed in the literature by machine learning.[311] Data-driven strategies can also be used to improve the interpretability of battery manufacturing processes[312] and Convolutional Neural Networks combined with X-ray technology has been used for internal wrinkle detection.[312]

Currently, there is a lack of interconnection between machine models and process models in battery manufacturing. However, it is important to bridge this gap and couple multiscale models to represent the entire manufacturing value chain. By integrating machine models, which represent the operational behavior of manufacturing equipment, with process models, which simulate the physical phenomena occurring along the manufacturing process, a more holistic understanding of the entire process can be achieved. Combining physics-based models and data-driven modeling approaches in a hybrid analysis and modeling framework can be particularly beneficial.[313] Physics-based models provide a fundamental understanding of the underlying physical principles, while data-driven models leverage large volumes of data to capture complex relationships and patterns. By integrating these approaches, a robust digital twin platform can be developed, enabling battery researchers and manufacturers to make more informed decisions in the battery manufacturing chain.

All in all, the production of electrodes in battery manufacturing involves complex processes with multiple parameters that reflect various multi-scale and multi-physical phenomena. While simplified experimental design-based methods are currently used for optimization and scaling up, it is crucial to acquire comprehensive knowledge of the manufacturing processes to achieve better control and efficiency. To obtain complete knowledge, it is necessary to focus on automated data acquisition, data infrastructure, and data processing. This involves sensorisation to ensure accuracy and speed in capturing and analysing relevant information. Parameters such as electrode thickness and density can be measured at the macroscopic level, and these measurements are relatively common. However, evaluating the microstructure of electrodes and properties such as pore and binder distribution at the micro-level poses a challenge. Microstructural evaluation in real time is still under development and often comes with high costs. Establishing a direct correlation between macro-level information (obtained through sensors) and micro-level properties is an ongoing endeavour. Therefore, efforts should be directed towards advancing sensor technologies, improving data infrastructure, and developing analytical techniques for microstructural evaluation. This would enable more precise control and optimization of electrode manufacturing processes, leading to improved performance, efficiency, and quality in battery cell production.[314]

Finally, with the interconnected nature of a battery manufacturing facility, where different physical assets interact with each other, it is crucial to establish communication protocols and standards. These standards enable seamless interaction and data exchange between the physical assets and their corresponding digital twins. By adhering to standards, interoperability is enhanced, allowing different systems and components to work together effectively

7.5.2 Challenges

The development of **new battery cell designs** that minimize waste, reduce energy consumption, and achieve zero or low emissions is crucial for sustainable and environmentally friendly battery manufacturing. In this context, multiphysics modeling plays a significant role in battery design and manufacturing processes. However, to fully leverage the potential of multiphysics modeling, there is a need for the further development of comprehensive computational platforms integrating multi-scale physicochemical models with AI algorithms.

Such platforms would cover the entire manufacturing process chain of LIBs, from cell design to production. Computational simulations can be employed to explore and optimize new-generation battery cell designs, incorporating materials discovered through BIG-MAP and the integration of sensing and self-healing functionalities into battery cells can be explored through computational science, presenting exciting new challenges.

As the demand for LIBs increases, there is a need to continuously evolve and develop new technologies that offer higher energy density, longer lifespan, and improved safety. However, it is crucial to address the environmental impact of battery manufacturing itself, as it contributes significantly to the carbon footprint of electric vehicles and other applications. To achieve a sustainable and low-carbon future, it is necessary to redefine materials and manufacturing routes in a way that balances a low carbon footprint with high throughput and desired performance targets.

In the field of LIBs, ongoing research and development efforts are exploring innovative approaches such as metallic lithium anodes, intercalated thin film electrodes, and solid electrolytes made of polymers, ceramics, or hybrid materials. However, these advancements often necessitate a fundamental redesign of current manufacturing processes to accommodate the unique characteristics and requirements of these new materials and technologies. The development of these emerging battery technologies and their successful integration into the market requires a holistic approach that encompasses not only the design and performance aspects but also the manufacturing processes and their environmental impact. It involves exploring new paradigms in battery design and manufacturing, optimizing production techniques, and incorporating sustainable practices to achieve a greener future.

Given the disruptive nature of the concepts to be developed under the Battery 2030+ initiative, it is also necessary to think outside the box in the areas of cell design and manufacturing. Anticipating future battery technologies is challenging, making it difficult to predict the exact manufacturing concepts that will be required. However, we can identify advanced tools and technologies that are currently at the forefront and are expected to play a central role in the future.

This manufacturability roadmap aims to provide methods and approaches for developing manufacturing processes that go beyond the current state of the art. These methods involve pushing the boundaries of existing technologies and exploring novel techniques to enable the production of future battery technologies. By leveraging cutting-edge tools and technologies, manufacturers can develop more efficient, scalable, and sustainable manufacturing processes.

Three main challenges in battery production can be identified: the first one pertains to general methods ***for current battery production***, particularly roll-to-roll manufacturing processes. Ongoing research and development efforts are focused on improving the efficiency, reliability, sustainability and scalability of current production methods. These challenges are currently being addressed but will continue to be relevant as the production of future battery technologies evolves. Optimizing and adapting these production methods to accommodate new materials and concepts will be necessary.

The second challenge is associated with ***advanced and novel manufacturing concepts***, as well as optimized materials and approaches for future battery technologies. These challenges may be more difficult to anticipate since they are linked to technologies that are not yet fully realized. As new battery chemistries and designs emerge, novel manufacturing processes will need to be developed to accommodate their unique requirements. The development of these advanced manufacturing concepts will involve exploring new materials, production techniques, and quality control methods to ensure the successful production of future battery technologies.

The last challenge is independent of the specific technology or chemistry and relates to the ***scaling-up process***. Transitioning from laboratory-scale to pilot-scale or industrial-scale production poses significant challenges in terms of cost, efficiency, and quality control. Currently, this scaling-up process can be resource-intensive and time-consuming. However, by leveraging modeling and digital twin tools, it is possible to accelerate the scaling process from the laboratory to the pilot line.

Overall, addressing each challenge of battery production is crucial for achieving efficient and cost-effective manufacturing processes. Collaboration between researchers, industry stakeholders, and policymakers will be essential to overcome these challenges and drive the advancement of battery technologies towards sustainable and high-performance manufacturing chains. This is the core of the scope of Battery 2030+ and the focus of this roadmap.

According to this, the following challenges can be outlined:

A) Manufacturing challenges associated with current (mostly Li-ion) battery manufacturing methodologies

First, it will be necessary to overcome today’s use of trial and error as a general tool to fine-tune current battery manufacturing processes and shorten development time. The current process chain is highly complex and associated with very high investments. Competitive production currently requires the exploitation of economies of scale, which leads to so-called gigafactories with tens of GWh of manufacturing capacity. These factories are usually very specialised in terms of chemistry and limited to a few cell formats. Despite the strong optimisation of current production lines using trial and error, very large quantities of materials and cells still do not comply with specifications. This makes the change to new cell chemistries and materials, as well as the manufacturing of novel cell formats, very difficult and associated with high start-up costs and material waste. For this reason, the production of small series for special applications with a few tens of thousands of cells is very difficult and expensive, limiting the market launch of novel materials and chemistries.

Overall, lowering the general manufacturing process cost, with less solvent and energy use, reduced scrapping, and faster manufacturing, especially during the formation step, are needed.

Finally, with an eye on closing the loop, establishing cell designs and manufacturing process that enable component-level recycling and reuse (i.e., electrode recovery and reuse from end-of-life high-performance cells) will be crucial for the fully sustainable development of battery technologies.

B) Challenges related to future battery manufacturing technologies arising as a result of the foreseen highly innovative battery R&D scenario

There is a need for a flexible manufacturing process design strategy, as BIG–MAP produces innovative materials/interfaces with specific manufacturing demands. Rapid prototyping and high throughput methods will be needed to implement the design rules from BIG–MAP; in particular, direct wet-chemical-to solid methods are of interest. and fulfil the tag of being also within the improved CO_2 footprint conditions per produced material entity of the battery.

The introduction of self-healing materials/sensors plus their potential need for external physical connections at the cell level requires activation/bi-directional communication. Design rules are needed for these sensors from the production point of view, addressing scalability, automated integration, cost, and recyclability.

Additionally, it is believed that the introduction and viable upscaling of 3D or other mesoscale composite materials in electrode and cell processing, without affecting microstructure/functionality, will generate a specific need to preserve textural/functional properties.

All in all, research and tools to predict the impact of this new manufacturing parameters on the functional properties of battery components will be needed, partly in parallel with the introduction of new materials and concepts at the cell level. Additionally, there is a need for new manufacturing routes facilitating direct recycling methods that preserve the structural elements of the cell (e.g., electrodes and sensors), or consider stripping these at low energy and CO_2 footprints.

C) Challenges related to scaling up process

Understanding the cause-and-effect relationships between battery development and cell production is crucial for efficient and reliable manufacturing step. In this regard, there is a real need to better understand how results can be transferred from lab-scale to larger scaled production at pilot lines and ultimately to the giga scale. Improving data availability and understanding of the pilot line production of battery cells through advanced modelling efforts can address this need. The lack of standardized protocols presents significant challenges in scaling up battery production. Standardization can play a critical role in streamlining processes, ensuring compatibility, and enabling efficient scaling up processes.

In addition, current efforts in the battery industry are primarily focused on developing machines and systems for large-scale production. This emphasis on large-scale manufacturing can limit the availability of production systems suitable for small and medium quantities with flexible operating points.

Furthermore, increasing processing volumes in battery production can result in more elaborate and time-consuming test setups. As production scales up, it becomes essential to ensure rigorous quality control and testing to maintain consistent product quality and performance.

All in all, **developing digital tools for predicting the impact of processing parameters on the characteristics and performance of the final product is a valuable approach to move**

away from trial and error in manufacturing. These tools can significantly improve efficiency and reduce costs by enabling more informed decision-making. In addition, the digital twins will enable fast adaptation to new chemistries and new manufacturing facilities, as well as accelerate the scaling-up process. However, the implementation of digital twin technology in predicting the impact of processing parameters on product characteristics and performance faces certain challenges. Some of these challenges include:

- The current *parameterization activities* in battery manufacturing require significant resources and can be cost-intensive. Parameterization involves identifying and characterizing various parameters that feed models to replicate performance, reliability, and safety of battery cells. To address the cost-intensive nature of parameterization activities, researchers and manufacturers are exploring alternative approaches. On the one hand, this includes the use of simulation models and virtual testing methods to optimize the parameters identification process and assess performance in a more cost-effective manner. On the other hand, advancements in data-driven and machine learning techniques enable the analysis of large datasets to extract directly models parameters. Overall, while parameterization activities in battery manufacturing are currently cost-intensive, new advancements in modelling and data analysis techniques are expected to contribute to more cost-effective and efficient parameterization approaches in the future.
- *Process models and machine models in battery manufacturing are often not fully coupled or integrated*. Process models focus on simulating and understanding the physical and chemical processes involved in battery manufacturing, such as electrode coating, cell assembly, and formation step. On the other hand, machine models focus on the behaviour and performance of the manufacturing equipment itself, such as roll-to-roll coaters, stacking machines, and formation chambers. While both process models and machine models are valuable for understanding and optimizing battery manufacturing, the lack of coupling between them can limit the accuracy and effectiveness of the overall simulation and optimization process.
- *Physics-based models* can be computationally demanding and time-consuming, which can affect their efficiency. Developing accurate and efficient physics-based models for cells and their manufacturing is a challenging task due to the complexity of the underlying multiphysics processes and the multi-scale nature of battery systems. One reason for the computational inefficiency of physics-based models is the need to solve complex partial differential equations (PDEs) requiring numerical methods and iterative techniques, which can be computationally intensive. To improve the efficiency of physics-based models, researchers are exploring various approaches. These include model simplification and reduction techniques, such as lumped parameter models and reduced order models (ROM), which aim to capture the essential physics while reducing computational complexity. Additionally, advancements in numerical methods, parallel computing, and high-performance computing can help accelerate the simulations and make the models more efficient. Furthermore, the integration of physics-based models with data-driven or machine learning techniques can provide a hybrid modeling approach that combines the accuracy of physics-based models with the computational

efficiency of data-driven methods. Overall, while there are challenges in achieving high efficiency with battery physics-based models, further research and advancements in numerics and modelling techniques will help improve the efficiency.

- Current *inline sensors* used in battery manufacturing often focus on measuring macroscopic parameters such as electrode thickness and weight, which provide valuable information about the overall quality and uniformity of the battery components. However, they are limited in their ability to directly measure microstructural properties in real time. However, obtaining real-time measurements of these properties is challenging. Microstructural characterization typically requires destructive or time-consuming off-line methods such as scanning electron microscopy (SEM) or focused ion beam (FIB) imaging, which are not practical for real-time monitoring during battery manufacturing. To address this limitation, researchers are exploring the development of advanced in situ or non-destructive characterization techniques that can provide real-time information on microstructural properties. These techniques may involve the use of imaging technologies such as X-ray or neutron imaging, or spectroscopic methods to probe the internal structure of the battery electrodes. In any case, implementing such advanced inline sensors would enable real-time monitoring of microstructural properties and facilitate the optimization of battery manufacturing processes. However, the development of cost-effective and practical inline sensors for real-time microstructural characterization remains an active area of research, and further advancements are needed to make these technologies accessible for industrial-scale battery production.

Standards and protocols

Finally, the development of standards and protocols for process development and monitoring needs are to be seen as an important contributing factor to developing manufacturing processes that are both efficient and sustainable. However, this needs to be done considering commercial aspects that in certain cases can outweigh the benefit of standardization. Despite the challenges associated with standardization, there are a lot of positive factors showing the need to standardize. Advantages include increased interoperability, reproducibility and a positive effect on sustainability, as less scrap is to be expected when standardised procedures are used, which at the same time also has a positive impact on profitability.

To take advantage of the entire consortium, the long-term goal of standardization in manufacturability is to implement automated data collection of standardized and interoperable data for the Battery 2030+ Electronic Lab Notebook (ELN),[144] that should rely on BattINFO[110,145] as developed within BIG-MAP and extended to battery manufacturing.

This data should include a complete battery history as introduced by the Battery Passport,[315] such as battery chemistry, manufacturing protocols, SoH, and others. As such it can directly be utilized by BIG-MAP and can also be directly be used for recyclability to enable efficient and history-dependent recycling.

7.5.3 Advances needed to meet the challenges

Advances in battery cell manufacturing technology are needed to link process parameter variations more directly to observed cell performance, reduce overall scrap and energy consumption, accelerate the scaling up process and improve the resilience of production lines to changes in battery technology. The progress required in the **short term** in the development of battery cells and their production can be summarised as follows:

- Exploration for novel but industry integrational green and CO_2-reduced manufacturing routes for the battery materials and cell assembly. This further defines as an alternative by establishment of these strategies for recycling. Adaptability of manufacturing lines to respond to shifts in technology. This may include alternative methods of LIB production or transitioning to alternative chemistries like SIB-ion or solid-state cells.
- Reproducible deployment of sustainable and cost-effective manufacturing processes in giga-scale production. Manufacturing techniques like dry electrode coating lower the cost and improve the sustainability of cell manufacturing. More data on the practical scalability of these processes, as well as the long-term effects on cell performance and aging, are needed.
- Fast processing and screening tools working bottom down and up.
- A more robust supply chain for cell manufacturing equipment to ensure a seamless flow of equipment and materials.
- Stable open-source software libraries and tools supporting the semantic annotation and exchange of data, models, and interfaces in battery manufacturing. Existing proof-of-concepts should be scaled up, with a focus on community acceptance and implementation. The goal is to achieve plug-and-play interoperability among different machines, models, and databases in a cell manufacturing line. This activity should integrate with Battery Passport definitions.
- The development of computationally efficient models combining ROM techniques with advanced AI algorithms like Neural Networks (NN) or Physics-Informed Neural Networks (PINNs) can significantly enhance cell design and manufacturing optimization as well as control processes.
- Automatic parametrization of models when developing efficient models for cell design and manufacturing optimization.
- Smart sensors for implementation throughout the manufacturing value chain providing electrode microstructural properties, with the aim to enhance process monitoring, improve quality control, increase productivity, and optimize resource utilization.

These advances will offer a variety of benefits for the European battery sector, including:

- Improved sustainability in the battery manufacturing chain
- Improved battery material optimization via structure-property proxy definition and fast screens.
- Improved battery manufacturing efficiency
- Accelerated scalability process from lab to pilot line level, and ultimately giga-scale.

- Improved quality of battery cells, with reliable lifetime predictions.
- Improved resilience of the battery manufacturing sector regarding new chemistries or changes in technology.

7.5.4 Forward vision

Industry 4.0 represents the use of automation and data sets in a manufacturing scenario. This smart automation of the battery cell manufacturing routes is needed for efficient and autonomous management of massive production systems such as the ones found in lithium-ion battery giga-factories.

In the medium term, a proof of concept of a partial digital LIB cell manufacturing plant is expected. The smart automation of state-of-the-art LIBs cells manufacturing machines, will then require efficient and accurate models of products, materials, and processes which are then used to convert them into virtual models of the entire battery cell production operation. The smart automation will also have sensors present within many assets along the production line. These assets will then be able to communicate with each other to provide an in-depth insight into production line operation. This is then to be sent to the cloud, where data will be collected, analysed, stored and used for instance in predictive maintenance of the manufacturing plants.

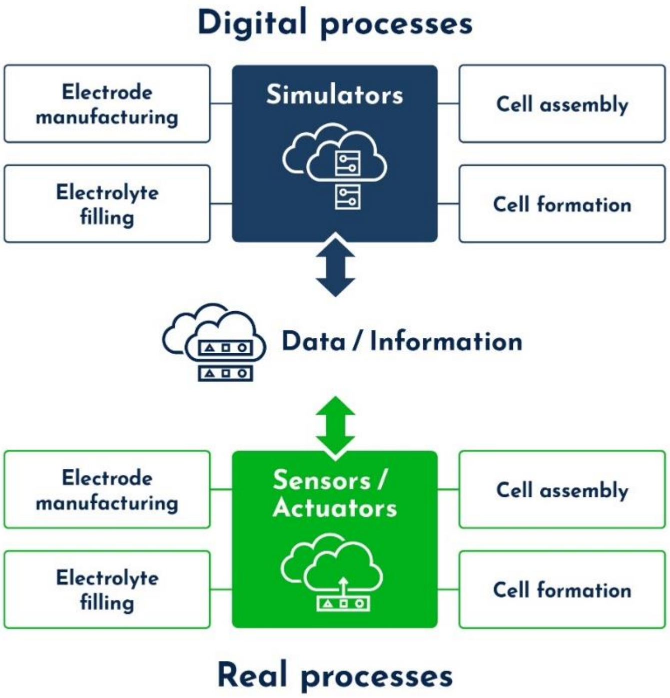


Figure 20. Digital twin of cell manufacturing processes.

Through this development, the main goal of the digital twin models designed for cell manufacturing processes is to resolve physical issues faster by detecting them earlier in the process, and to predict outcomes with a much higher degree of accuracy (see Figure 21). Additionally, their ability to evaluate the performance of equipment in real time may help companies obtain value and benefits iteratively and faster than ever before. All in all, through this implementation, substantial optimization of the selected critical steps is expected, making the manufacturing route more efficient and sustainable and minimizing human labour, trial and error, and waste products. Together with sensor data, the modelled data will be transferred to the cloud and analysed at suitable points over the horizontal integration.

In general, the main benefits of this approach are as follows:

- Speed up processing – rapid manufacturing and prototyping
- Improve quality control and generate cost reduction strategies
- Improve the homogeneity on production of the cells, producing more equal cells

In the long term, i.e., in ten or more years, the methodology is expected to be fully mature and close the loop by integrating the cell design and manufacturing design sub-loops (see Figure 22). In addition, a proof of concept for a digital twin of novel closed-loop recycling cell manufacturing routes for optimised LIBs is provided. All in all, digital twins in particular are expected to evolve from vertical integration to a horizontal, networked structure. It is assumed that digital twins will not only be designed for individual processes and coupled in real time, but that the entire process chain can be included. It will no longer be necessary to make optimisations only locally, but globally across the process chain. Parameters will be adjusted on the fly to achieve an optimal result for each individual battery cell. Finally, some parts of this methodology can be gradually made available to industry before the whole package becomes available as a commodity in a new state of the art.

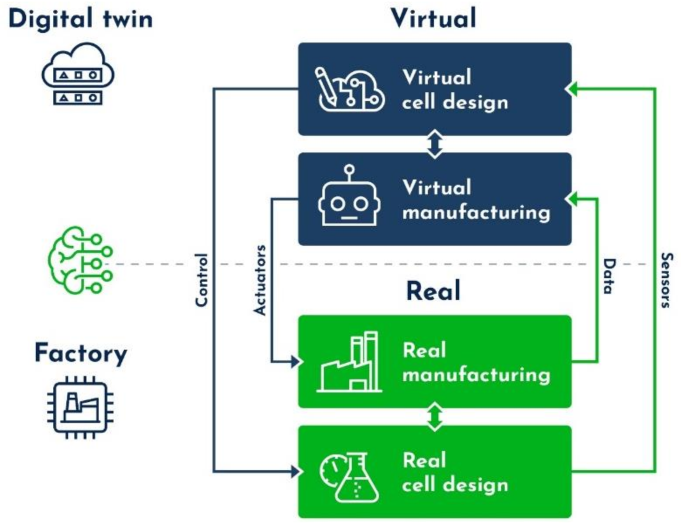


Figure 21. AI-driven design and manufacturing methodologies linked together as a whole.

Potential impacts of this approach:

Future battery manufacturing machines and tools, using advanced software and network sensors, will be used to plan, predict, adapt and control business outcomes, leading to optimisation of the entire value chain, with the following potential impacts:

- Accelerating the discovery of new cell designs and manufacturing processes; reducing new battery cell development time and costs; reducing battery research and innovation (R&I) costs.

- Increasing process speed, improving the efficiency of battery cell production facilities and reducing the number of problems as well as downtime, ultimately leading to cost savings.
- The finished battery cells will have a higher quality level and will be more convenient and cheaper to use and maintain.

Potential challenges of this approach:

- Data management (usable, accessible, integrated, and curated);
- Data harmonisation: Data standards are needed for the classification and unambiguous description of battery cells and their manufacturing chain;
- Standardisation and reference architecture: a reference architecture is needed to provide a technical description of these standards and facilitate their implementation;
- Intellectual property management (data ownership);
- The readiness level of the Internet of Things (IoT), which connects machines and systems and enables seamless data transfer across all assets (virtual and real replication);
- Increasing the adaptability of a battery production line to successfully produce battery cells of a new generation or technology.

7.6 Cross-cutting area: Recyclability

Glossary:

- Re-use: action or practice of using something again for its original purpose.

- Re-purpose: the process by which an object with one use value is transformed or redeployed as an object with an alternative use value.

- Recycle: process of converting waste materials into new materials and objects.

- Reconditioning: Servicing, readjusting, and recalibrating materials/equipment to bring them to near-new or original operational level.

- Circular economy: an economic system where products and services are traded in closed loops or cycles. A circular economy is characterized as an economy which is regenerative by design, with the aim to retain as much value as possible of products, parts and materials.

- Sustainability circle: a method for understanding and assessing sustainability and for managing projects directed towards socially sustainable outcomes.

- Extended Producer Responsibility (EPR): an environmental policy approach in which a producer's responsibility for a product is extended to the post-consumer stage of a product's life cycle.

- Direct recycling: refers to a novel recycling approach for batteries, in which the high value anode and cathode active powders and other components are recovered in whole from spent cells, separated from each other and from the other recoverable materials.

- Eco-design preparatory study for batteries: This study provides the European Commission with a technical, environmental and economic analysis of Batteries in accordance with relevant European Directives.[316]

- Eco-design Directive: provides consistent EU-wide rules for improving the environmental performance of products, such as household appliances, information and communication technologies or engineering.[317]

- Prospective Life Cycle Assessment (LCA): An LCA is prospective when the (emerging) technology studied is in an early phase of development (e.g. small-scale production), but the technology is modelled at a future, more-developed phase (e.g. large-scale production).[318]

The development of battery dismantling and recycling technologies with high efficiencies going well beyond the EU Battery Directive 2006/66/EC target of 70% [315] for most battery technologies is essential to ensure the long-term sustainability of the battery economy by 2030. This calls for new, innovative, simple, and low-cost processes targeting a very high recycling rate, small carbon footprint, economic viability as well as for logistics and business incentives. One technical approach will be the direct recovery of the active materials and single, instead of multi-step recovery processes. Furthermore, the new materials, interfaces/interphases, and cell architectures envisioned in Battery 2030+ call for new recycling concepts, such as reconditioning or reusing electrodes. Industrial participation will be brought on board early. To pave the way for such a shift, there will be a direct coupling to material suppliers, cell and battery manufacturers, main application actors, and recyclers to integrate the constraints of recycling into new battery designs and manufacturing processes: (1) design for sustainability (including eco-design as well as economic and social aspects considering the whole lifecycle), (2) design-for-dismantling and (3) design-for-recycling approaches. In such a way, the Battery 2030+ roadmap will promote a circular economy with reduced waste, small CO_2 footprint, and more intelligent and reduced use of strategic resources.

Implementation of design for sustainability and, more specifically, design for recycling is to be integrated in the algorithms for automated materials discovery (the input parameters can be the criticality of the raw materials, raw material toxicity, reduced number of elements, and other socioeconomic aspects). At the same time, both the recycling topic as well as the overarching theme of sustainability need to be accompanied by developing standards and protocols for assessing the economic and environmental validity of recycling processes. This can include also the development of ways to certify carbon footprint and overall sustainability of the complete battery life cycle.[53]

7.6.1 Current status

The battery recycling industry has developed significantly in the EU since the implementation of the Batteries Directive (Directive 2006/66/EC[315] updated with 2020/0353(COD)[319]), which introduced extended producer responsibility **(EPR)** for battery waste. The Directive forces battery producers, or third parties acting on their behalf, to finance the net cost of collecting, treating, and recycling waste batteries. The EPR concept is aimed at promoting the integration of the environmental costs associated with goods throughout their life cycles into the market price of the products. In addition, the EU has issued a number of supporting and guidance documents as well as the recycling efficiency regulation, specifying minimum requirements for battery recycling processes, according to the battery chemistries. Contributing to the Commission's "Circular Economy Action Plan", the revision of the Battery Directive 2006/66/EC was adopted on the 14th of June 2023 with updated categories and recycling efficiencies.[319,320] According to the just updated regulation, the recycled content should reach:

- **Minimum levels of materials recovered from waste batteries**: lithium - 50% by 2027 and 80% by 2031; cobalt, copper, lead and nickel - 90% by 2027 and 95% by 2031;
- **Minimum levels of recycled content from manufacturing and consumer waste for use in new batteries**: eight years after the entry into force of the regulation - 16% for cobalt, 85% for lead, 6% for lithium and 6% for nickel; 13 years after the entry into force: 26% for cobalt, 85% for lead, 12% for lithium and 15% for nickel.

Following timeline has been proposed to implement additional requirements for putting industrial and electrical vehicle batteries on the EU market, no matter the country of origin:

- July 2024: a carbon footprint declaration will be required[321] and by 2026: these batteries must feature a carbon intensity performance class
- As of July 2027, they will have to comply with maximum carbon thresholds.

- 2026: each industrial or electric vehicle battery with a capacity higher than 2 kWh shall have an individual “battery passport” linked to information about the characteristics of each battery type and model. An Electronic Exchange System (EES) in the form of an online battery database listing detailed information about all battery manufacturers and their battery types placed on the market may complement the passport.
- 2027: industrial and electric vehicle batteries with internal storage will have to declare their content of recycled cobalt, lead, lithium and nickel.

After potential dismantling and sorting into categories according to the battery chemistries, the batteries or battery parts are directly fed into the recycling process or further fragmented by physical means (e.g., shredding or grinding). In terms of recycling schemes, depending on the battery chemistry and process chosen, several steps involving physical, mechanical, and/or chemical transformations may be needed. Although each recycler may use variations or combinations of different individual steps, recycling processes (or schemes) are currently classified as shown in Figure 22. Currently, pyrometallurgy is the most applied method.[14]

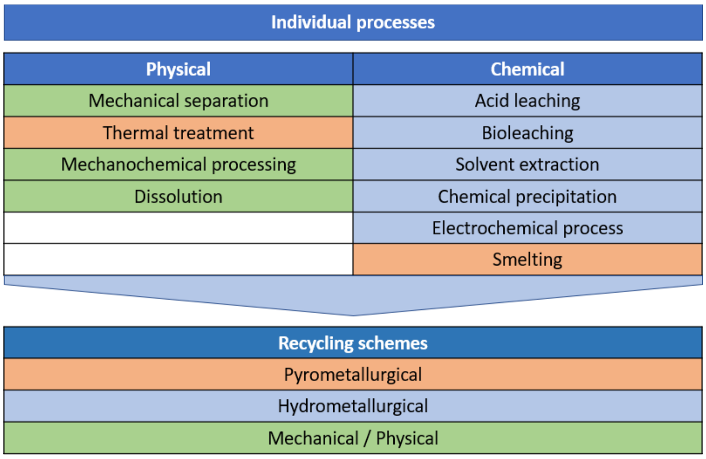


Figure 22. Recycling processes and schemes.

7.6.2 Challenges

The development of closed material loops in the interest of a circular economy will be required to ensure the security of supply after the ramp-up phase of the battery market. Innovative collection, processing, and recycling technologies will be needed for the recovery of not only valuable elements but of all cell components to increase sustainability.

The definition and implementation of design for sustainability for future batteries/cells will provide market advantages for European manufacturers and embed their products in closed loops. This approach will also decrease the dependency of the EU on critical metal imports, and support the usage of more abundant raw materials respectively. A quality of recycling enabling a closed loop usage is needed to recover the critical raw materials especially in the context of very large expected volumes of EOL automotive batteries expected in the timeframe 2030-2040 as shown in Figure 23.[322]

Currently, the global capacity for battery recycling is around 180 kt/yr. China accounts for almost 50% of this capacity and it is expected to retain its dominant position given the large amount of additional capacity it has announced.[323]

At 250Wh/kg, 1300 GWh to be potentially recycled by 2040 would need a capacity of ca. 5200 kt/yr (but a part of these EOL batteries will be shifted to second life).
Life cycle thinking, encompassing resource extraction, manufacturability, the use phase, and reuse/recycling, needs to be integrated into the design phase of new battery systems to increase their overall sustainability. In the following, current challenges as well as challenges foreseen for the medium and long terms are listed.

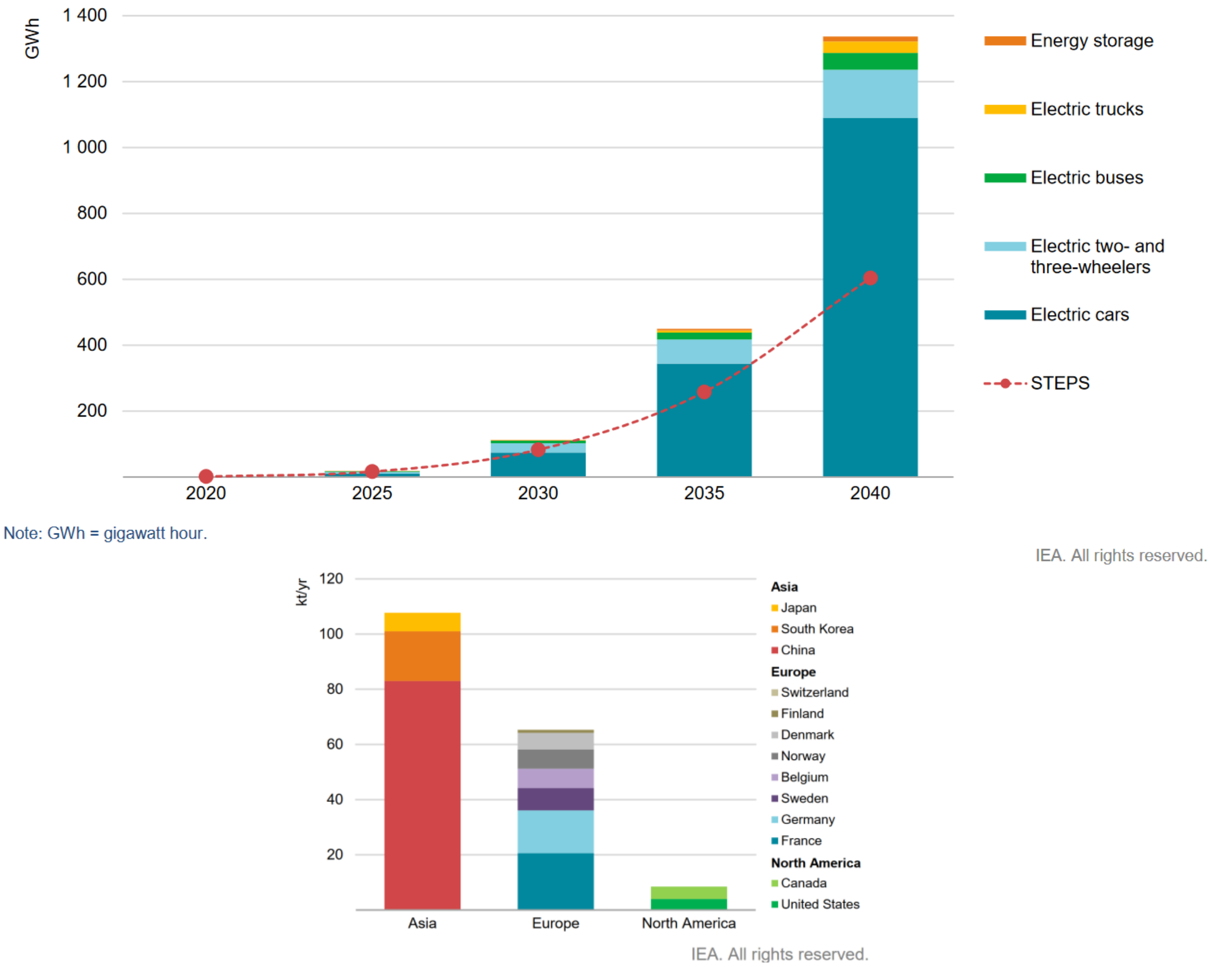


Figure 23. Amount of spent EV and storage batteries reaching the end of their first life by application until 2040 (top) and existing and announced lithium-ion battery recycling capacities to come online by 2021 by region (bottom).[322]

Current challenges:

- Battery collection targets need to be reached at end of life (Battery Directive), which seems to be less of a problem with automotive than with portable batteries. Many issues are related to collection and transportation of spent batteries.

- Batteries are complex products incorporating micro-components, embedded electronics, etc., and no available processes for efficient component separation exist today, causing high recycling costs.
- Labelling and automated, high-throughput detection of cells and batteries is necessary to sort mixed battery types and enable a highly efficient recycling process. For new batteries, this is partially addressed by labelling requirements in the Battery Directive revision.
- In particular EV automotive battery systems, are designed for high safety, and their dismantling poses a huge challenge to efficient recycling processes. State-of-the art battery disassembly is a manual process.
- The limited and decreasing value of the active materials of lithium batteries when compared with the cost of recycling promotes the need of **"direct recycling"** processes, however demonstrating the economic benefit of these processes will be a challenge. Direct recycling refers to a novel recycling approach for batteries, in which the high-value anode and cathode active powders and other components are recovered as such from spent cells, separated from one another and from the other recoverable materials, and reconditioned to battery-grade materials.
- Batteries' active materials degrade over their lifetime. For example, structural changes in the crystalline structure of the cathode materials of Li batteries may be irreversible, limiting the possibility of recovering them without a reconditioning process restoring the expected level of quality and functionality. Additionally, materials will be technologically outdated when recycled, e. g. $LiCoO_2$ or NCM-111 cathode powders introduced 10 years ago.
- New systems like Na-Ion batteries enter the markets for mobile and stationary applications. New recycling concepts are necessary of the recycling of low value, low environmental impact materials.[318,324]
- Methodological challenges: the economic, ecological, and social impacts of emerging battery technologies must be analysed and estimated in a prospective manner. All material, component, and cell developers as well as recyclers and other stakeholders need to work together in an interdisciplinary way, to reach shared visions on new battery systems.

Specific short/medium-term challenges:

- The number of battery chemistries on the market is increasing. Multiple Li-ion chemistries will make specific recycling processes more difficult, and sorting quality will become a major challenge to overcome in order to have specific processes applicable to component recovery.[325] Standards for identification are important on the battery and cell levels in order to overcome these challenges.
- New battery technologies seem likely to enter medium term markets, for example, solid-state, lithium-sulphur, redox flow, and metal-air batteries in mobility and stationary applications. Proposed new recycling processes to cope with all these chemistries (and related BMS) will create new process challenges; for example, the presence of Li metal

will affect safety aspects of the recycling processes.[326] Recycling processes may have to be redesigned, for example, to use an inert gas atmosphere, depending on the battery type.

- While the transition to aqueous processing of electrodes on the large scale is inevitable with regard to economic and ecologic improvements in battery manufacturing, the same relevance of this transition accounts for recycling and recovery processes of electrodes.[327–329] Obsolete binders and additives will have to be removed in advance to further recovery steps of active materials.[330,331]
- Despite recent progress regarding direct recovery of electrode active materials,[332,333] an additional upscaling of electrode chemistries will be necessary in many cases, as decommissioned batteries will likely contain outdated electrode chemistries. Although first results have been published, for example, the upscaling of lithium cobalt oxide (LCO) to $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$,[334] this represents one of the major challenges to be tackled within the next few years.
- Several recycling processes are likely to cause impurities in directly recovered electrodes such as aluminum or copper fragments from the current collectors.[335] Even though such impurities can be beneficial in some cases,[336] generally, these direct recycling specific aspects need to be overcome to obtain reusable and competitive electrodes.
- Following the large quantities of EV batteries available on the market, new business cases are appearing, for example, the reuse of battery modules or cells after sorting to provide a longer service life or a second life. As a result, the batteries eventually coming to final recycling can be expected to be at a more advanced degradation stage and in a more mixed condition. In addition, although desired, global battery standardization cannot be expected in the short/medium term given the multiple applications in the market, consequently chemistry identification and quality sorting will become even more challenging. In near future, decommissioned batteries will not provide sufficient information about cell chemistry and electrode condition to handle their recycling in an ideal way, which is why fast analytical measures like lithium content determination in cathodes have to be implemented.[337] The required level of expertise can only be expected if advanced AI development, including marker particles for example with magnetic codes,[338] complements more traditional recognition means such as labelling and visual observation.
- The amount of information associated with batteries will increase, first through more and more sophisticated BMS, then with information from sensors and future battery passport. Processes to handle information from these innovations during the recycling phases will have to be developed and standardized. Such advanced data will provide valuable input for second-life applications and options to exchange individual aged battery cells in a battery pack.
- The huge amounts of battery systems/modules to be recycled will require enormous logistical efforts, and transportation of these systems/modules will significantly increase costs, safety issues, and the CO_2 footprint. Novel decentralized collection and recycling processes/units need to be established, and low environmental footprint as well as societal acceptance issues to be obtained.

- A legislative framework must be established to foster/safeguard sustainable design, including design for recycling.

Tentative longer-term challenges:

- Beyond 2030 novel emerging battery technologies may appear in the market such as Mg, Al and Ca based batteries.
- Large volumes of spent batteries will require the transformation of recycling plants and a move to highly automated processes from sorting and dismantling down to the recycling itself. Generation 4.0 recycling plants will call for major investments. Innovation will be needed to demonstrate highly flexible but economically feasible processes for all the steps of recycling, enabling the treatment of multiple sources of batteries with potentially different chemistries.
- The recycling technologies will need to recover future intelligent battery components such as sensors, self-healing components, and any kind of information-linked components.
- Additional circular economy business ecosystems for reconditioning and/or reusing recycling products/materials will have to be developed and located near battery recycling units (decentralized, if possible).

7.6.3 Advances needed to meet the challenges

It is the ambition of Battery 2030+ to transition to a new recycling model based on data collection and analysis, automated pack disassembly to the cell level, investigating reuse and repurposing whenever possible, automated cell disassembly to maximize the number of individualized components, and the development of selective powder-recovery technologies that recondition powders to battery-grade active materials that are reusable in batteries for automotive/stationary applications with significantly reduced logistical efforts.

The present EU activities "Eco-design preparatory study for Batteries"[316] has the goal to provide the European Commission with a technical, environmental, and economic analysis of Batteries in accordance with relevant European Directives, especially the Eco-design Directive (2009/125/EC).[317] Sustainability is addressed within this description, but social aspects are not considered.

In contrast to the "Eco-design preparatory study for Batteries", not only technical, environmental and economic aspects will be considered in Battery 2030+, but also social aspects to ensure sustainability. Furthermore, the proposed approach will be technology neutral to accommodate any innovative developments.

Battery 2030+ aims to provide a basis for holistic sustainable battery design starting from raw and advanced materials, design for manufacturing, and material recycling. It will provide criteria and requirements for BIG–MAP and sensing functionalities to enable high-efficiency recycling to recover critical raw materials and minimize the carbon footprint. The focus is not only on the use phase, but on the whole life cycle (i.e., life cycle sustainability) by means of prospective life cycle assessment (LCA), contributing by defining rules and standards for the recycling part of the loop. The implementation of standards and protocols in recyclability constitutes one important aspect on reaching a circular economy, in improving the efficiency of recycling processes and in decreasing the dependency on imports. An important aspect here is the link to manufacturability and information about the entire battery history which are directly transferred to recyclability (see Table 5). This is to be accomplished by standardized and interoperable automated data acquisition for the Battery 2030+ Electronic Lab Notebook.

The ambition of Battery 2030+ is to develop a ground-breaking new recycling process compared with the current state of the art. The current recycling flow, through pyro and hydro processes encompassing multi-processing steps, is summarized as shown in Figure 24. Considering the increasing variety of battery designs and chemistries, as well as the technological readiness, a multilateral approach to battery recycling consisting of pyro and hydro processes, as well as direct recycling methods will dominate the next decade.[339] However, in light of sustainability, an increased focus on direct recycling methods, where not only the most valuable but all components are recovered, is inevitable. Furthermore, the dependence of hydro and especially pyro processes on the market value of metals like cobalt and nickel will result in higher economic volatilities and less planning reliability.[340]

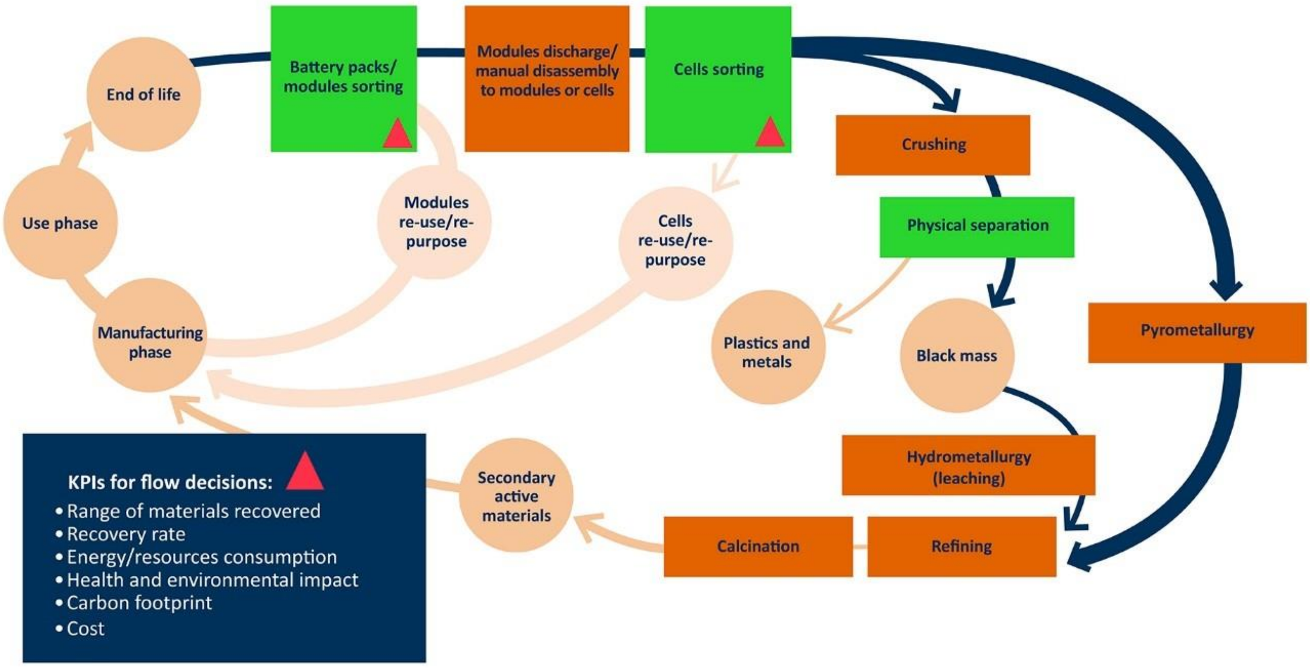


Figure 24. Present recycling process.

Based on a novel integrated approach to recycling designed materials (as developed in BIG–MAP) and sensor technologies (as developed in the "Sensor" section), Battery 2030+ will come up with a *new model* (see Figure 25) based on:

- Data collection and analysis (e.g., from labels, BMS, sensors, battery passport).
- Modern small-carbon-footprint logistics concepts, including decentralized processing.
- Automated pack disassembly to the cell level.
- Investigating reuse and repurposing wherever possible.
- Automated cell disassembly to maximize the number of individual components.

- Development of selective technologies for powder recovery and powder reconditioning to battery-grade active materials reusable in batteries for automotive/stationary applications. When not possible, precursor synthesis is eventually envisaged with composition adjustments.
- Finally, optimized pyro- and hydro-metallurgical processes applied to ultimate waste should demonstrate the high recovery rate expected for critical raw materials.
- International collaboration to be stimulated and developed.

In order to be able to properly and comparably assess the individual process steps in terms of their economic and environmental implications, to ensure the validity of such assessments, and to provide a framework for future regulatory efforts in battery production, use, re-use and recycling, standards and protocols will be developed in close coordination with other European and international consortia, initiatives, and regulatory bodies. The aim of such activities will be to create a harmonized framework for the assessment and certification of economic, environmental, and societal impacts of large-scale battery production, use and recycling in high-volume applications such as traction batteries.[53]

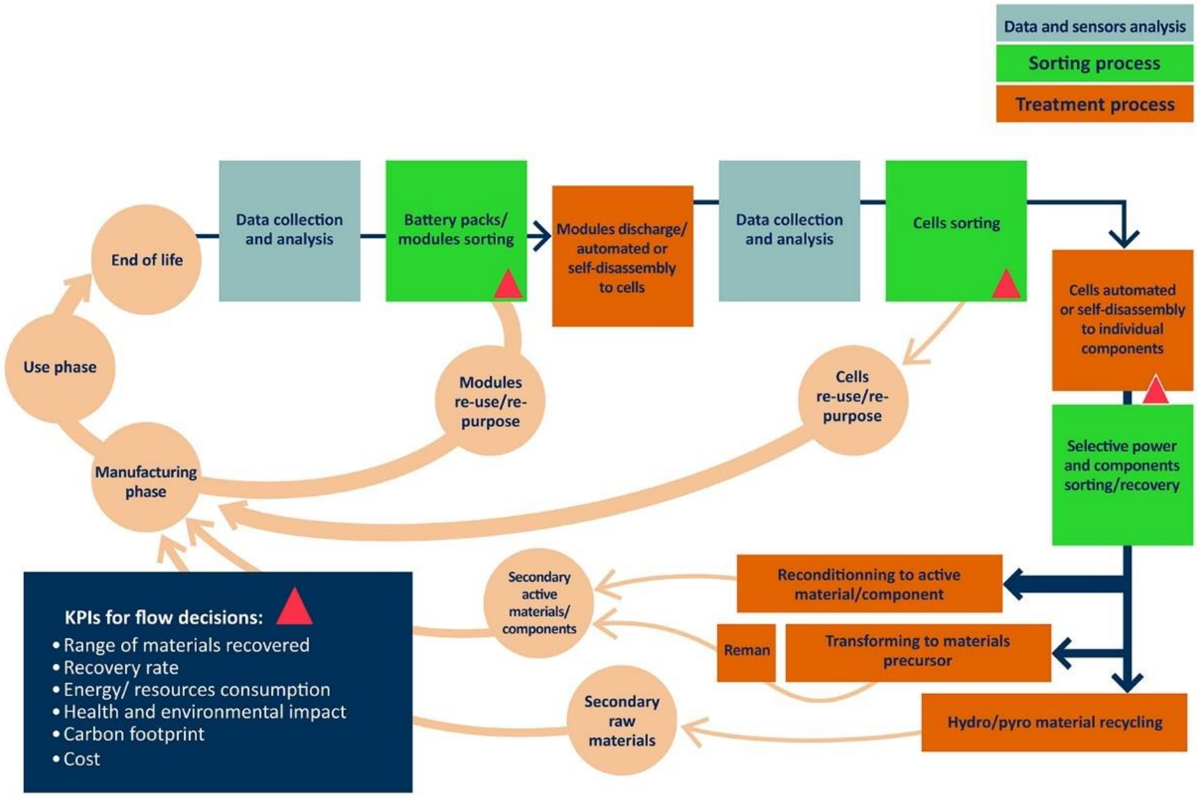


Figure 25. Future recycling process: direct recycling fully integrated with reuse.

While Figure 25 summarizes the total approach of the complete circularity loop, obviously not all the steps are currently on the same TRL level. Table 3 describes the respective current TRL levels and the priorities set by Battery 2030+.

Table 3. Current TRL levels and priorities set by Battery 2030+.

	TRL	Battery 2030+ priority
Design for sustainability/recycling	3	3
Packs/cells data collection and analysis	2	2
Battery packs/modules sorting	3	2
Fast SoH determination (<30 min)	1	2
Automated disassembly packs/modules	2	2
Re-use/re-purposing/second life echnologies	8	5
Cells sorting	2	2
Cells opening/automated disassembly	2	2
Selective separation/recovery materials from cells	1	1
Reconditioning technologies materials/DR	1	1
Validation materials in automotive/ESS new cells	1	1
Back-up pyro/hydro process if DR not successful	9	6
Recommendations for design/standardisation	3	4
Social approval	3	4

The activities with priority 1 correspond with fundamental low TRL work focusing the implementation of Direct Recycling, aiming at developing material sorting technologies, material reconditioning for its chemical and physical composition (including re-lithiation, re-coating) and finally product validation.

Summarizing the priority actions:

1. Selective separation/recovery materials from cells: based on cell cutting or shredding

- ➢ Electrode separation and recovery active materials
- ➢ Technologies such as ultrasonic, froth flotation.

2. Reconditioning technologies materials/DR

- ➢ Cathode relithiation
- ➢ Cathode upcycling and impurity impact

3. Validation materials in automotive/ESS new cells

7.6.4 Forward vision

The new process for recyclability will be the basis of a series of R&I actions with the main purpose of implementing direct recycling in the long term (see Figure 26).

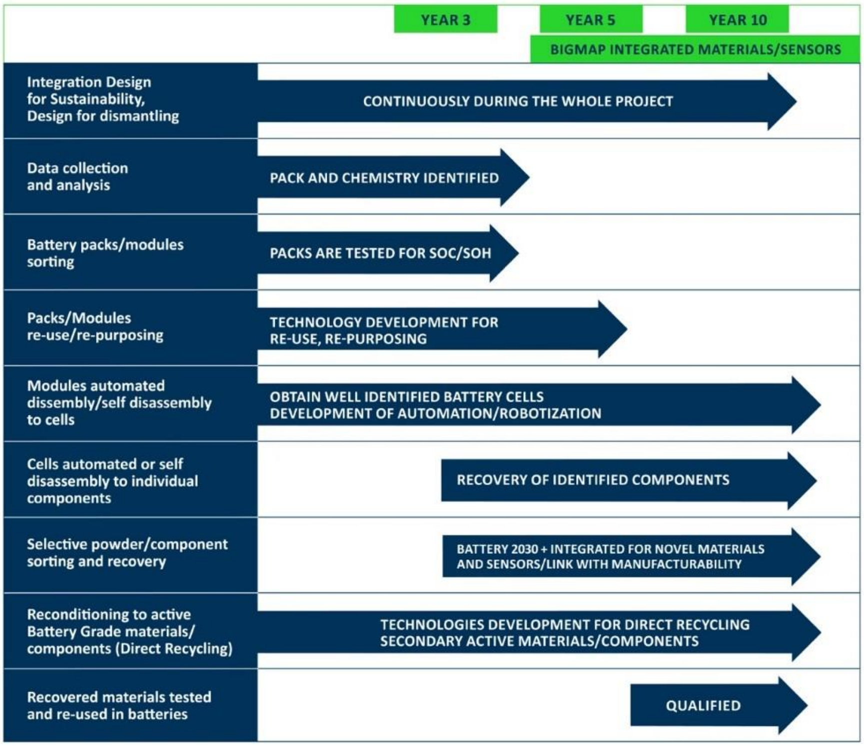


Figure 26. The ten-year roadmap for recyclability within Battery 2030+.

If the materials/components are not suitable to be reconditioned to battery grade because of, for example, structural or purity constraints, a fall-back alternative in the last stage of the new process could be to convert them to precursors with a view to eventual changes of composition ratios, anticipating future chemistry changes and new generation materials.

A full description of proposed recycling process and its state of the art is presented in the Advanced Energy Materials publication "A Roadmap for Battery Research in the context of the European Battery 2030+ Initiative".[14]

***In the short term*:** Start integrating design for sustainability and dismantling, develop a system for data collection and analysis, start-to-end traceability, develop technologies for battery pack/module sorting and reuse/repurposing, and start developing the automated disassembly of battery cells. Develop new tests for rapid cell characterisation.

***In the medium term*:** Develop the automated disassembly of cells into individual components, as well as sorting and recovery technologies for powders and components and their reconditioning to new active battery-grade materials. Test recovered materials in battery applications. Develop prediction and modelling tools for the reuse of materials in secondary applications. Significantly improve, relative to current processes, the recovery rate of critical raw materials (e.g., graphite recovery) as well as energy and resource consumption.

***In the long term*:** Develop and qualify a full system for direct recycling; the system should be economical, viable, safe, environmentally friendly, and have a smaller carbon footprint than current processes.

8 A closed loop between the research areas

The research areas outlined in the previous section aim to play a crucial role in inventing the battery of the future. Individual advancements in each respective research area are extremely important for the development of new battery technologies, however the collective effort and synergies among the research areas will be key to the overall success of Battery 2030+ and its ambitious vision to reinvent the way to invent batteries. All of them are interconnected and rely on a continuous exchange of information and ideas between the different areas, ultimately forming a closed loop system. The long-term vision is to achieve accelerated and automated research that is able to discover and invent new self-healing batteries which can be directly manufactured and recycled, while also being safe, sustainable, cost-effective and have electrochemical properties tailored for specific applications. To foster effective collaboration between the research areas, specific goals were set for the short, medium and long term, which are presented in Table 4.

Long term

A **closed loop** must develop between all research areas in the **long term**. This requires the research areas of **BIG-MAP**, **Sensing** and **Self-Healing** to closely interact in the first place. A continuous and efficient feedback loop must be established between sensor data, the BMS, and AI modules. The self-healing functions will be appropriately triggered in response to external stimuli detected by sensors or based on predictive models from BIG-MAP. These self-healing properties should be able to restore not only the cell components but also the sensors implanted in the cells. To detect criteria for self-healing to be triggered, autonomous procedures will be set in place for multimodal characterization and analysis of the smart batteries and multiple self-healing properties will be detected with universal and unique models. With this closed loop, the cycle life, longevity, reliability and safety of future batteries will be highly improved. New materials and interfaces will be discovered at accelerated rates, with direct feedback integrated into Sensing and Self-Healing research areas. To reach the envisioned closed loop, a close interaction with the cross-cutting areas is mandatory.

Another central aspect where communication is crucial to forward development, is the **link** between the **Cross-cutting areas** Manufacturability and Recyclability to **BIG-MAP**, **Sensing** and **Self-Healing**. Establishing a closed loop system will enable a feedback mechanism to efficiently manufacture and recycle next-generation battery cells incorporating new materials, engineered interfaces, sensors, and self-healing functionalities in the long term. This approach will lead to new, environmentally friendly and cost-effective batteries.

One of the objectives is to ensure that, after their first life, the sorted materials will be given a second life and reintroduced at the beginning of the production chain. This process includes preserving their full history and material information which will be obtained from sensor data. To capture all necessary cell data during their lifespan, the interlink between Sensing and the cross-cutting areas requires the automated deployment of new advanced sensors in next-generation cells at pilot line level, under recyclability constraints. This will be enabled by the feedback loop between BIG-MAP and the cross-cutting areas, resulting in the development of new cell designs that are optimised for recycling and a full proof of concept of a digital twin in manufacturing. To ensure constant information flow, sensors will be connected to an external connection point to constantly feed information into the BMS and transferring it to BIG-MAP. The automation of the integration and connection of the internal wiring interfaces during the cell assembly and potentially extending it to the module level while adhering to recyclability constraints is another important factor within the loop.

Self-Healing components are aimed to be automatedly fabricated with considering recycling constraints at pilot line level supplemented by a proof of concept of automated insertion of self-healing components into cells. Ultimately the demonstration of a manufacturing process for new battery technologies evolving from the feedback loop with BIG-MAP is envisioned by integrating recyclability criteria (i.e., metalized plastic to replace current collectors).

Short and Medium term

In order to achieve close collaboration between related research areas, various short- and medium-term goals have been established, defining steps between each pair of the individual research areas.

Between **BIG-MAP & Sensing** the first steps are to correlate data from sensing and from operando characterization. One difficulty thereby is to obtain compatible data from different sensor types, which requires finding solutions to standardise and ensure compatibility in the output format. For comparability an ontologized data management will be put into place. Next steps involve conducting on-the-fly analysis of multimodal data obtained through sensing on instrumentalized batteries. The multi sensor input will be transferred to the BMS. With the sensing input data feeding BIG-MAP, material characterization and discovery will be accelerated, leveraging the real-time utilisation of sensing data.

With the aim of combining preemptive and curative approaches in future batteries, Self-Healing is another research area which has to be interlinked to the other research areas early on. Specifically, between **BIG-MAP & Self-Healing** the first steps involve detecting the effectiveness of self-healing mechanisms, accompanied by the development of predictive models to understand how self-healing works in the cell (e.g., how dendrite growth is suppressed). Based on these models, new materials and interfaces for self-healing (e.g., electrolytes) can be developed. Additionally, the established predictive model will be utilised to predict potential failures in self-healing and estimate the lifespan of the self-healing properties. In the medium term, the focus will be on implementing the established predictive model to predict failures in self-healing and triggering preventive self-healing measures. By establishing these interconnections and collaborations, the research areas of BIG-MAP, Self-Healing, and Sensing will work together to advance the development of improved self-healing capabilities in future batteries.

The necessity of a close connection between the smart functionalities **Sensing & Self-Healing** is apparent. In the short term a constant feed of data regarding the self-healing process has to be measured by sensors, giving feedback on self-healing efficiency. One difficulty here is the frequency of sensing data from self-healing needed to interact with the BMS to reduce data traffic. The amount and location of sensors has to be specified to ensure proper functioning of the sensors and accurate measurement of self-healing properties. In the medium term, it becomes necessary to evaluate the sensitivity and accuracy of sensors during long-term cycling as well as the effects of aging of sensors along with the sensor response to the cell being considered. Sensors will constantly monitor the state of health (SoH) and the self-healing functionalities to evaluate their long-term effectiveness. When sensor data detect cell malfunction, self-healing will then be triggered.

Already in the medium term, **BIG-MAP, Sensing & Self-Healing** will closely interact, to combine preemptive and curative approaches. Sensor data on self-healing and SoH of the cells will be fed into BIG-MAP to develop predictive models. Thus, curative self-healing based on sensor data can be triggered as well as preemptive self-healing based on BIG-MAP's predictive models.

For interlinking **BIG-MAP & Cross-cutting areas** the exploration of new cell designs for the disruptive materials evolving from BIG-MAP while considering recyclability constraints will be in the focus in the beginning. New manufacturing routes will be developed for the BIG-MAP components, derived from AI data-driven models. In the medium term, the feasibility of the new and flexible manufacturing processes for the novel battery chemistries will be demonstrated.

In the short term, it is necessary to develop procedures to automatically insert the benchmark sensors inside the cells at pilot scale, linking the research areas of **Sensing & Cross-cutting areas**. Initially, the focus will be on LIB cells, while new chemistries will follow in the medium to long term. To manufacture the new batteries with smart functionalities, in addition to sensor integration, the connection to the BMS has to be established. Therefore, special focus will be given to the adaptation of internal interfaces and connections (like communication pathways, electrical connections and power, etc.) to the cell manufacturing tools and constraints all while considering recyclability. In the medium term, the implementation of new advanced sensors under manufacturability and recyclability criteria is to be demonstrated. Again, the communication interfaces between sensors and BMS are crucial. The sensor fabrication process and the establishment of sensor communication interfaces at the cell level to the battery management system (BMS) will be implemented for enhanced monitoring and control.

Since not only sensors, but also self-healing materials and functionalities are to be integrated in the new generation cells, the connection between **Self-Healing & Cross-cutting areas** has to be considered. Two aspects have to be considered here, the first being the manufacturing of self-healing components, the other being the integration of self-healing components into the cells. The starting point would be the exploration of self-healing functionalities that can be using existing equipment. With the variety of self-healing materials at hand and new self-healing materials yet to be developed within BIG-MAP, it will be necessary to reevaluate the existing manufacturing routes and explore new manufacturing routes for new self-healing components. This might also lead to the development of new cell design configurations including self-healing components. Ultimately, a procedure for the manufacturing of the self-healing components (i.e., self-healing electrodes) in LIB cells will have to be developed. Building upon the short-term goals, the medium term objectives involve demonstrating the integration between manufacturability & recyclability criteria with the development of new self-healing components. The manufacturing of self-healing functionalities in spatial distribution in a roll to roll process is to be shown.

The last aspect is the connection of the cross-cutting areas themselves, **Manufacturability & Recyclability**. Since the goal is to develop and qualify a full system for direct recycling, not only state-of-the-art LIB batteries will have to be considered, but also new materials and interfaces discovered in BIG-MAP, as well as sensors and self-healing materials built into the cells to accomplish smart functionalities. All these factors have to be considered from the beginning. In the short term, concepts for the design for sustainability and recyclability will be integrated into the manufacturing routes. These concepts will not only be integrated into the real processes but also in the digital twin, thereby implementing design for sustainability and recyclability concepts in the AI data-driven models. The design of the cell will be approached with a focus on sustainability and recyclability right from the beginning. In the medium term, an initial proof of concept will be demonstrated, showcasing the integration of manufacturability criteria into recyclability goals (easy to dismantle, sort and reuse, aligning with the principles of a circular economy).

The establishment of a closed loop between the research areas will be essential for the successful development of Europe's new, safe and sustainable batteries and battery technologies with properties that are tailor-made for their specific applications.

To realise this vision, it is necessary to implement a final piece: using consistent terminology throughout all research areas and creating a common ontology and standardized protocols. Goals on implementing standardization in the research areas are summarized in brief in Table 5, emphasizing the importance of unified language and protocols to facilitate effective collaboration and communication across the various research areas.

Table 4. Cross-linked short-, medium-, and long-term goals.

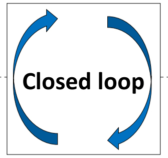
Cross-link	Short term (3 years)	Medium term (6 years)	Long term (10 years)
BIG-MAP & Sensing	Data from sensing and from operando *characterization* are correlated.	On-the-fly analysis of multimodal data from sensing on instrumentalized batteries.	The self-healing properties also include the healing of the sensors. Multiple self-healing properties can be detected with universal and unique models, thus autonomous procedures are in place for multimodal characterization and analysis of smart batteries.
	Data of different sensor types regarding the output format is standardized and compatible.	Accelerate material characterization & discovery by on-the-fly utilization of sensing data in BIG-MAP.	
	An ontologized data management is in place.	Multisensor input is transferred to the BMS.	
	Data is efficiently transferred from sensing to modelling, and from modelling to sensing.	Preemptive & curative approaches are combined with an emphasiz on interfaces and forwarding ontologies.	
BIG-MAP & Self-Healing	Monitoring and assessment of self-healing.	Preemptive & curative approaches are combined.	
	A predictive model is established to predict failures in self-healing and estimate the end of self-healing properties to work. Preventive self-healing is triggered.	Efficient feedback loop between sensing, the BMS, and/or AI modules to appropriately trigger the self-healing functions by external stimuli which are already implanted in the cell are established.	
	Development of electrolytes for self-healing and predictive modeling of how self-healing works in the cell (e.g., to suppress dendrite growth).		
	Self-healing data is transferred to the BMS.		
Sensing & Self-Healing	Self-healing is triggered based on sensor data.	Sensitivity and accuracy of sensors during long-term cycling and effects of sensor aging along with the sensor response to the cell.	
	The state of health and the self-healing functionalities are monitored with sensors to evaluate the long-term self-healing functionalities.	Efficient feedback loop between sensing, the BMS, and/or AI modules to appropriately trigger the self-healing functions by external stimuli which are already implanted in the cell are established.	
	Preemptive & curative approaches are combined.		
BIG-MAP & Cross-cutting areas	Exploration of new cell designs for the BIG-MAP disruptive materials, considering recyclability constraints.	A demonstration of the new and flexible manufacturing processes of the novel battery chemistries	
	New manufacturing routes of the BIG-MAP components, based on the AI data-driven models.		
Sensing & Cross-cutting areas	A procedure for the automatic insertion at pilot scale of the benchmark sensors inside the LIB cells.	A demonstration of the integration between manufacturability & recyclability criteria and the development of new advanced sensors.	Efficient feedback loop between BIG-MAP, Sensing, Self-Healing and the cross-cutting areas to efficiently manufacture and recycle next-generation battery cells incorporating new materials, engineered interfaces, sensors, and self-healing functionalities. Automated deployment of new advanced sensors in next-generation cells at pilot line level under recyclability constraints. Automated fabrication of easily recyclable self-healing components at pilot line level & POF of automated insertion of self-healing components into cells. Automation of integration and connection of internal wiring interfaces during cell assembly and possible transfer to the module level under recyclability constraints. The sorted materials are introduced in the beginning of the manufacturing chain for second life. Demonstration of manufacturing process for new battery technologies (SSBs, SIBs, etc.) by integrating recyclability criteria. Full POC of a manufacturing digital twin for LIBs by integrating recyclability criteria. Green & Large scale manufacturing with accelerated self-healing effect to be introduced. AI-based & high throughput manufacturability methodology for cells having accelerated self-healing mechanisms.
	Adaptation of internal interfaces and connections (communication pathways, electrical connections and power, etc.) to cell manufacturing tools and constraints under consideration of recyclability.	Integration of sensor fabrication process and their communication interfaces at cell level to the battery management system (BMS).	
Self-Healing & Cross-cutting areas	Exploration of self-healing functionalities that will enable manufacturability on the existing equipment.	Demonstration of spatial distribution of self-healing functionalities manufactured with roll to roll processes.	
	A procedure for adaptable manufacturing of the self-healing components (i.e., self-healing electrodes) in LIB cells.	New methodologies on multiscale modelling of manufacturing to be introduced and validated.	
	New cell design configurations including self-healing components to be explored.	Special cell design configurations to facilitate self-healing reactions	
	An energy-storage perspective for modelling of manufacturability to be introduced.		
	New manufacturing routes for self-healing components, considering recyclability constraints.		
	A demonstration of the integration between manufacturability & recyclability criteria and the development of new self-healing components.		
Cross-cutting areas: Manufacturability & Recyclability	Integrated design for sustainability and recyclability concepts in the manufacturing routes.	An initial POC of the integration between manufacturability criteria and the recyclability goals (easy to dismantle, sort and reuse).	
	Implement design for sustainability and recyclability concepts in the AI data-driven models.		
	Consider sustainability and recyclability concepts in the design of the cell.		

Table 5. Short-, medium-, and long-term goals for Standardization in the research areas.

	Short term (3 years)	Medium term (6 years)	Long term (10 years)
BIG-MAP & Standards	Continue the development of the ontology eco system.	Go **from** the **Electronic Lab Notebook (ELN) to** the **Lab as a Service (LaaS)**.	Accelerate research by use of ontologies & standards.
	Establish international collaborations.	Utilize the ontologies and standards to make data fully FAIR.	
	Realize a broad implementation of the **Battery 2030+ Electronic Lab Notebook** (ELN).	Have well-defined & standardized interfaces to enable reproducibility & interoperability.	
	Find attractive ways for researchers to use ontologies and standardization.		
	Find ways to include new metadata and observations in otherwise standardized processes.		
Sensing & Standards	Ensure a transparent flow of information and enable the comparability of sensor results (sensor sensitivity and type, data postprocessing, environmental conditions etc.).	Integrate sensor connectivity and data management with the BMS interface at the cell, module, and pack levels while maintaining compatibility with battery manufacturing processes.	Standardized communication with the BMS and generation of standardized sensor data for the **Battery 2030+ Electronic Lab Notebook (ELN)**.
	Define how to determine data from measurements for each sensor type.	Standardisation of the sensor integration process and connections.	
	Ensure the metrological traceability of sensors with regards to primary references in order to ensure comparable measurements and hence more meaningful experiments.		Automatized and standardized insertion of advanced sensors in the new generation cells.
	Define and report measurement conditions for each sensor type in use (e.g. definition of the compression frame for pouch cells.).		
	Implement unified calibration procedures for certain sensor types (especially for sensors inside the cell).		
Self-healing & Standards	Short-term standarization activites not relevant due to low TRL.	Evaluation of the need for standardization activities, based on the results of the ongoing BATTERY 2030+ projects.	First standardization activities for self-healing components in the cell.
	Clear definition of self-healing needed (both for autonomous and triggered).		
Manufacturability, Recyclability & Standards	Standardization of metadata reports and data produced by digital tools (battery models, etc.) in use.	Process neutral and machine open standardization.	Be able to hand over full battery history (battery passport: chemistry, manufacturing protocols, SoH,...) to recyclability.
	Standardized protocols and reports in use.	Standardized & interoperable **Battery 2030+ Electronic Lab Notebook (ELN)** in use.	Standardized interoperable automated data acquisition for the **Battery 2030+ Electronic Lab Notebook (ELN)**.
	Inline quality control for common chemistries and processes in place.	Standardization in validation of digital tools.	
	Find ways to handle sensitive data.		

Ontologies & Standardization have been identified as key aspect to be addressed for reaching the ambitious goals of Battery 2030+. They will enable reproducible results, facilitate communication, accelerate new discovery through the use of standardized electronic protocols that feed into an Electronic Lab Notebook, and lead to a more holistic understanding of the battery production process, to name just a few benefits. To reach these goals, data generation, data processing, data storage, data exchange and metadata treatment have to undergo the research data management strategies described in the respective research areas and which are summarized in Table 5. These standards are to be used to harmonize work strategies already in place but varying throughout the consortium. Eventually, ontologies and standards are tools for improving the quality of research, enforcing the FAIR[43] data principles and enabling closer collaboration between all research areas on an overarching level, with Battery 2030+ as a testbed. Ontologies and standards will be implemented in close cooperation with other national and international partners in practical and feasible ways. Developments from Battery 2030+ can then be deployed outside the Battery 2030+ consortium, e.g., by use of formal standardization bodies such as ISO and IEC.

Summary

Europe is presently creating a strong battery research and innovation ecosystem community, where Battery 2030+ has the role to provide a roadmap for long-term research for future battery technologies. LIBs still dominate the market for high-energy-density rechargeable batteries. However, current generation LIBs are approaching their performance limits, despite new generations coming in near time. The transition toward a zero-carbon emission society calls for the development of batteries with higher performance, with respect to both energy and power density. Future batteries must have an improved ecological footprint. They will be characterized by outstanding lifetime and reliability, as well as enhanced safety and environmental sustainability. This will most likely require batteries that are approaching their theoretical limits, providing the opportunity to explore more disruptive approaches in the search for high-performance batteries, as predicted by Battery 2030+.

With this roadmap we aim to contribute to the development of a dynamic European ecosystem that fosters long-term, transformational research starting at fundamental TRLs gradually forming the basis for novel concepts and technologies that later can be transformed into products. To develop the required breakthrough technologies, we strongly believe in multi-disciplinary and cross-sectorial research efforts across the European battery community. Battery 2030+ has developed a chemistry-neutral approach to facilitate the invention of the batteries of the future. We create a generic toolbox transforming the way we develop, design, and manufacture batteries, which later branch out into the development of specific battery chemistries and technologies. In pursuit of this approach, we strive to develop capabilities for diverse battery technologies and build synergies in our understanding. In order to accelerate progress, we have identified three cross-cutting themes that shall be addressed. The first theme pertains to the accelerated discovery of battery materials via a fundamentally improved understanding of their functional interfaces. Within the field of material research, we think Europe can play a leading role by the development of the Battery Interface Genome and the Material Acceleration Platforms (BIG-MAP) with specific focus on designing and improving key battery components. The second theme deals with the integration of smart functionalities into batteries that will increase safety, reliability, and cycle life. Here, the development of self-healing mechanisms holds significant promise to enhance battery life-time. Finally, we believe that blue-sky research shaping new technology must consider the manufacturability aspects of batteries and, facing the challenges of a climate-neutral society, the recyclability of batteries. In conclusion, over a time frame of ten years, we will develop a circular model incorporating specific R&I actions, based on the considerations developed in the roadmap detailed above.

9 Abbreviations and glossary

AI	Artificial intelligence
AIMD	Ab initio molecular dynamics
BD	Brownian Dynamics
BIG	Battery Interface Genome
BIG–MAP	Battery Interface Genome–Materials Acceleration Platform
BMS	Battery management system
BSH	Battery self-healing
CB	Carbon-black
CEI	Cathode–electrolyte interface
CFD	Computational Fluid Dynamics
CNT	Carbon nanotube
DEM	Discrete Element Method
DFT	Density Functional Theory
EASE	European Association for Storage of Energy
EBA	European Battery Alliance
ELN	Electronic Lab Notebook
EMIRI	Energy Materials Industrial Research Initiative
EMMC	European Materials Modelling Council
Energy density	Energy per unit volume (Wh/l)
EOL	End of life
EPR	Extended producer responsibility
EPR	Electron paramagnetic resonance
EUCAR	European Council for Automotive R&D
EURAMET	European Association of National Metrology Institutes
FBG	Fibre Bragg grating
FIB	Focused Ion Beam
HPC	High-performance computing
HTS	High-throughput screening
JRC	Joint Research Centre, the European Commissions
KMC	Kinetic Monte Carlo
LBM	Lattice Boltzmann Method
LCA	Life cycle assessment
LCO	Lithium cobalt oxide
LEAPS	League of European Accelerator-based Photon Sources
LENS	League of Advanced Neutron Sources
LIB	Lithium ion battery
Li-ion	Lithium ion
LM	Liquid metal
LSB	Lithium-sulphur battery
MAP	Material Acceleration Platform
MC	Monte Carlo
ML	Machine learning
MOF	Microstructural optical fibres

NCA	Lithium nickel cobalt aluminium oxide (cathode material)
NMR	Nuclear magnetic resonance
NN	Neural Networks
NPS	Nano-plasmonic sensing
p2D	Pseudo two-dimensional
p4D	Pseudo four-dimensional
PCF	Photonic crystal fibre
PIL	Polymerizable ionic liquid
PINN	Physics-Informed Neural Network
POF	Proof of concept
QRL	Quality, reliability, and lifetime
QRLS	Quality, reliability, lifetime and safety
RE	Reference electrode
RFB	Redox Flow Battery
ROM	Reduced order models
RUL	Remaining Useful Life
SEI	Solid electrolyte interphase
SEM	Scanning electron microscopy
SET PLAN	Strategic Energy Technology Plan
SHE	self-healing electrolyte
SIB	Sodium-ion battery
SoC	State of charge
SoH	State of health
Specific energy	Energy per unit mass / Energy stored gravimetrically (Wh kg^{-1})
SSB	Solid-state battery
SWCNT	Single-walled carbon nanotubes
TEM	Transmission electron microscopy
TRL	Technology readiness level
TBMS	Thermal battery management system
XAS	X-ray absorption spectroscopy
XRD	X-ray diffraction
ZIB	Zinc-ion battery
ZIFB	Zinc-iodine flow battery

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