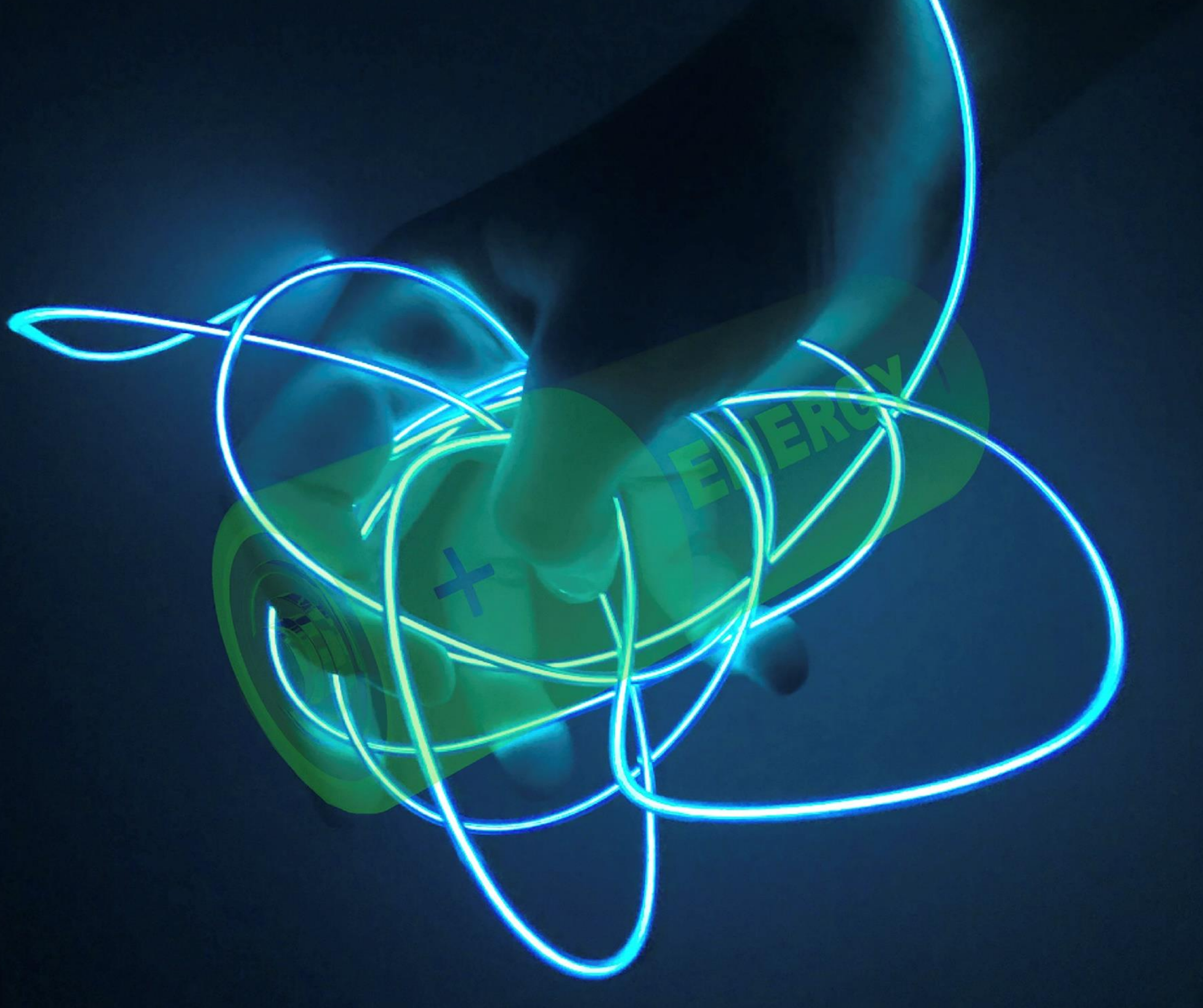
BATTERY 2030+



INVENTING THE SUSTAINABLE BATTERIES OF THE FUTURE

Research Needs and Future Actions

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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] Since then, through its projects BIG-MAP, BAT4EVER, HIDDEN, INSTABAT, SENSIBAT, SPARTACUS and the coordination and support action, 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 version of the roadmap follows the main tracks from the earlier one 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, and design battery materials, components, and systems 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.

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.

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Contents

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.[2] 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.[3]

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, 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 storage) and future emerging sectors (e.g., robotics, aerospace, medical devices, and Internet of things)

With this first update of the roadmap, BATTERY 2030+ has refined the originally expressed research directions, following actual developments[4–15], progress in the international research community as well as in the currently running ramp-up projects under the LC-BAT call. The chemistry-neutral 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)[16], meet the "sustainability requirements for Batteries in the EU"[17] 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.

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.[18] Digitalisation tools will be developed utilising the power of modelling and of AI to deliver solutions to replace classical trial-and-error approaches for manufacturing. 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.

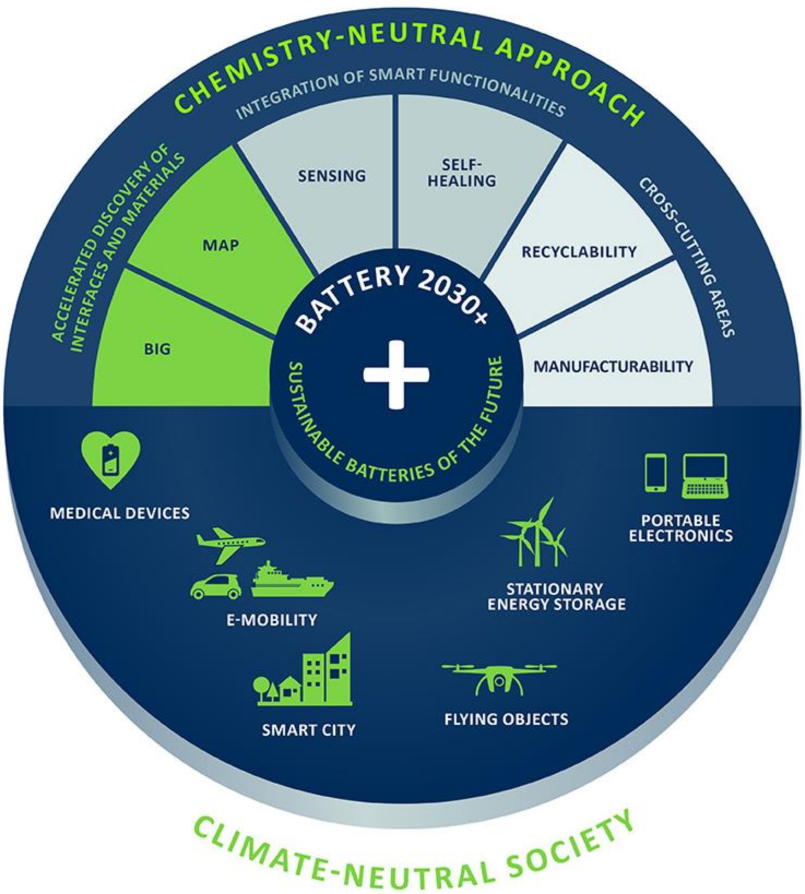


Figure 1. BATTERY 2030+: a holistic approach.

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.[19] By 2030, the EU wants to reduce its greenhouse gas emissions by 50% or more compared with 1990 levels, aiming at zero net emissions by 2050. This goal has been formulated as part of the European Green Deal[2] 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.[3]

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.[20] 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).

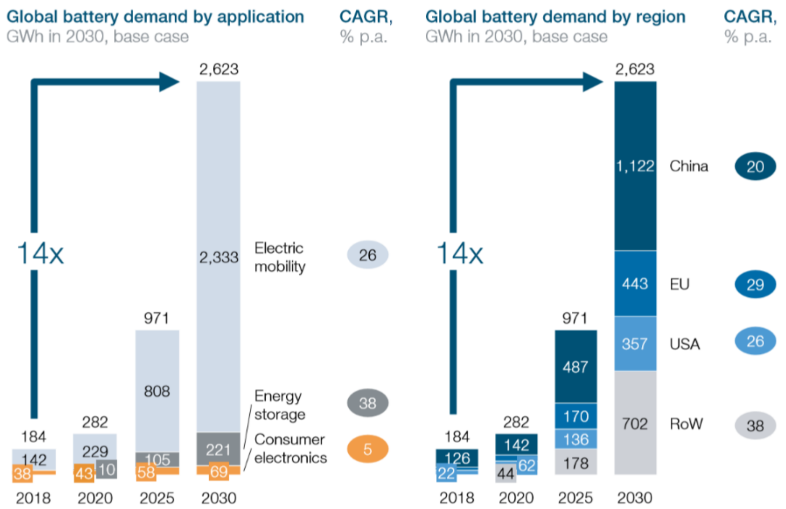


Figure 2. Expected growth in global battery demand by application (left) and region (right).[20]

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[21] 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.[22] 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.[23]

One action in the Strategic Action Plan[22] 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 Action plan on Batteries[22] and the SET Plan.[16]

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.

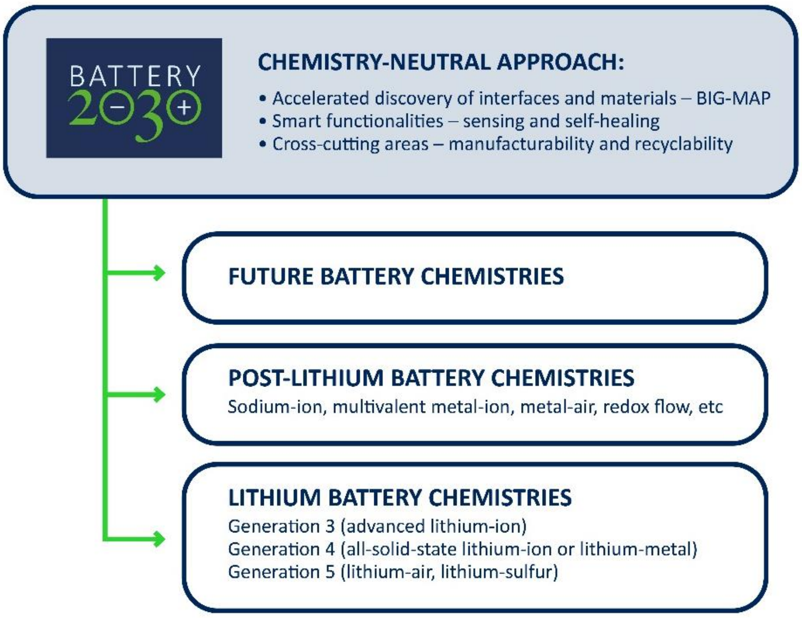


Figure 3. The BATTERY 2030+ chemistry-neutral approach will have an impact on both current state-of-the-art and future, as yet unknown battery technologies.

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 an autonomous, "self-driving" laboratory 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)**[24] to reinvent the way we perform battery materials research today. This will be achieved by creating an autonomous, "self-driving" laboratory 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, materials and interface simulations, 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)** 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 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 data infrastructure capable of performing the autonomous acquisition, handling, and analysis of data from all domains of the battery development cycle. Novel AI-based tools and physical 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.

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, reliable and long-lived batteries.

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).[25] 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 future battery technologies is addressed in this roadmap from the standpoint of the fourth industrial revolution, i.e., Industry 4.0[18] and digitalisation. The power of modelling and the use of AI should be exploited to deliver "digital twins"[26] for both innovative cell designs, avoiding or substantially minimising classical trial-and-error approaches, and manufacturing methodologies.

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).

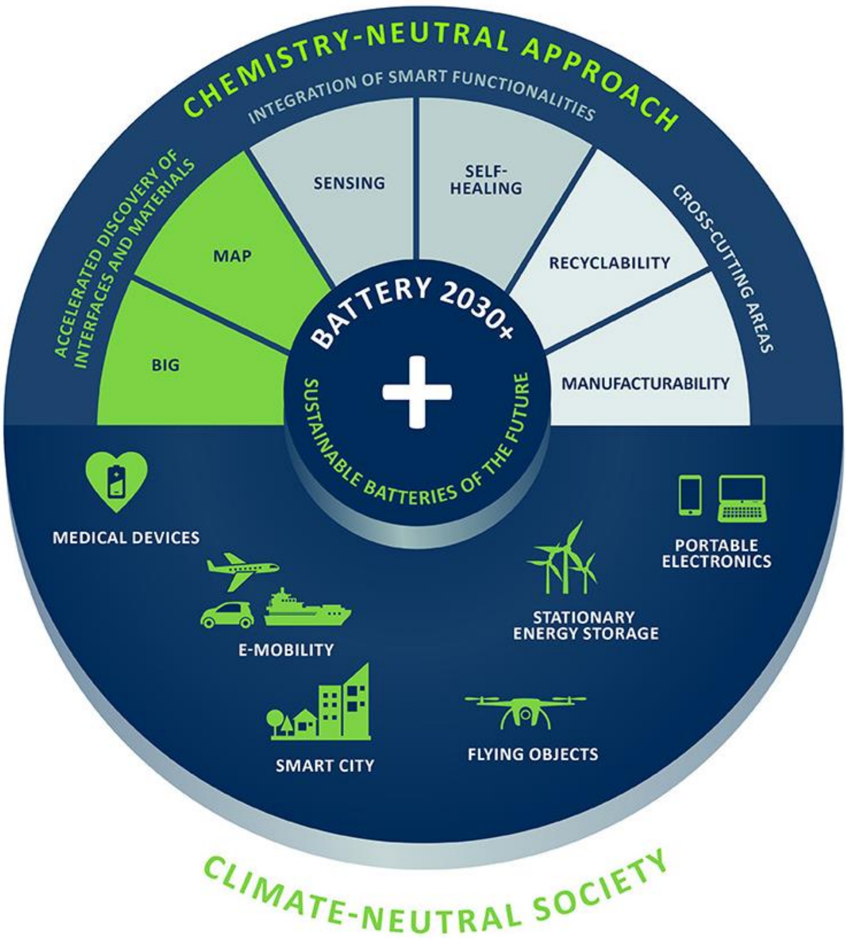


Figure 4. The BATTERY 2030+ vision is to invent the sustainable batteries of the future through a chemistry-neutral approach that will deliver ultra-high-performance batteries optimised for their intended applications, such as electro-mobility, stationary storage, medical devices, and robotics. BATTERY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

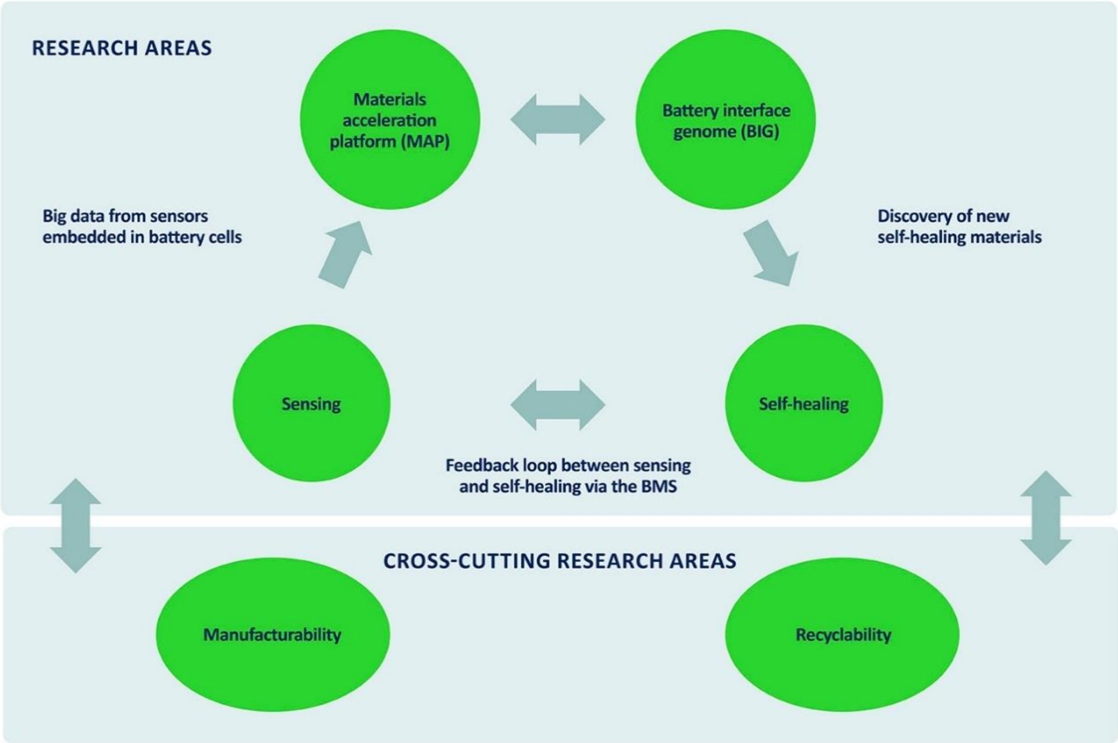


Figure 5. Interactions between the different BATTERY 2030+ research areas and thematic assignment of the six already running projects.

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, materials autonomous synthesis, and characterisation.	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	Apply non-invasive multi-sensing approaches transparent to the battery chemical environment offering spatial and time resolution.	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.	Master sensor communication with an advanced BMS relying on new AI protocols by wireless means to achieve a fully operational smart battery pack.
	Integrating sensors into existing battery components (e.g., separator, current collector, and electrode composite).	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).		
Self-healing	Establishing a new research community that includes a wide range of R&D disciplines to develop self-healing functionalities for batteries.	Integrating self-healing functionalities into battery components (e.g., separator or electrode composite).	Established efficient feedback loops between cell sensing, BMS, and/or AI modules to appropriately trigger, by external stimulus, the self-healing functions already implanted in the cell.
	Developing autonomous and non-autonomous (on demand) self-healing functionalities for specific battery chemistries, targeting loss of capacity and loss of power.	Electrochemically stable non-autonomous self-healing functionalities triggered via an external stimulus obtained from an advanced BMS.	Designing and manufacturing low-cost biosourced and/or biomimetic membranes with controlled functionalities and structure as autonomous self healing functionalities.
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.		The new concepts in cell manufacturing are transferred to the industry and academia.
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.	
	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.	
		Develop prediction and modelling tools for the reuse of materials in secondary applications.	

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 (Findable, Accessible, Interoperable and Reusable) 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, charactericastion, 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 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".[27–32] Along with their document on "Development of reporting methodologies"[33] they supplement the European Battery R&I landscape and the vision of Battery 2030+ by important strategic focus areas.[34]

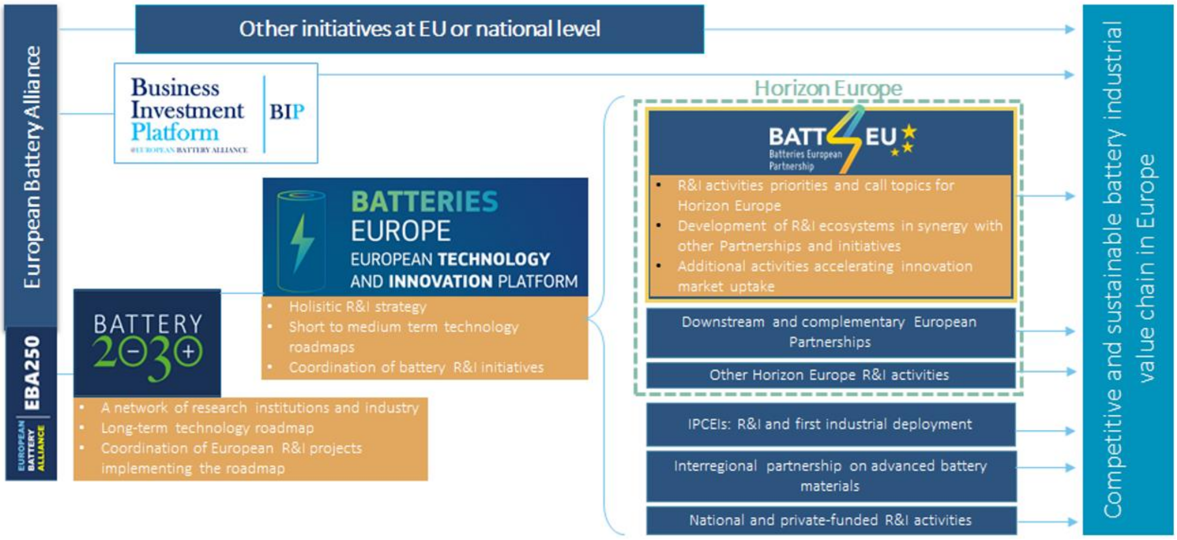


Figure 6. The battery R&I landscape in Europe.

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, 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, it is necessary to pursue a concerted approach of scientists and engineers. 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.

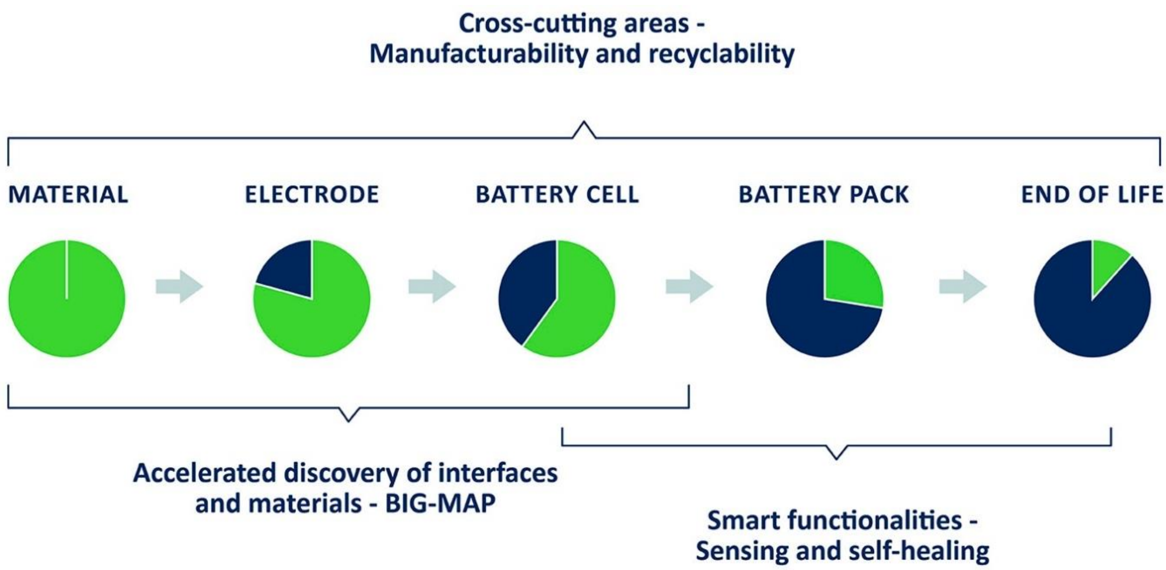


Figure 7. The decrease in total capacity as more inactive material is added when going from the material to the complete battery pack. The identified research areas will address these losses throughout the battery value chain. End of life represents the additional capacity loss due to degradation.

5.3 Impact on the European SET Plan targets for batteries

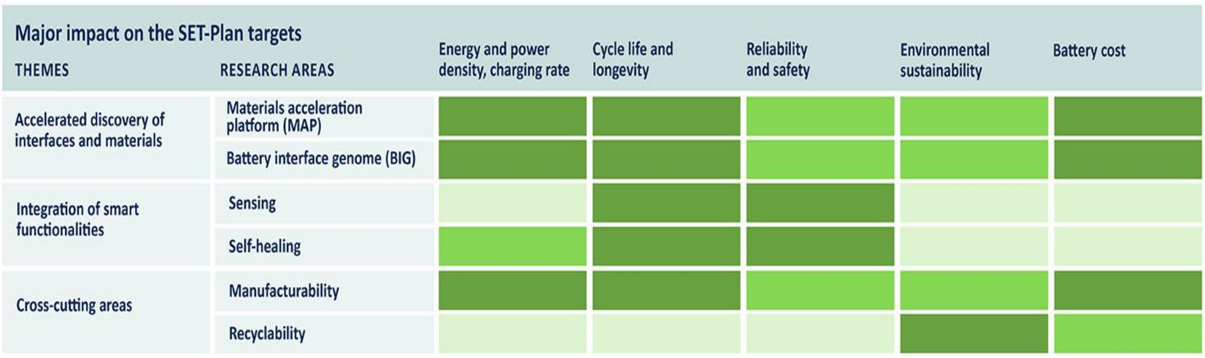
BATTERY 2030+ suggests actions pushing battery technologies far beyond the current state of the art. This will have an impact throughout the battery value chain by enabling and accelerating the attainment and surpassing of the SET Plan targets.

The integrated SET Plan Action 7[16] 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 state of the art of today's market for rechargeable batteries is dominated by lead acid and LIBs, with market shares of 49.9 % and 45.7 %, respectively.[35] Nickel-cadmium and nickel-metal hydride batteries as well as some non-rechargeable chemistries are also of commercial importance, and there are strong efforts to develop redox flow batteries (RFBs), mainly for stationary energy storage solutions. Battery technology based on other, novel chemistries is mostly still in its infancy. While sodium ion batteries have recently been commercialized and also Li-metal systems have reached maturity in some applications, batteries based on alternative mono- and multivalent ions, such as potassium, magnesium, aluminium, calcium and other chemical elements are still the subject of intense research and development efforts.

Lead-acid batteries are still being developed for several markets due to their robust performance over a wide temperature range, high recycling percentage, and low cost. From advanced lead-acid batteries, some experts expect an increased turnover over the next ten years. However, currently there is a rising potential competitor, the Na-ion battery which targets to be used as starter battery in cars. Generally, Pb acid batteries cannot compete for use in electric vehicles due to their considerably lower energy density.

LIBs are used in applications ranging from consumer electronics to electric vehicles, but also in large-scale energy storage and back-up power solutions for the grid. They have hugely emerged in the last decade, which can be attributed to their superior energy density compared to lead-acid batteries (see Figure 8), their high efficiency and their reliability. The first commercial LIBs came on the market in the 1990s. Since then, the energy density of LIBs has more than doubled[36] while the cost has dropped by a factor of about 15 or more between 1995 and 2019.[37–39] Building on this battery concept, multiple efforts are underway worldwide to further increase battery performance by developing improved storage materials and electrolytes, by optimising battery design parameters, as well as by developing more cost effective and optimised production methods.

RFBs are mainly targeting large-scale energy storage applications, for which they have technical advantages such as scalability, recyclability, long life and simple maintenance. The current state-of-the-art is based on vanadium and zinc-bromine chemistries, but issues regarding raw materials availability and corrosive electrolytes have motivated studies focusing on more ubiquitous materials and non-corrosive, aqueous electrolytes. Such new RFB concepts promise excellent levelized cost of storage (LCOS), rendering the technology an important component to Europe's technological independence and a catalyst for the broad deployment of renewable energy generation.

In line with its chemically neutral approach, BATTERY 2030+ will be open to chemistry and materials developments and discoveries, regardless the technology, that will facilitate Europe’s technological independence to develop batteries ecosystem. In this domain the technological sovereignty would facilitate the electrification of the economy, thus contribute 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.[40–43]

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[44] 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.[45] The expected cost will decline to well below USD 100/kWh by 2024,[45] 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 commercialized technologies.

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.

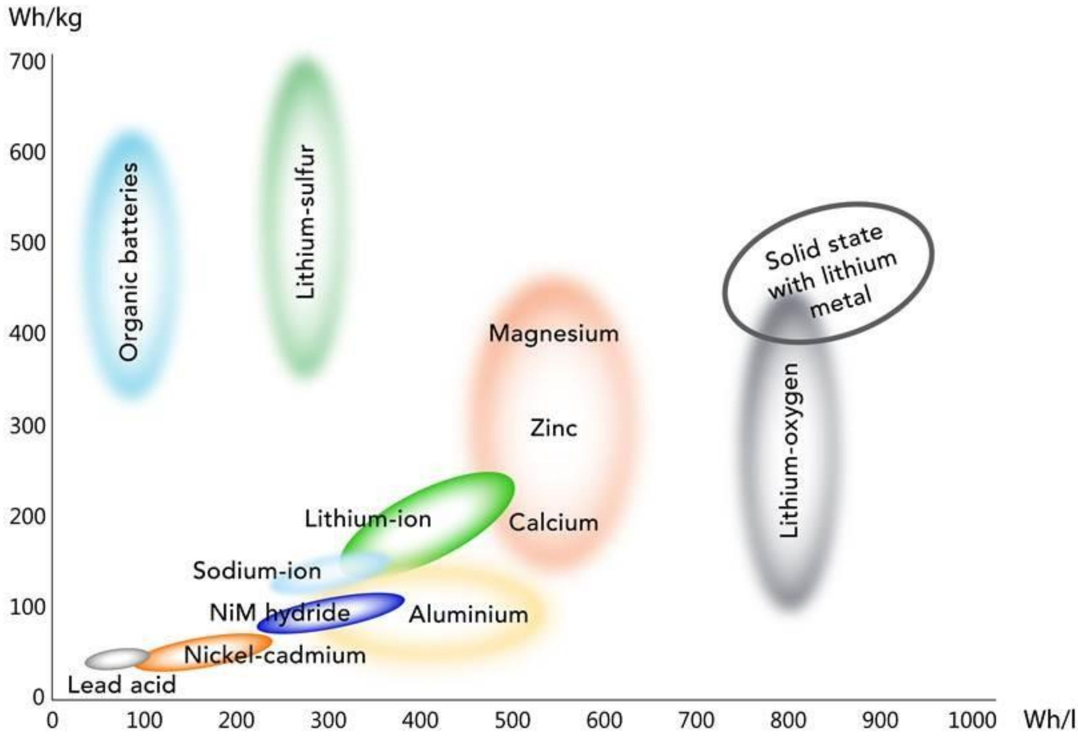


Figure 8. Current commercial batteries and targeted performance of future possible chemistries. The post lithium batteries chemistries are given as names indicating all kinds of metal-type batteries in respective category. There is a large uncertainty of their respective position in the graph. NiM hydride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

Several associations and countries have published roadmaps for batteries or for energy storage including batteries. Some recent roadmaps are from: ETIP[27–32], EASE[46], EMIRI[47], EUCAR[44], implementation of the SET Plan Action 7[23], JRC[48–51], China (Li, H., Ouyang, M., Zhan, M. 2019), Finland[53], India[54,55], Japan[56,57], and the USA.[58]

Some international targets for automotive batteries expected for the coming years are shown in Figure 9.(Courtesy of Prof. Hong Li 2019) 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.

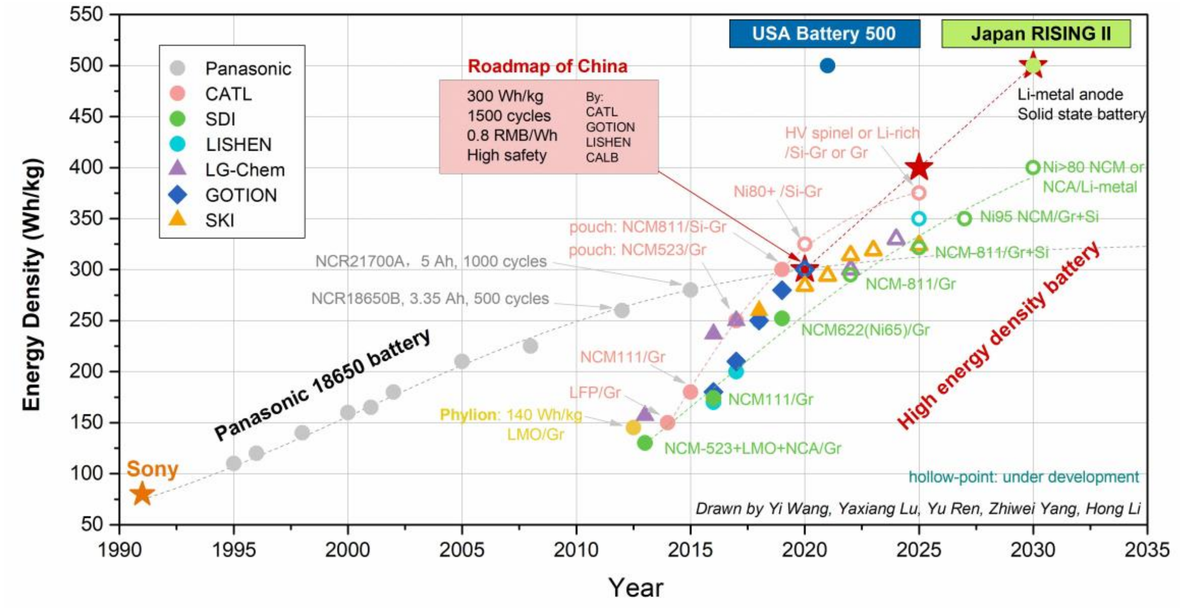
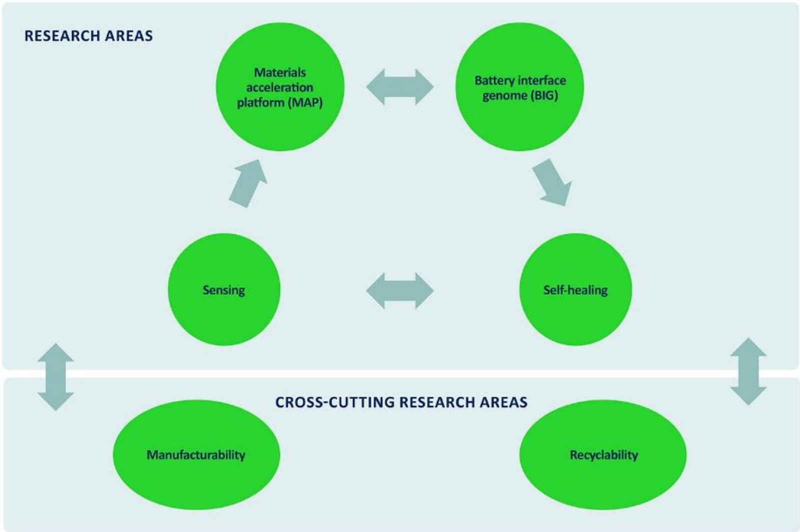


Figure 9. Comparison of the gravimetric performance of different batteries for automotive applications. The targets from the SET Plan coincide with the green line (different NCM-based generations of lithium-ion batteries). Japanese Rising II follows targets similar to those of the SET Plan, while China's 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.[60]

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.** 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+.[4] 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.[61] 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 and development crosscuts the entire clean energy technology portfolio, ranging from energy generation, conversion and storage to delivery and end use. Advanced materials are the foundation of nearly every clean energy innovation, particularly for emerging battery technologies. Relying on existing trial-and-error–based development processes, the discovery of novel high-performance battery materials and cell designs entails considerable effort, expense, and time – traditionally over ten years from initial discovery to commercialisation. In BATTERY 2030+, we outline a radically new path for the accelerated development of ultra-high-performance, sustainable, and smart batteries, which hinges on the development of faster and more energy- and cost-effective methods of battery discovery and manufacturing.

In this section, we outline the opportunities, challenges, and perspectives connected with establishing a community-wide European battery **Materials Acceleration Platform** (MAP)[62], to be integrated with the **Battery Interface Genome** (BIG) described below. The emerging BIG–MAP infrastructure is modular and highly versatile, in order to accommodate all emerging battery chemistries, material compositions, structures, and interfaces. Drawing inspiration from initiatives like the Materials Genome Initiative[63] and following the format of Mission Innovation: Clean Energy Materials (Innovation Challenge 6) MAP Roadmap,[24] and further conceptual iterations[64], a MAP utilises AI 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.

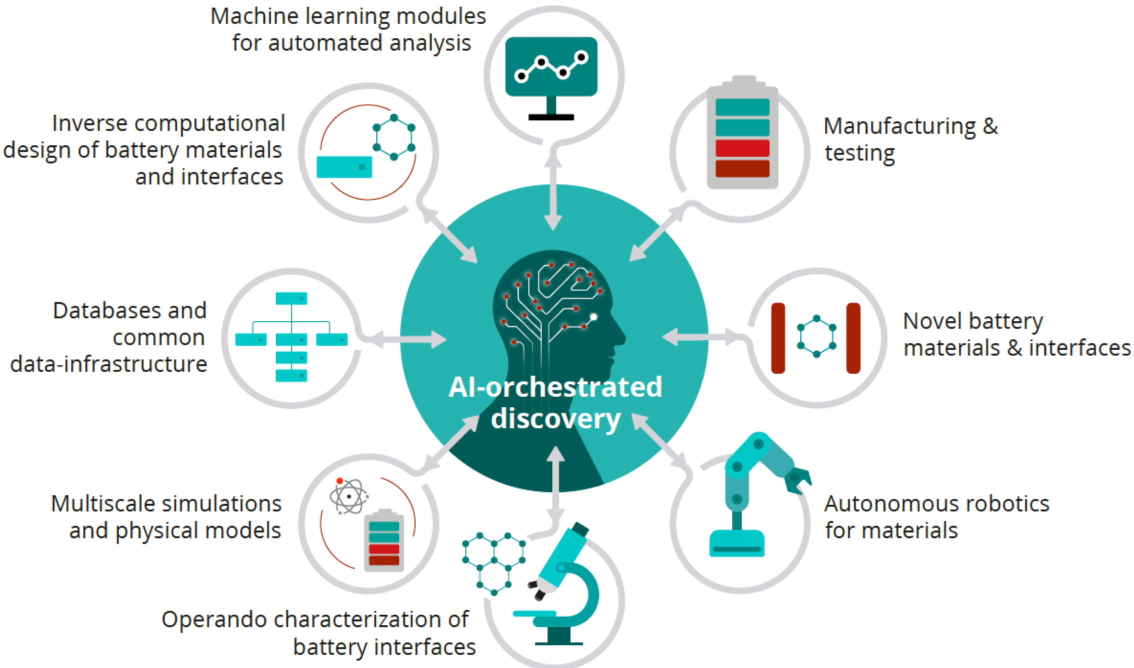


Figure 10. Key components of establishing a battery MAP.

Realising each of the core elements of the conceptual battery MAP framework entails significant innovation challenges and the development of key enabling technologies. Combined, these technologies enable a completely new battery development strategy, by facilitating the inverse design and tailoring of materials, processes, and devices. Ultimately, coupling all MAP elements will enable AI-orchestrated and fully autonomous discovery of battery materials and cells with unprecedented breakthroughs in development speed and performance.[14]

Successful integration of computational materials design, AI, modular and automated synthesis, robotics, and advanced characterisation will lay the foundation for dramatically accelerating the traditional materials discovery process[65]. The creation of autonomous, "self-driving" laboratories[66] capable of designing and synthesising novel battery materials, and of orchestrating and interpreting experiments on the fly, will create an efficient closed-loop materials discovery process. Its implementation, presently pursued in Europe in the context of the BIG-MAP project[67], constitutes a quantum leap in materials design, which can be achieved only through the integration of all relevant European expertise.

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 many 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.[11]

In recent years, several examples have emerged in which the close integration of virtual (typically atomic-scale) computational material design and in operando characterisation[68] techniques in a circular design loop can accelerate the discovery cycle of next-generation battery technologies, such as high-capacity Li-ion cathodes[69] and materials for secondary metal–air batteries,[70] but further acceleration is needed to reach the highly ambitious goals of BATTERY 2030+. Ideally, such a circular materials development process will integrate experimental and theoretical research in a closely coupled development platform that enables near-instantaneous cross-fertilisation of the results of complementary techniques. In the following sections, we summarise the state of the art in key areas of MAP.

Interoperable data infrastructures 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[71] and multi-sourced data from different scales and domains, such as experiments, testing, and modelling. A large number of ongoing efforts in Europe and beyond aim to create extensive, flexible, and sharable databases and repositories[72,73] for experimental data. Additionally, computational infrastructures such as PRACE and EuroHPC, and platforms such as ASE, SimStack,[74] AiiDA,[75] and Materials Cloud[76] facilitate efficient and reliable high-throughput calculations[13], while only few examples like the OPTIMADE[77] REST API bridge computational and experimental data. At present, no data infrastructures exist that are 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),[78] 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[79] is under development in Battery Interface Genome–Materials Acceleration Platform (BIG-MAP) that will 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 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 Data Management Plan (DMP) has been established to coordinate these efforts and ensure a linkage between data and tasks of the project[61]. To make it more operational, the DMP is being interconnected with BattINFO and, ultimately, should connect all the projects under the BATTERY 2030+ umbrella.

Multiscale modelling: Battery performance and lifetime are determined by many processes that occur on vastly different time and length scales.[80] Simulating batteries 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. A single computational model of virtual materials design that encompasses all these phenomena is beyond the limits of current computing power and theory, but advances in machine and deep learning models and explainable AI (AIX) provides new possibilities for autonomous parameterization and advanced multi scaling.[81,82] Traditional single-scale models must be combined to form multi-scale workflows[83], for example, through generative deep learning. An overview of the potential impact of these techniques is given in Bhowmik et al.[9]. Multi-scale modelling techniques are currently being developed, for example, to optimise real and virtual electrode microstructures[84] and to study the effects of the fabrication process on cell performance[85] and electrode surface film growth.[86]

Experimental characterisation of materials and interfaces at large-scale research facilities, such as synchrotron and neutron scattering facilities, plays a critical role in ensuring sufficient acquisition of high-fidelity data describing battery materials and interfaces.[6] This calls for the ability to perform autonomous, on-the-fly analysis of the vast amounts of data generated at laboratory, synchrotron, and neutron facilities across Europe, and to develop integrated and standardized multimodal workflows including correlative analysis of multi-scale multi-techniques data. The state of the art of the most relevant structural and spectroscopic characterisation techniques related to battery materials and interfaces is discussed in detail in Section 4.3.

Autonomous synthesis robotics,[8] which can be controlled and directed by a central AI, are 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,[87,88] and examples are also emerging in the development of solids and thin-film materials.[89–91] For energy storage materials, robotic-assisted synthesis and automation have opened the field to the high-throughput screening of functional electrolytes and active materials constituting anodes and cathodes. In combination with computational approaches such as data mining and the correlation of structure–property relationships with the performance of battery active materials, robotics has had a significant impact on the discovery of novel and promising materials.[87] A key aspect is the transformation from automation to autonomy in both synthesis and characterization.

Experimental and computational high-throughput screening of large compound libraries for activity in the accelerated formulation of relevant battery materials[8,13] via the use of automation, miniaturised assays, and large-scale data analysis can accelerate materials discovery by up to one order of magnitude.[92,93] On the computational side, workflows have been developed to automate different steps of the calculations needed to screen for new compounds.[94] Several examples of fully automated high-throughput screening (HTS) systems for electrolyte formulation, cell assembly, and selected relevant electrochemical measurements are now available.[95]

AI in materials discovery offers great prospects,[96] 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.[65] 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[97], Phoenics[98] and Olympus[99] 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.

7.1.2 Challenges

Availability of FAIR and curated data: The development of predictive models to design future batteries 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[100] of battery materials and interfaces requires the integration of high-fidelity data[101] covering complementary aspects of the material 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.

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).[102]

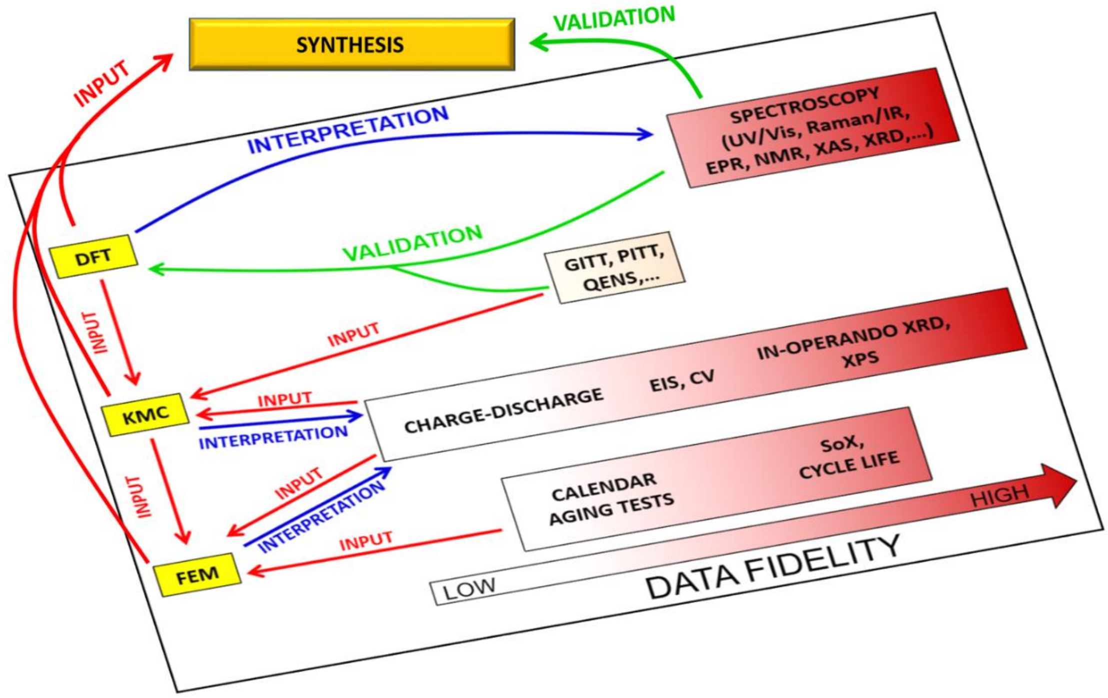


Figure 11. Illustration of the data flow between representative experimental and theoretical methods for studying battery interfaces. The fidelity of each method is generally proportional to its cost, but the fidelity–cost relationship can be optimised by acquiring data only when the given method/data is most valuable (adapted from[102]).

While machine learning could potentially massively accelerate the screening and identification of, for example, the structure–property relationships of inorganic energy materials,[103] a key challenge in the discovery of battery materials and interfaces is the development of autonomous workflows[13] for extracting fundamental relations and knowledge from sparse datasets[104] 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[66,90] and, more importantly, automated approaches for characterising battery materials and cells are either lacking or dramatically underdeveloped. Several machine-learning–based tools have recently been developed for a number of relevant characterisation techniques, for example, XRD and XAS.[105,106] 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.

An important bottleneck in closed-loop discovery is the lack of robust and predictive models of key aspects of battery materials and interfaces. 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[107] with automated synthesis and characterisation technologies will enable the envisioned breakthroughs required for the implementation of fully autonomous materials discovery.[102]

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 both a unified terminology as well as 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.[108,109] The adoption of a unified ontology will be enabled and facilitated by the implementation of Electronic Lab Notebooks (ELNs). Ontologies and standards must eventually be made available for use by the scientific community. By their broad application, data will be made fully FAIR. For BIG-MAP, one main goal to be reached are well-defined and standardized interfaces which will then enable full reproducibility and interoperability. At this point the Electronic Lab Notebook (ELN) could represent one step to the Lab as a Service (LaaS).

7.1.3 Advances needed to meet challenges

European strongholds in the battery community have always been in 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 Faraday Institution in the UK, and the CELEST, MEET and POLiS 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 robotics: The comprehensive electrochemical characterisation of battery materials and testing on the cell level are among the major bottlenecks slowing the development of new battery materials and interfaces.[6,8] To explore larger classes of materials in the context of specific applications, 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.

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,[110–112] many electrochemical tests do not work on short time scales; in particular, cycling experiments can take days to months or even years.[113] 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 cells has to be established, including the development of versatile multimodal cells, standardized cycling protocols and sample transfer methods. This infrastructure must address the issues of width and depth, and should include filtration by identified lead candidates. The combination of physics-guided data-driven modelling and data generation is required to enable the high-throughput testing of batteries and their incorporated active materials in the future, and thus to develop 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.[88,114] 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. The establishment of a common data infrastructure will help to 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:[6,13] 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 efficient 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 sacrifice of detail and resolution. Releasing the full potential of multi-scale modelling, multimodal characterization, and inverse design to support new materials radically new approaches to link scales beyond the state of the art that can be achieved by isolated research communities in individual countries.[100] 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[115] 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 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 find it important to codify the workflows to a degree that they can be used outside the group which have developed them via accessible App-stores. In addition to purely computational workflows, workflows that integrate on-the-fly experiments (and vice versa) hold a huge potential to accelerate materials discovery. 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. While presently most simulation efforts are directed towards understanding of battery function, with an increasing emphasis on design, additional efforts are needed to develop models to address the full battery life-cycle.

AI exploitation: AI-based generative models,[116] 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 are substantial gaps in the model spectrum that preclude the development of comprehensive battery models. These can be closed by AI-based techniques, but these are typically unaware and thus may violate physical laws. The key to overcoming this dilemma is the development of hybrid models in which the prediction space of AI-based models is constrained by laws of physics or in which AI is used to adapt physical models. These models must be trained on large curated datasets from advanced multi-scale computational modelling, materials databases, the literature,[117] and operando characterisation. These data must span all aspects of battery materials from synthesis to cell-level testing.[102]

Unification 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.[118] 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.

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 data 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 uncertainty-based hybrid data-driven and physical models of materials and interfaces. These developments will be supplemented by establishing the necessary ontologies and standards.

***In the medium term*:** Implement BIG in the MAP platform (BIG–MAP), capable of integrating 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 pre-emptive launch of the developed self-healing additives. Demonstrate transferability of the BIG–MAP approach to novel battery chemistries and interfaces, e.g., to 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 coupled experimental-computational workflows. 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)

Past experience has shown that when developing new battery chemistries or introducing new functionalities into an existing battery technology, interfaces hold the key to exploiting the full potential of the electrode materials and to developing ultra-high-performance, sustainable, and smart batteries. The European battery R&D landscape consists of a multitude of research institutions, laboratories, and industries, many of which pursue complementary approaches to tackle this challenge at a local scale. We will bring together this expertise with cross-sectoral competences, industrial partners, and end users to establish BIG and accelerate the development of radically new battery technologies.

Current research methodology relies largely on incremental advances at the local scale, which are not pertinent for tackling the ambitious challenges outlined in this roadmap. MAP will provide the infrastructural backbone to accelerate application of our findings, while 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.[119] In this respect, we must take into account studies of ion transport mechanisms through interfaces and, even more challenging, visualise the role of the electron 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 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 physical and data-driven models. Thereby integrating dynamic events at multiple scales, e.g. across the atomic micron scales. In this respect, we must consider studies of ion transport mechanisms through interfaces and, even more challenging, visualize 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[6] 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,[120] 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 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, AI, and systematic multi-technique characterisation of battery interfaces, including in operando characterisation, to generate/collect comprehensive sets of high-fidelity data 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. 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 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 graphitic anodes is one of the most crucial properties in ensuring the cycling stability 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 are currently capable of providing a full description of the events happening at the interface.

Experimental and computational techniques have a challenge of being both surface and interphase sensitive. Thus, no singular technique is currently capable of providing a comprehensive description of events happening at the interface. 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 interfaces upon cycling. The most prominent approach is the use of electrolyte additives that react inside the cell during initial operation, and of coatings that can passivate the surface of electrode materials and thus prevent reactivity with the electrolyte. However, many years of Edisonian trial-and-error research have demonstrated the need to use several additives working in synergy to results in an effective electrode-electrolyte interface. Accelerated development of such an interphase would greatly benefit from high-throughput techniques and the AI-assisted rationalisation.

Physics-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 idealized physics/chemistry-based simulations and data on real materials. Interoperability and scale-coupling is 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 big 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 include usually coupled multistep reactions.[121,122] These multistep reactions are often either oversimplified or the reaction steps are modelled in too ideal environments.[123] In specific cases, it is possible to combine DFT methods with classical approaches to improve the description of surface reactions,[124] 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 specific characteristics of the composition of the electrolyte, 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 need to include experimental conditions. Similarly, the experimental conditions should be made as reproducible and exact (i.e. ideal) as possible 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. 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 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.[125] 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 the large datasets. These could feed the physics-aware data-driven hybrid models that will help to better understand and predict interfacial properties and will 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[126,127] for interface/interphase development and dynamics. This has the potential to lay the foundation for the inverse design of battery interfaces/interphases[102], for example, using region-based active learning algorithms.[128]

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[126] and generative deep learning models,[129] is crucial for controlling and improving the fidelity of the predictive design of interfaces. Simultaneous utilisation of data from multiple domains, including data from apparently failed experiment,[130] 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 in 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[131]. Likewise, the interpretation of experimental results can be done with higher precision if theoretical models can be used in combination with experiments.

To develop the battery interface genome, high-quality/high-fidelity data and insights are required, which calls for the development of superior in operando experimental techniques for establishing atomic-level understanding on smaller scales and on 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, as well as common European battery standards for data acquisition and transfer that could serve as worldwide standards.

In addition to the continuous improvement and 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, radical 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, and transfer reactions. Advanced physics-based hybrid models and simulation techniques have to be used for the interpretation of cutting-edge in operando experiments. Efficient methods for using the 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.[132] 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.[133,134] Implementing such European data infrastructure would require the 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.[6] High-throughput experiments should be designed to allow investigation of a large number of samples at great comparability and reproducibility alongside 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.

Standardized 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. For that purpose, guidelines should be defined, becoming the project's characterisation quality label. This checklist should be aligned and complement previously published ones by either scientific journals or other large-scale initiatives recently published, in order 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 in order to certify the performance of batteries, helping better integrate academia and industry. Therefore, efforts towards standardization 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, as well as 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, needless to say that 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.[80] 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 owing to the variety of the involved phenomena. 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 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 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, in which 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 can then be reduced to determining a finite set of parameters.

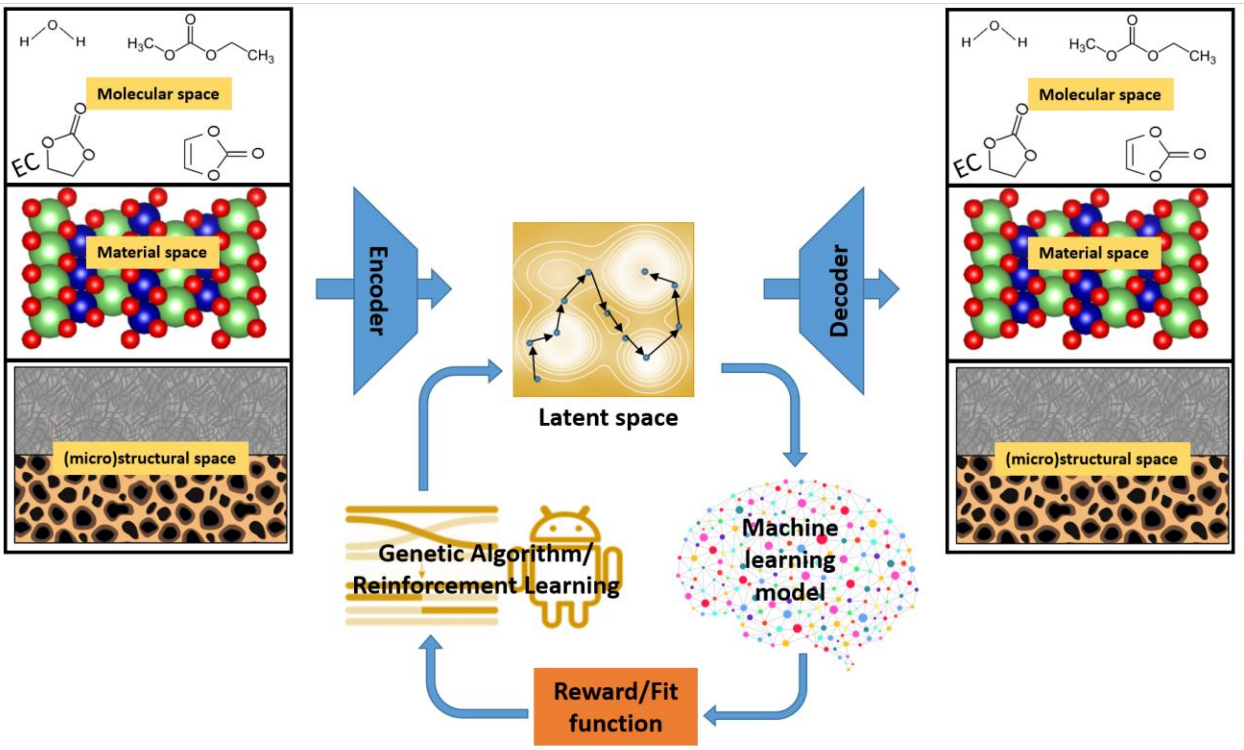


Figure 12. Generative model of interphase design. Variational auto encoder (VAE)-based encoding and decoding of chemical and structural information on a battery interphase into latent space, to enable generative battery interphase design through the use of, e.g., genetic algorithms or reinforcement-learning–based exploration[102]. Reprinted from Energy Storage Materials[102]

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 autonomous modules and apps for 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 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 the time and length scales of electron 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).[14,15,135] 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.[14]

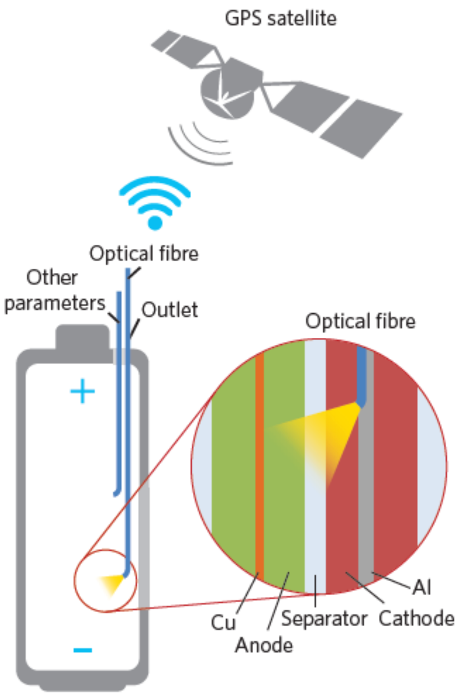


Figure 13. A future battery with an output analyser connected to sensor (optical fibres, wires, etc.) in addition to the classical positive and negative electrodes.

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.[136] 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.[137] 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).[138] 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)[139].

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.

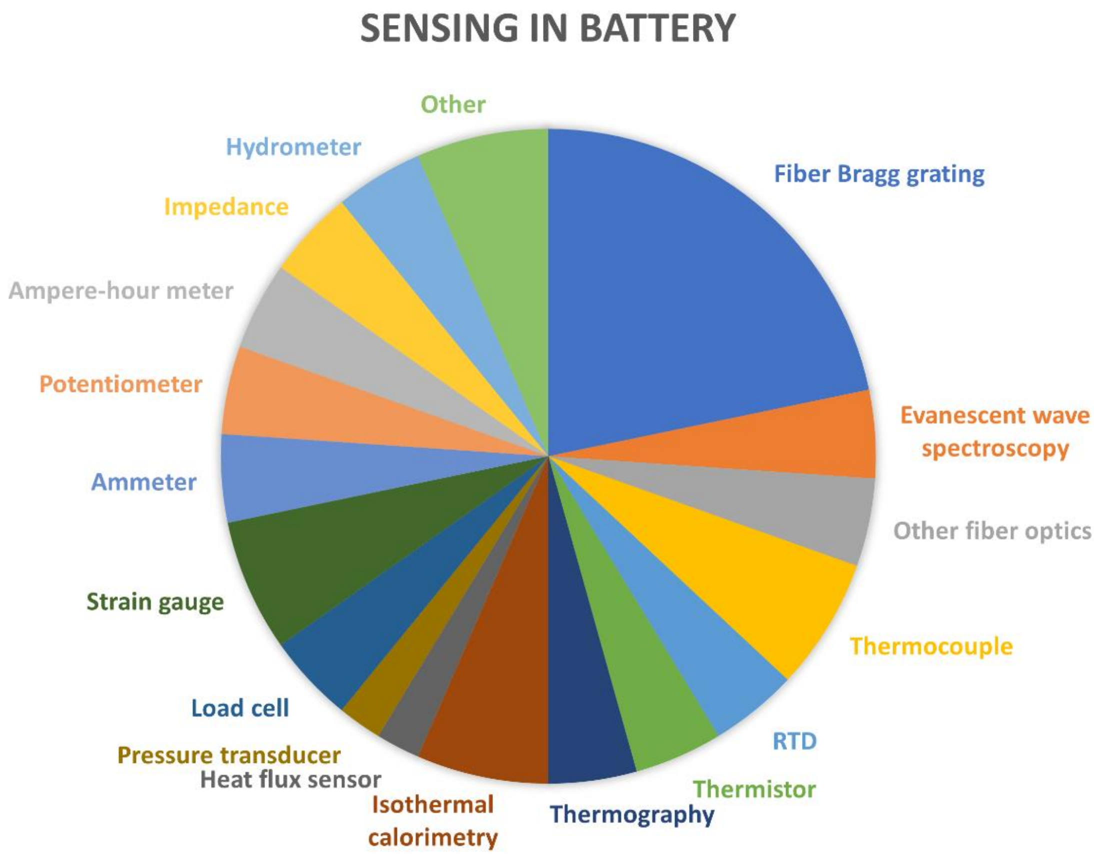


Figure 14. A glance at available sensing technologies for battery modules and systems.

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.[140] 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.[141] 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.[142] 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.[143] 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 that can be used in voltammetric/amperometric and/or potentiometric detection regimes. Reference electrodes

(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.[144] 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.[145] 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.[146] Furthermore, Rayleigh sensors, unlike FBGs, can provide axial resolution, in addition to being less expensive to manufacture.[147] 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).[148] 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.[149] 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.[150] **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 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)[139].

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:

***In the short term*:** At the battery cell level, develop **non-invasive multi-sensing approaches** relying on various sensing technologies and simple integration that will be **transparent to the battery chemical environment** and will offer feasible in vivo access to different relevant phenomena (e.g., interface dynamics, electrolyte degradation, dendritic growth, metals dissolution, and materials structure change). 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. 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. On the side of sensors, development of specific sensor dedicated to capture the internal phenomenon on the battery cell, stable over a wide range of condition (temperature, pressure, chemical concentration, electric field).

***In the medium term*:** 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. 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.

***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.

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.[151–158] 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).[159,160] Detection of irreversible changes (sensing) is a first step towards better QRLS[15]. 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?[161] 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."[162] 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,[163,164] 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).[165–169] 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.

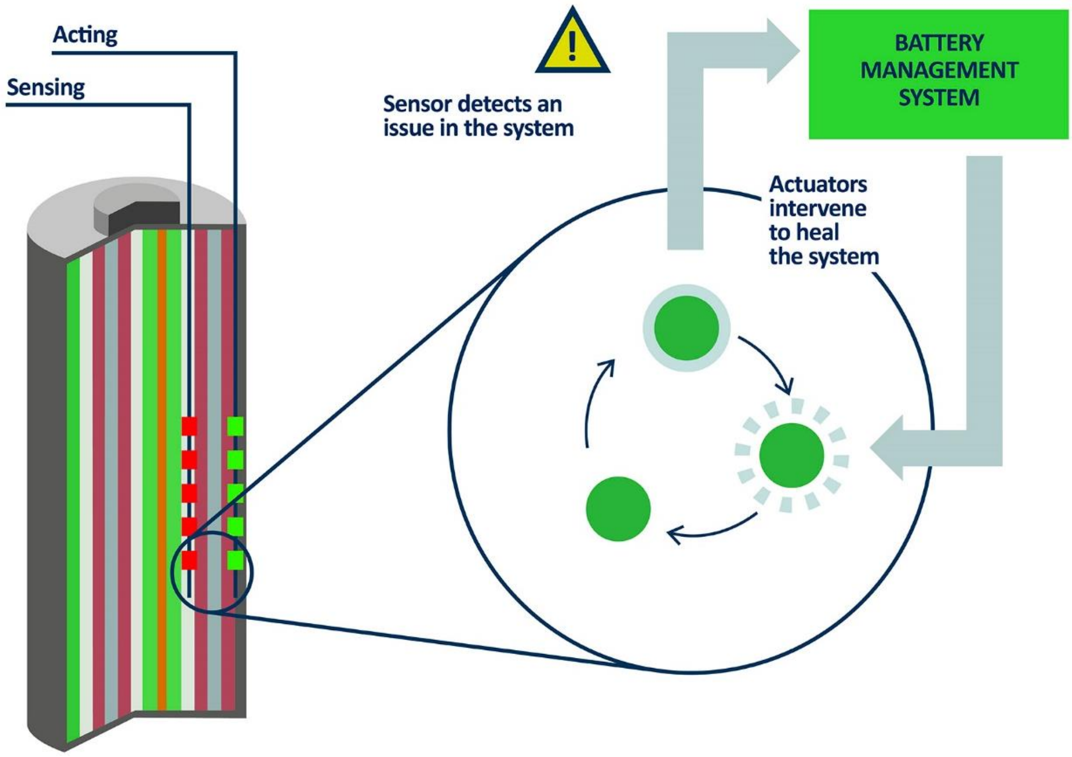


Figure 15. The synergy between sensing, BMS, and self-healing.

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[170]. 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[171,172].

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 in 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 aging processes of a chemical, electrochemical and mechanical nature are closely linked.[15]

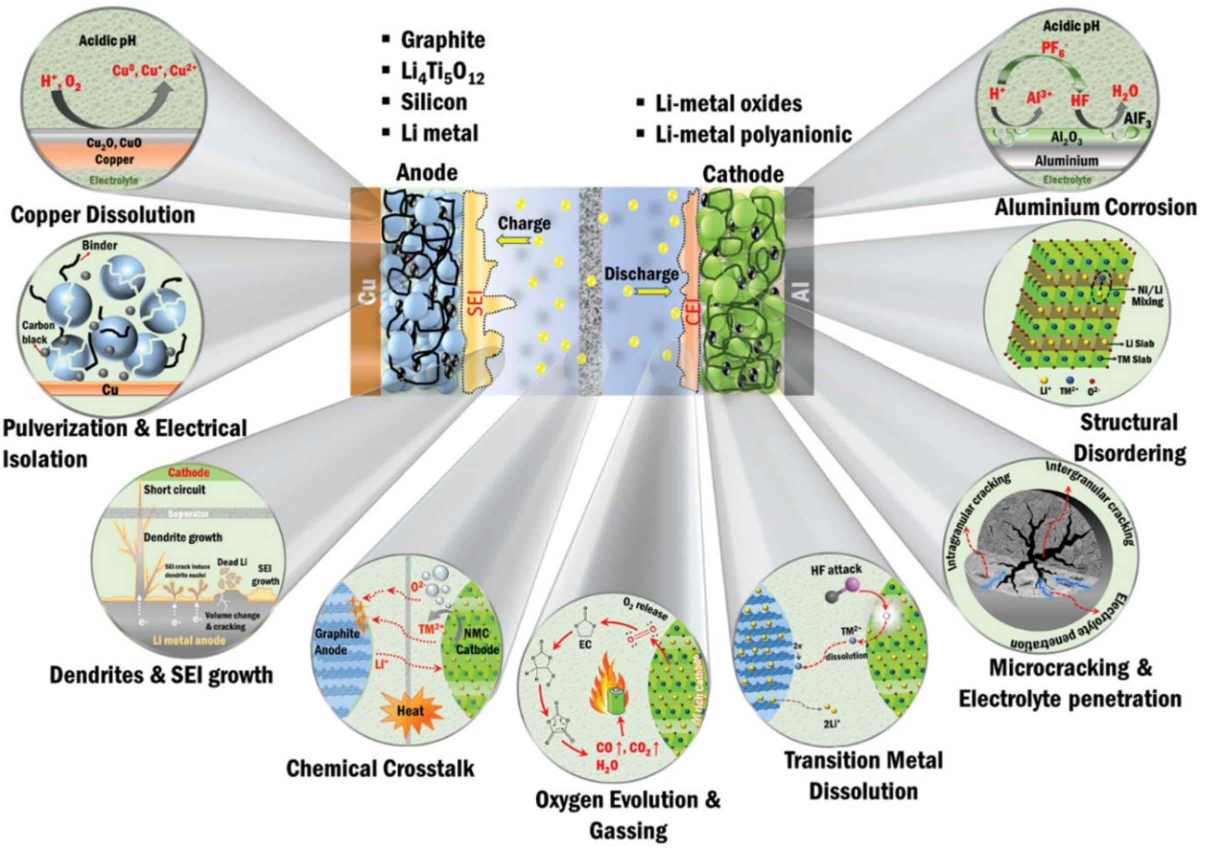


Figure 16. Overview of the major degradation mechanisms in Li-ion batteries.[15]

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[173]. Significant advances have been achieved in the field of thermo-switchable polymers with thermal self-protection integrated into the electrolytes and current collectors.[174–176] A plethora of self-assembling materials [177–180] 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 fiber-reinforced polymer composites, self-healing coatings, self-healing cementitious materials, self-healing ceramics[15], self-healing organic dyes, self-healing concrete molecules, and many others.[181–183]

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.[184]

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^+ [185] 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.[15,186,187]

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.[15]

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).[173,188]

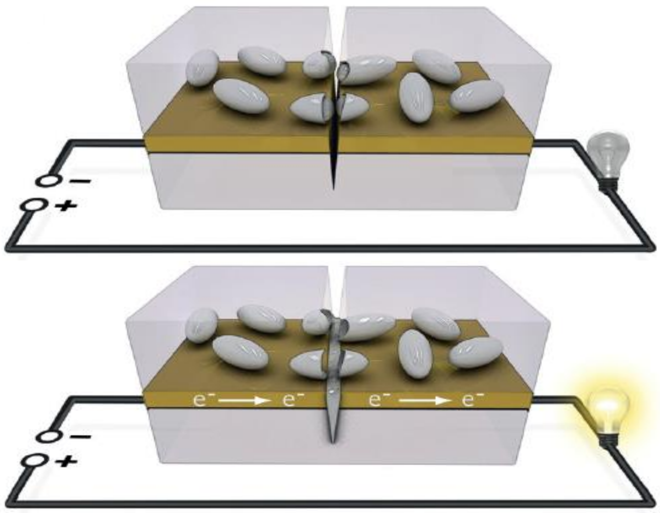


Figure 18. Testing self-healing of the gold line after damage.[173]

Other conductive chemical systems, such as carbon-black (CB) dispersions, were similarly encapsulated and tested.[189,190] 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.[172,191,192] 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.[193] Besides hydrogen-bonded polymers, self-healing binders based on several other supramolecular interactions have also been employed for Si anodes[194–198] and sulphur cathodes.[199] Long-term testing is sorely needed to fully evaluate the practicality of these approaches.

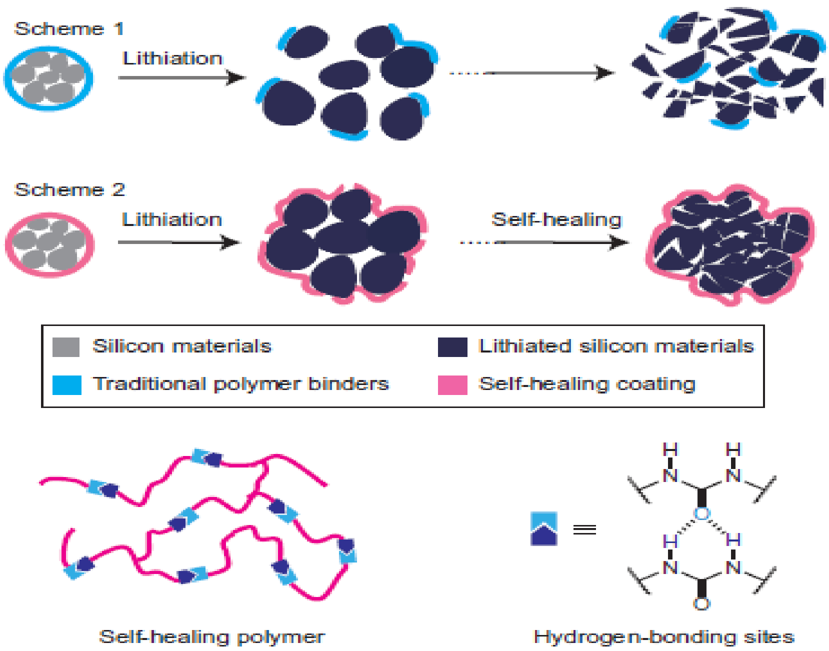


Figure 19. Design and structure of a self-healing silicon electrode.[188]

Another auto-repair concept developed by Deshpande et al.[200] 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[201] 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.[202] 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.[203] 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,[179] 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[204]. 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.;[205] 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.[206] To further improve the efficiency of Li-S batteries, Peng et al.[207] from the working group of Prof. Zhang, 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.

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[208]. 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.[209]. 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})[210]. 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 aging. 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 aging 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, there is no coherent European research effort to explore BSH in spite of the foreseen emerging opportunities that could give Europe worldwide leadership. 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.

***In the short term*:**

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. 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.

***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. 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*:** Design and manufacture low-cost bio-sourced membranes with controlled functionalities and porosity for ion detection and regulation, mimicking channels made by proteins from life science. 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.

7.5 Cross-cutting area: Manufacturability

Battery manufacturing is a topic covering a large area. Depending on the actual context, it may refer to individual cells, cell modules, or battery packs. Therefore, in this section it is of particular interest to properly set the reference scenario. The battery cell is the smallest and most fundamental functional element in the battery value chain that gathers the essential materials, components, and features of a given "battery technology". Any superstructure made thereof—modules and battery systems—basically comprises the engineering solutions to make such cells work in a practical environment. The present section will be focused on the cell level.

In this section, we will apply the criterion that any material or component that inherently takes its final form and function during or after its integration in the cell will be considered part of the battery manufacturing process. An example of this is polymer electrolytes for solid-state batteries cast from melt during the battery manufacturing process. From this perspective, this section relates to the synthesis of innovative/breakthrough materials (see Section 7.1) and to the interfaces created inside the battery in the manufacturing process (see Section 7.2). Furthermore, we want to introduce the cross-sectional concept of remanufacturing, as an industrial process to transform a used battery component into a quasi-new condition or to improve the functional conditions. This will have a future impact on the design of new cell concepts and battery modules.

The development of new materials with different properties and processing needs and requirements, along with the integration of new features such as sensors and materials with self-healing properties, will require a significant rethinking of cell design, including remanufacturing issues as previously stated. The redesign of cell architecture is essential to drive both competitiveness and sustainability, while maintaining or even increasing the energy density, and will play a central part in this work.

The availability of a new generation of breakthrough battery materials will create a new world of opportunities for innovative battery technologies. These new battery technologies will need to undergo at least two main validation phases: first, they will need to prove their potential at the prototype level, and second, the feasibility of cost and energy-efficient upscaling to the industrial process level will need to be assessed. The approach will be useful at both the prototype and industrial manufacturing levels, and also covers cell design, understood as a necessary step between innovative materials and actual battery technology.

Manufacturing of future battery technologies[7] is addressed in this roadmap from the perspective of Industry 4.0 and digitalisation and in conjunction with the accelerated materials discovery and interface design in BIG–MAP as well as self-healing and sensorisation.[211] The power of modelling and of AI will be exploited to deliver digital twins both for innovative, breakthrough cell geometries, avoiding or substantially minimising classical trial-and-error approaches, and for manufacturing methodologies. Fully digital manufacturing analogues will allow the understanding and optimisation of parameters and of their impact on the final product. These virtual representations can be manipulated (e.g., simulation and optimisation) and will therefore actuate the physical world supporting greater control of battery manufacturing facilities and production lines.

To facilitate the connection between BIG-MAP and manufacturing to work, interfaces need to be created between the different research areas that allow for an efficient exchange of FAIR data and metadata. The development of RDM infrastructure (section 4.4.2) 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 parts 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

LIBs are today's state-of-the-art high-energy battery technology for various mobile applications, including portable electronics and electric vehicles.[48,212–215] Other commercial battery technologies exist as well (e.g., lead acid, redox flow, Na-S)[216], with new technologies yet under development,[217] 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 issues.

Cell design

Today, most cell designs are based on three main formats: cylindrical, pouch, and prismatic hard-case. In detail, these geometries are based on certain standards (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, and mandrel integration in cylindrical cells) ensure steadily increasing energy densities and quality.

Extensive research has been conducted in literature, both by experimental and modelling techniques, to shed light on the effect each cell parameter has on the cell performance. Most of the physics-based models used for battery cell optimization still rely on the Doyle's pseudo two-dimensional (P2D) approach,[218] combining the porous electrode theory proposed by Newman et al. [219] and the concentrated solution theory. The P2D model has been used to optimize cell components such as the cathode and anode thickness, porosity, particle size and many other important electrode parameters. [220–225] Surrogate models based on Machine Learning (ML) or reduced order models (ROM) have the potential to reduce the computational burden of battery design by several orders-of-magnitude.[226–230] Multiphysics multiscale models are necessary tools for accelerated understanding, design optimization, and design of automatic control of battery cells, as they allow for the analysis of an almost unlimited number of design parameters and operating conditions at relatively small cost.

Battery manufacturing

Battery manufacturing is well-established today. This is particularly true of LIBs, seen as the reference technology at present and in the near future. Current LIB's cell manufacturing routes[7] can be divided into the categories: electrode production, cell assembly and cell finishing [169]. Electrodes are usually manufactured by 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.[231] Hereby coating and drying are the most cost-intensive processes.[232] Then, cell assembly is carried out, where steps such as stacking and electrolyte filling are becoming critical, because in general they are the most time and economy consuming. Finally, the cell finishing phase is developed, where formation and ageing of the cells are the most cost-intensive processes, regarding the challenges of processing time and yield rate.

In contrast, dry electrode manufacturing represents a more sustainable and energy saving manufacturing process, thereby eliminating solvents and the disadvantages that come with using them.[233] Until now, the representative methods of solvent-free concepts, such as pulsed laser[234], sputtering deposition[235,236] and extrusion suffer from several drawbacks, like very slow deposition rates and high processing temperatures, as well as limited ratio of binder to active powders, which significantly limits their scalability. To overcome these drawbacks, a new process called electrostatic spray deposition (EDS), also known as dry painting, has been proposed.[237] Dry painted electrodes exhibit good flexibility as well as the ability to coat very large areas.

Another manufacturing method has been proposed to realize 3D thick electrode tailored architectures to achieve more energy and power density, which integrates an extrusion-based additive manufacturing process for macro-control with the exposure to an electric field for micro-control. In this line, a new method of 3D printing battery electrodes that creates a micro lattice structure with controlled porosity has been recently developed and demonstrated that this approach vastly improves cell capacity and charge-discharge rates.[238] Nevertheless, more studies are needed before tackling scale-up and industrial application.

When it comes to the cell manufacturing models, there are relevant works in modelling each of the main steps of the LIBs manufacturing process.[239–246] As an example of electrode processing models, algebraic models for slurries have been proposed for estimating rheological properties.[247] Also, several works on Monte Carlo (MC) simulations[248–252] and Brownian Dynamics (BD)- based models[253–256] are already in place for studying spatial distribution of particles and particles suspensions for describing LIB electrode slurries. Computational Fluid Dynamics (CFD) models have been proposed to simulate the drying process of the electrode coatings to analyze potential binder migration upon solvent evaporation.[257–259] In the recent years, a number of Discrete Element Method (DEM) models have been proposed to simulate the calendering process of LIB electrodes.[260–263] In addition, the electrolyte filling step has been recently modeled using Lattice Boltzman Method (LBM) approach.[264–268]

In spite of this well-organised sequence of steps, current approaches to battery design and manufacturing should be overcome in order to:

- Accelerate new cell designs in terms of performance, efficiency, and sustainability. Couple multiphysics multiscale models with advanced optimisation algorithms in the AI framework as well as with inverse cell design. This would represent a crucial step towards autonomous battery design discovery and optimisation, as it connects the desired properties to specific cell configurations, electrode compositions, and material structures as targets to synthesise, characterise, and test (see Figure 20).

- Accelerate the optimisation of existing and future manufacturing processes in terms of cell chemistry, manufacturing costs, and sustainability/environmental impact by building a digital twin of the manufacturing process (see Figure 21).

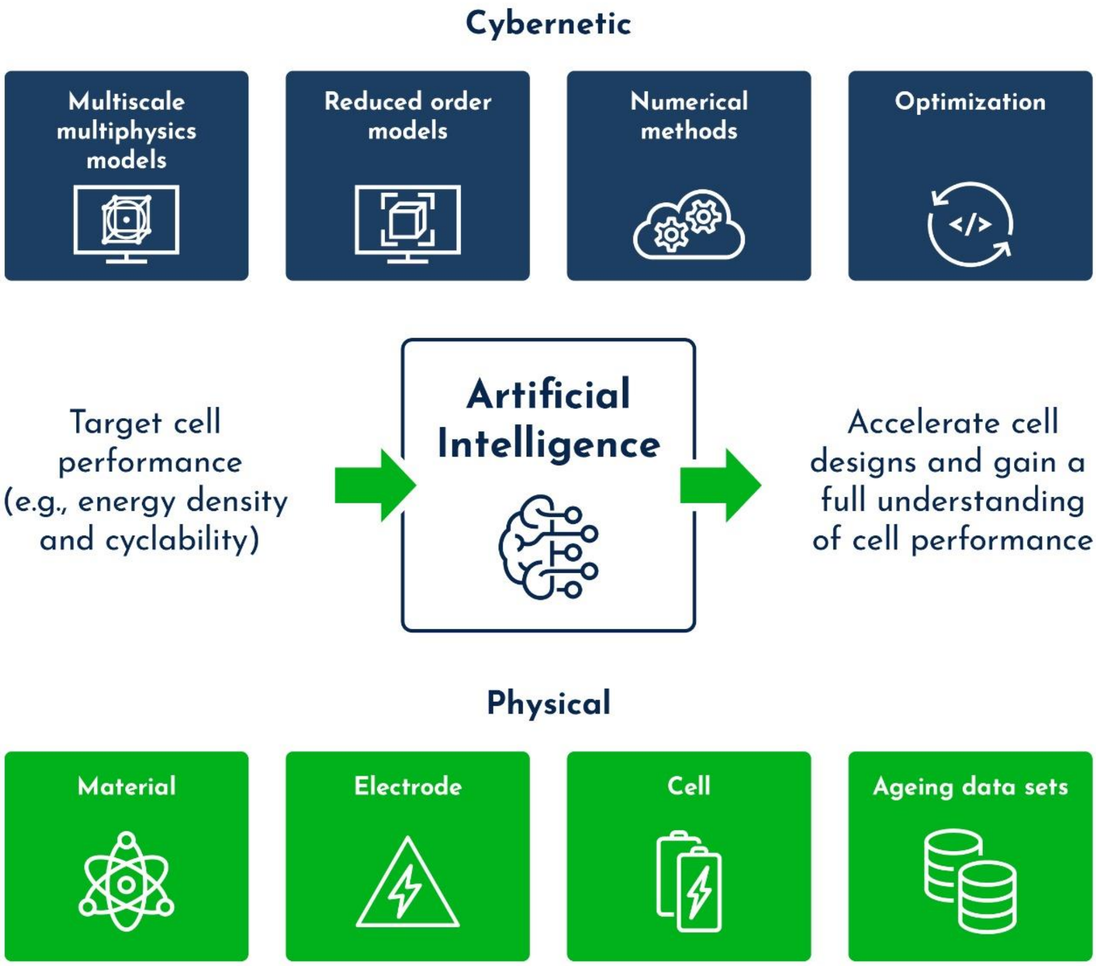


Figure 20. Inverse cell design based on digital twin of a cell.

New concepts will include radically new designs to minimise scrap and primary energy use and produce zero or nearly zero emissions. In this regard, current multiphysics modeling[80] can be of great importance in battery design and manufacturing. However, more effort is needed to develop a multi-scale physicochemical computational platform coupled to AI algorithms for the full manufacturing process chain of LIBs.

All these impressive efforts together with rapidly growing computational and algorithmic capabilities, particularly in the field of AI, call us to go even further. The computational simulation of cell design and manufacturing processes for new-generation batteries, for example, integrating interfaces discovered through the BIG–MAP concept and/or cells including sensing and self-healing functionalities, will certainly pose exciting new challenges for multi-scale computational science.

7.5.2 Challenges

Current LIB manufacturing processes face numerous challenges in order to meet highest standards on quality, low environmental impact, and economic competitiveness.

On the other hand, there is continuous evolution of the state of the art towards new technologies aiming for higher-energy-density, longer-lasting, and safer batteries. In some cases, the evolution may lead to a different paradigm for how batteries are designed and manufactured. To mention some examples, today's trends in lithium-based batteries on the lab and pilot scale involve the use of metallic lithium anodes, intercalated thin-layer electrodes, and solid electrolytes that are polymeric, inorganic, or hybrids combining both. Before market introduction, these and other approaches call for a substantial redesign of current manufacturing processes.

Given the disruptive nature of the concepts to be developed within the BATTERY 2030+ initiative, there is also the need to think outside the box in the cell design and manufacturing fields. It is not easy to anticipate what future battery technologies will be like, so no one can foresee exactly what manufacturing concepts will need to be put in place. Nevertheless, there are advanced tools at the technological forefront that will certainly play a central future role that may well be anticipated from today's perspective. The main focus of the manufacturability roadmap will therefore focus on **providing methodology to develop beyond-state-of-the-art processes in the future**.

In this sense, the challenges faced by the battery manufacturing industries can be divided into two levels. The **first level** of challenges is related to general methodologies for current battery production with a strong impact in the short term, but that will continue challenging the manufacturing of future battery technologies. These challenges are already being tackled today, but they will probably remain an open issue for some time, needing optimisation and adaptation to new materials and concepts. The **second level** involves advanced manufacturing concepts and approaches for future battery technologies that we can barely envision today. This is at the core of the scope of BATTERY 2030+ and is central to this roadmap.

According to these two levels, the following challenges may be outlined.

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.

Furthermore, there are difficulties adapting/modifying current manufacturing processes to accommodate next-generation batteries. Innovations such as using metal foil anodes (e.g., metallic lithium) and solid electrolytes (e.g., polymer, hybrid, or inorganic) are needed.

We need to overcome the paradigm of individual cells, involving excess packaging material, connections, and cabling, and move towards bipolar and other structures. This is a design issue with significant impact on manufacturing.

We should establish cell designs and manufacturing processes that allow for component-level recycling/reuse (i.e., electrode recovery and reuse from end-of-life well-performing cells).

We should develop tools to predict the impact of processing parameters on the characteristics and performance of the final product – or, otherwise, to predict the optimum processing parameters given the characteristics of starting materials – to leave behind trial and error, as stated in the state-of-the-art section.

We need to lower the general process cost, with less solvent and energy use, reduced scrapping, and faster manufacturing, especially during the formation step.

Standards and protocols

Finally, the development of standards and protocols for process development and monitoring needs 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. Since a deep understanding of individual process steps during manufacturing is fundamental to progress and innovation in the battery field, the development of standards can be expected to have a strong impact on battery manufacturing as it contributes to a more holistic understanding of the process chain.

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), that should rely on BattInfo as developed within BIG-MAP and extended to battery manufacturing. This data should include a complete battery history as introduced by the Battery Passport[269], 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. A summary of the goals in the short, and medium term to reach the long-term goals are given in Table 5.

Challenges related to future battery materials and 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 methods will be needed to implement the design rules from BIG–MAP.

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.

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

Tools to predict the impact of 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.

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).

7.5.3 Advances needed to meet the challenges

In a future scenario, current trial-and-error approaches should be avoided and cells and manufacturing processes need to be "smart", giving them a digital identity and creating a digital twin, i.e., a virtual counterpart to a physical object.

The advances needed **in the short term** in terms of battery cell design and manufacturing processes can be summarised as follows:

- Proof-of-concept (POC) of a digital twin of a cell design based on accurate multi-physics multi-scale models and AI data-driven models for LIBs. All in all, a digital twin of inverse cell design, providing capabilities for meeting battery cell performance targets (e.g., for energy, power, and cyclability) is envisioned to be developed.
- Improvements towards new greener and more sustainable manufacturing processes for LIBs manufacturing routes (3D printing, dry processing) are foreseen. New sustainable electrode and cell manufacturing techniques will be explored with the aim of reducing energy consumption, decreasing carbon footprint and Volatile Organic Compounds (VOCs) emissions.
- Coupling between process models and machine models along the LIB cell manufacturing for providing information on optimal manufacturing processes. Having validated multiphysics and multi-scale models coupled to AI algorithms of current LIB cell manufacturing processes capable of providing an accurate understanding of each step of the process will be a must.
- Further improvements of manufacturing plant technology with regards to flexibility. This includes the ability to process different materials, the ability to deal with quantity fluctuations as well as the ability to process varying cell formats.

- Development of standards and protocols for design and monitoring of manufacturing processes, prioritized by impact on process deployment and acceptance.

In general, the main benefits of having this approach are listed below:

- Provide new /disruptive cell designs for specific applications/cell chemistries
- Improve battery performance (e.g., power and energy density) through advanced design
- Increase sustainability in the battery manufacturing routes
- Increase battery manufacturing plant efficiency through the identification of the critical manufacturing parameters

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, analyzed, stored and used in predictive maintenance of the manufacturing plants.

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 labor, trial and error, and waste products. In addition, flexible and scalable manufacturing processes, as well as flexible, high-precision modelling tools for the optimisation of any technology regarding battery cell (i.e., SSB, SIBs) processing conditions and machine parameters is expected.

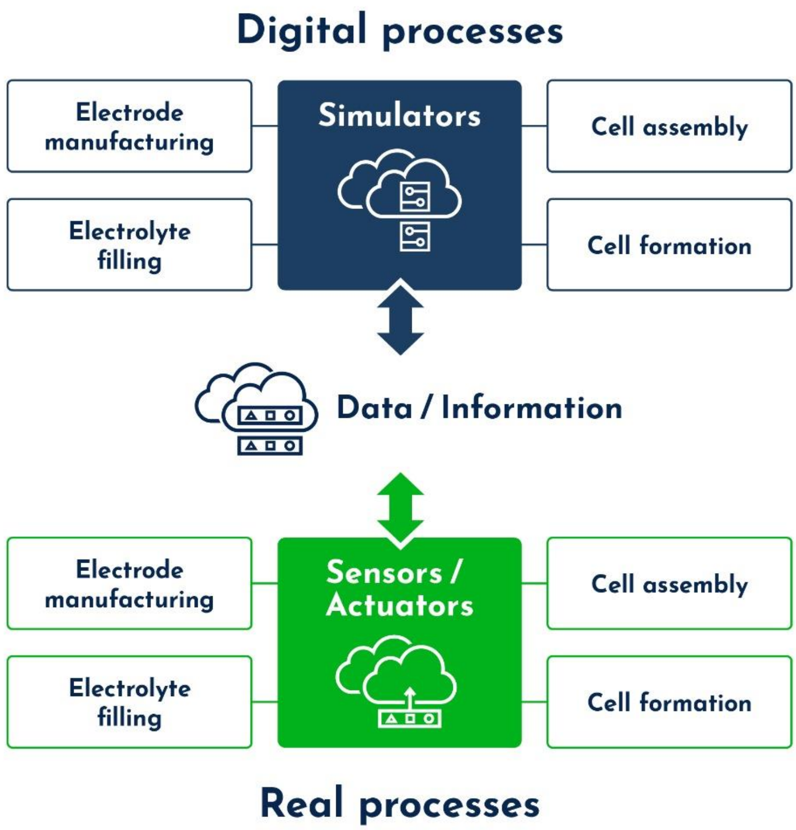


Figure 21. Digital twin of cell manufacturing processes.

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

The implementation of these techniques and methodologies calls for sequential step-by-step development in the short, medium, and long terms. Central to this process is the development of accurate and efficient modelling tools as a source of data feedstock for AI tools.

In the long term, i.e., ten or more years, full maturity of the methodology is expected, closing the loop by means of integrating the cell design and manufacturing design sub-loops (see Figure 22). In addition, POC of a digital twin of novel cell manufacturing routes with closed-loop recycling of optimized LIBs. Finally, some parts of this methodology can be progressively made available to industry, before the full package becomes available as a commodity in a new state of the art.

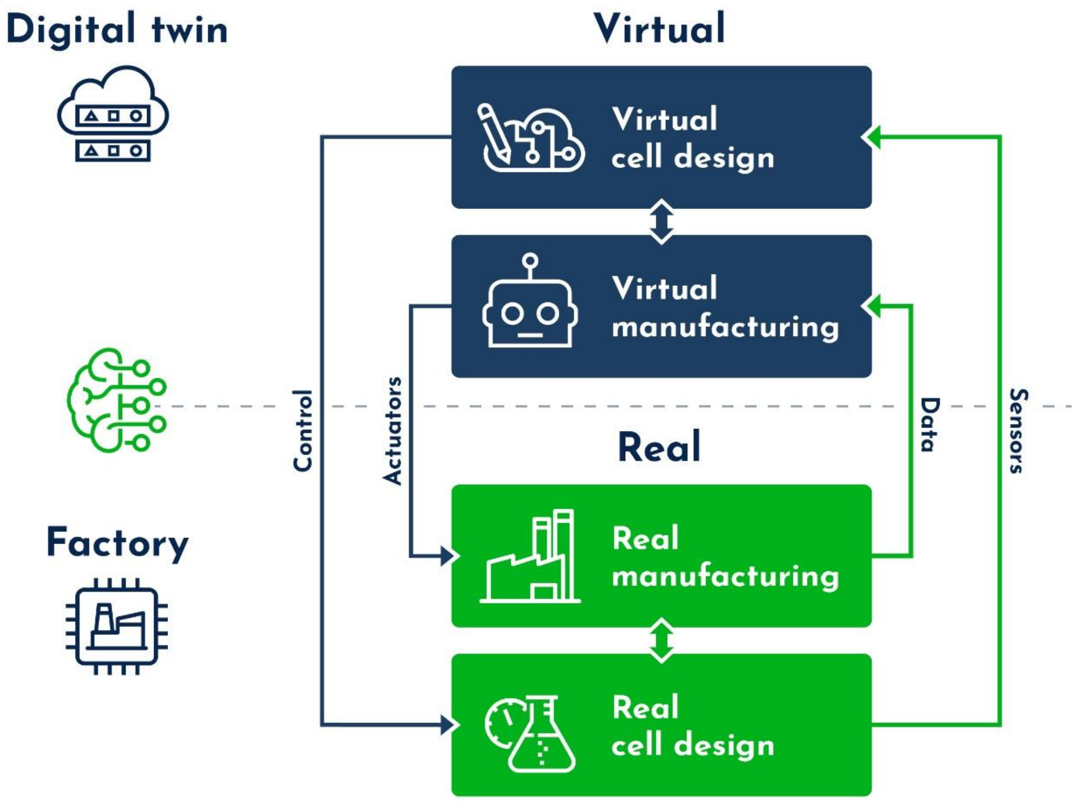


Figure 22. AI-driven design and manufacturing methodologies linked together as a whole.

Potential impacts of this approach:

The future battery manufacturing machines and tools, which will use sophisticated software and network sensors, will be used to plan, predict, adjust, and control business results, to lead to optimization of the whole value chain, where the following potential impacts are expected:

- Accelerate the discovery of new cell designs and manufacturing processes; reduce the development time and cost for new battery cells; reduce battery research and innovation (R&I) cost.
- Increase process speed, improve the efficiency of the battery cell production plants, and reduce the number of problems as well as the downtime, which ultimately leads to cost savings.
- The finished battery cells will have a higher level of quality and will be more comfortable and cheaper to use and maintain.

Potential challenges of this approach:

- Data management (usable, accessible, integrated, and curated);
- Data harmonisation: Data standards for the classification and unique description of battery cells and their manufacturing chain will be required;
- Standardisation and reference architecture: A reference architecture will be needed to provide a technical description of these standards and facilitate their implementation

- Intellectual property management (data ownership);
- The level of readiness of the Internet of Things (IoT) that connects machines and systems and allows seamless data transmission across all assets (virtual and real replica);
- Increase the adaptability of a battery manufacturing line to successfully produce new generation or novel technology battery cells.

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.[270]

- 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.[271]

- 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).

The development of battery dismantling and recycling technologies with high efficiencies going well beyond the EU Battery Directive 2006/66/EC target of 70% [269] 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 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.[272]

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 – now under revision [269]), 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. According to the current regulation, the recycled content should reach: **65% by weight for lead-acid batteries, 75% by weight for nickel cadmium batteries, and 50% by weight for all other batteries.**

The revision of the Battery Directive is expected to be published by 2023 with updated categories and recycling efficiencies (e.g 70 % by weight for Lithium- based batteries). 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 23. Currently, pyrometallurgy is the most applied method[273].

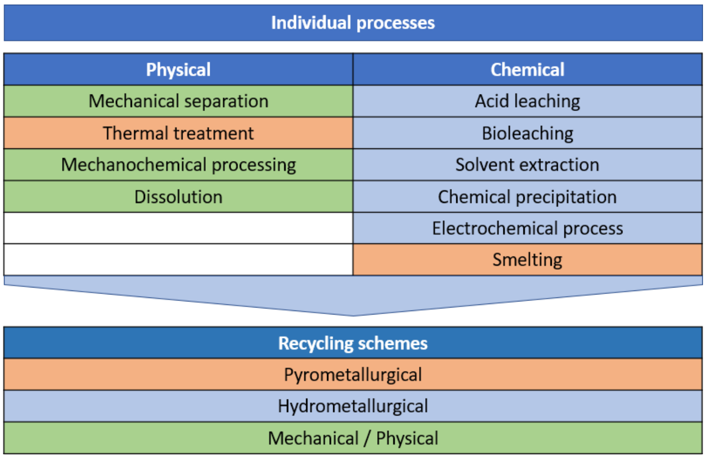


Figure 23. Recycling processes and schemes.

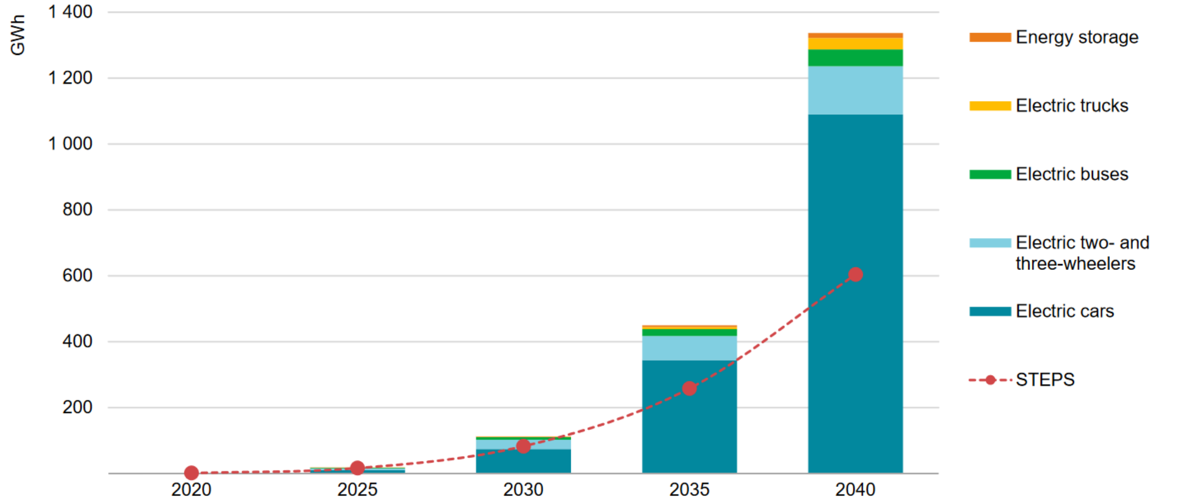
7.6.2 Challenges

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 24.[274]

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.[275]

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.



Note: GWh = gigawatt hour.

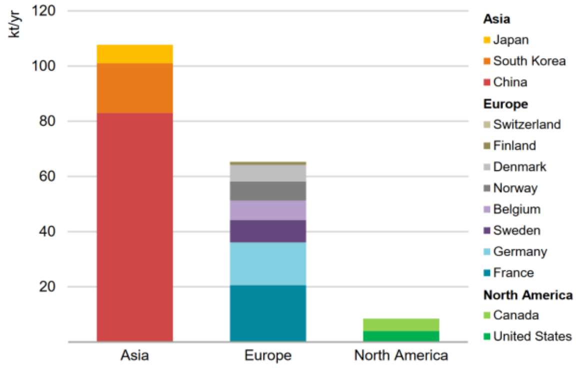


Figure 24. Amount of spent EV and storage batteries reaching the end of their first life by application until 2040 (top) and existing and announced lithium-ion battery recycling capacities to come online by 2021 by region (bottom).[274]

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.
- 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.
- Methodological challenges: the economic, ecological, and social impacts of emerging battery technologies must be analyzed 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.[276] 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, sodium-ion, 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.[277] 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.[278–280] Obsolete binders and additives will have to be removed in advance to further recovery steps of active materials.[281,282]
- Despite recent progress regarding direct recovery of electrode active materials,[283,284] 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 LCO to $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$,[285] 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.[286] Even though such impurities can be beneficial in some cases,[287] 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.[288] The required level of expertise can only be expected if advanced AI development, for example marker particles with magnetic codes,[289] 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"[290] 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)[271]. 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 25. 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.[291] 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.[292]

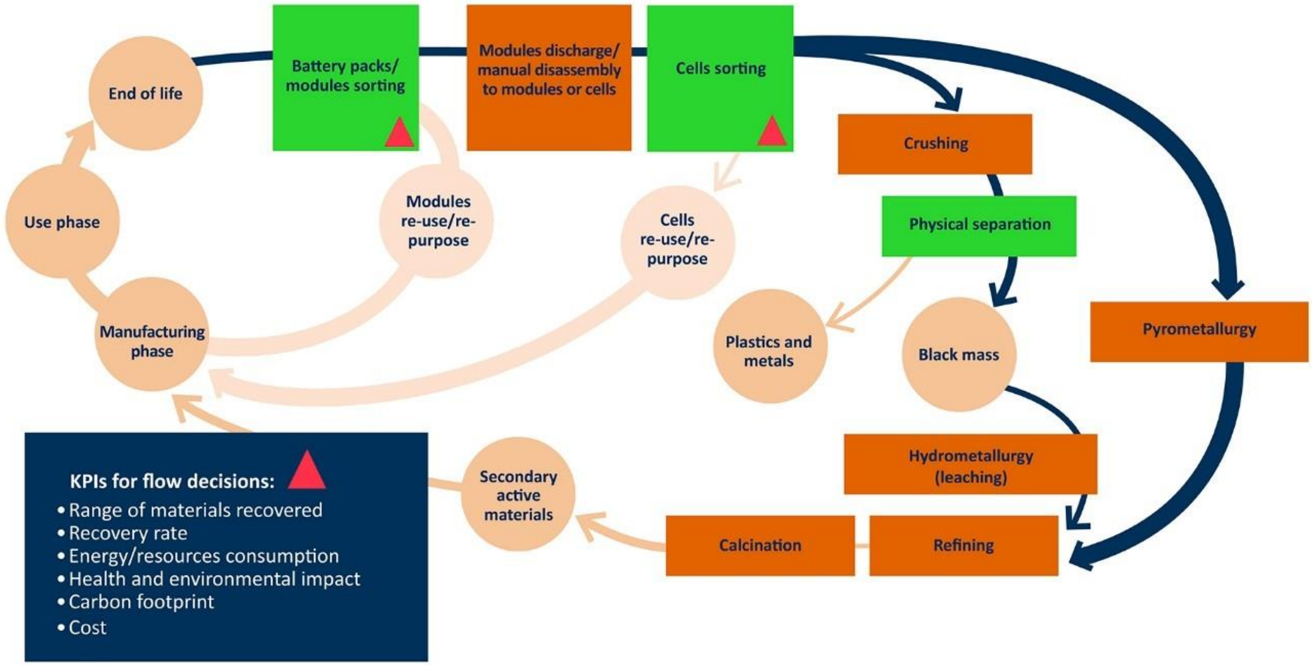


Figure 25. Present recycling process.

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 26) 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.[272]

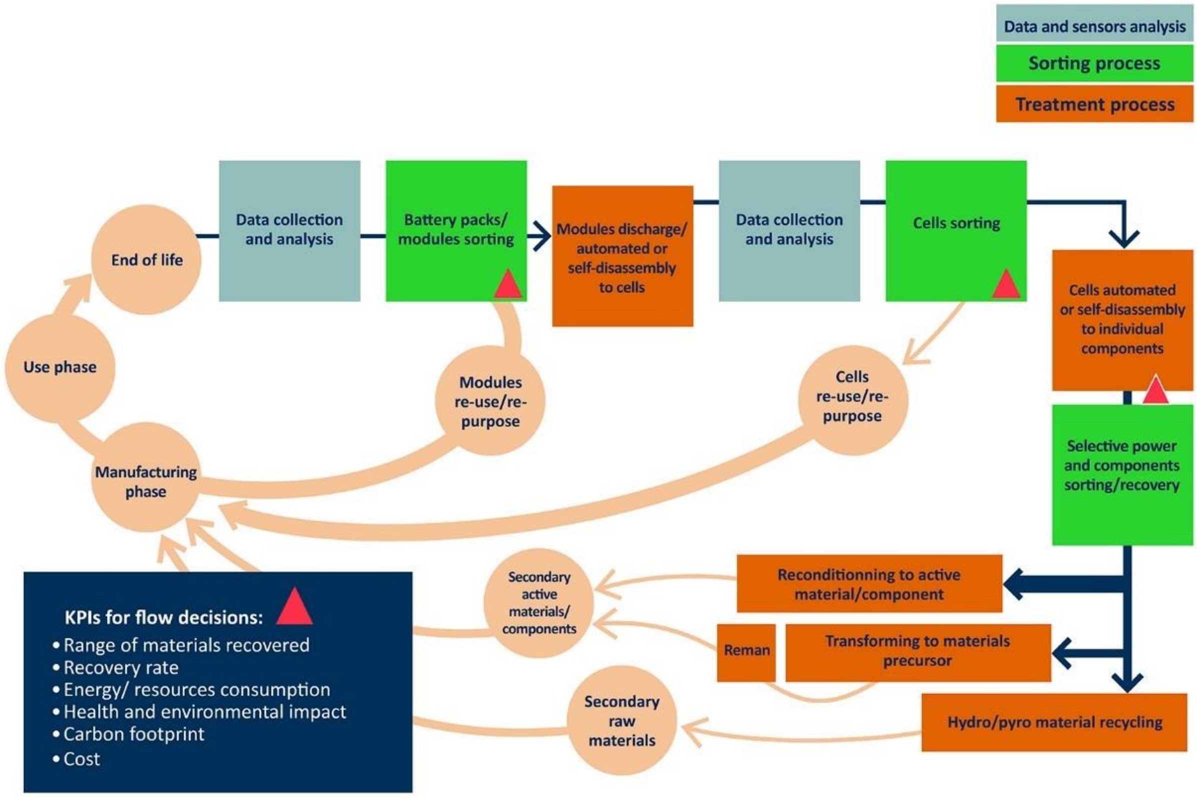


Figure 26. Future recycling process: direct recycling fully integrated with reuse.

While Figure 26 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.

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 27).

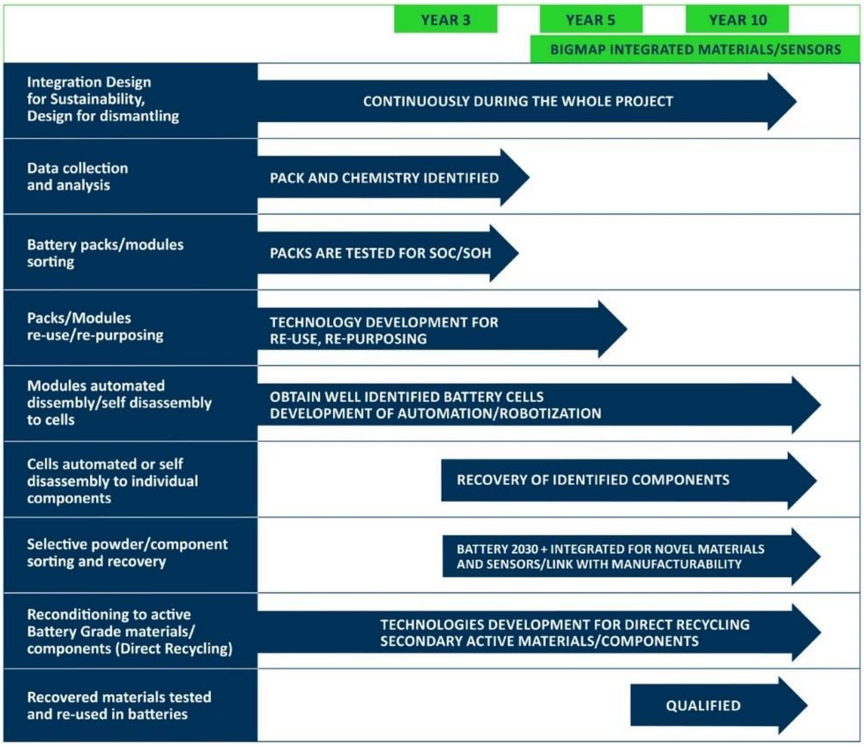


Figure 27. The ten-year roadmap for recyclability within BATTERY 2030+.

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”- draft under preparation.[273]

***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 as presented in the last section aim to be a governing factor in inventing the battery of the future. The progress in each respective research area will be essential for the newly developed battery technologies and for the vision of BATTERY 2030+ to reinvent the way to invent batteries. One crucial element of the success of each individual and of all research areas as a whole are not individual advances but the joint work for a common goal and the synergies in between the research areas. All of them are linked to one another and depend on in- and output being constantly transferred between the different areas, to create a closed loop between them in the end. The long-term vision is the accelerated and automatized research that is able to discover and invent new self-healing batteries which can be directly manufactured and recycled and which are safe, sustainable, low cost and have application-dependent tailored electrochemical properties. To effectively interlink the research areas early on, 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 constant and efficient feedback loop between sensor data, the BMS, and AI modules has to be set in place. External stimuli which are implanted in the cell will then be able to appropriately trigger the self-healing functions according to sensing information or based on predictive models in BIG-MAP. These self-healing properties should hereby not only include 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, cycle life and longevity as well as reliability and safety of future batteries will be highly improved. New materials and interfaces will be discovered at accelerated rates with a direct feedback into Sensing and Self-Healing. 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**. The closed loop is to enable a feedback loop to efficiently manufacture and recycle next-generation battery cells incorporating new materials, engineered interfaces, sensors, and self-healing functionalities in the long term. This will lead to new, environmentally friendly and cost-effective batteries.

This means, one aim is that after their first life, the sorted materials will be used for a second life and reintroduced at the beginning of the production chain, along with their full history and material information coming from sensor data. To capture all necessary cell data during their life, 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 new cell designs after design for recycling and a full proof of concept of a digital twin in manufacturing. A connection of sensors to an external connection point to constantly feed information into the BMS and transfer data to BIG-MAP is hereby necessary. The automation of the integration and connection of the internal wiring interfaces during the cell assembly as well as a possible transfer to the module level under 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. In the end a 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 closely related research areas, various short- and medium-term goals must be met along the way, each defined between two 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. Solutions have to be found to obtain data of different sensor types regarding the output format which have to be standardized and compatible. For comparability an ontologized data management will be put into place. Next steps will be the on-the-fly analysis of multimodal data from sensing on instrumentalized batteries. The multisensor input will be transferred to the BMS. With the input data from sensing feeding into BIG-MAP, material characterization and discovery by on-the-fly utilization of sensing data will be accelerated.

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. Between **BIG-MAP & Self-Healing** first steps are to initially detect whether self-healing works, accompanied by predictive models on of 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. With the predictive model that is to be established, failures in self-healing will be predicted and the end of self-healing properties to work can be estimated. In the medium term, the established predictive model to predict failures in self-healing will be set in place and preventive self-healing is triggered. Therefore, a close connection between Sensing, Self-Healing and BIG-MAP is needed early on to transfer self-healing data from the sensors to the BMS. For improved self-healing properties, new interfaces or materials (e.g. electrolytes) will be designed by BIG-MAP to facilitate self-healing.

The necessity of a close connection between the smart functionalities **Sensing & Self-Healing** is apparent. In the short term a constant feed of data from self-healing 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. For sensors to properly function and be able to measure self-healing properties, their amount and location has to be specified. In the medium term, the sensitivity and accuracy of sensors during long-term cycling have to be evaluated as well as effects of aging of sensors along with the sensor response to the cell being considered. The state of health and the self-healing functionalities will be constantly monitored with sensors to evaluate the long-term self-healing functionalities. 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 under consideration of 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.

For interlinking **Sensing & Cross-cutting areas** in the short term, procedures have to be developed to automatically insert the benchmark sensors inside the cells at pilot scale. In the beginning the cell chemistry aimed at will be LIB cells, while new chemistries will follow in the medium to long term. For manufacturing of the new batteries with smart functionalities additionally 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 under consideration of recyclability. For 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 sensors communication interfaces at cell level to the battery management system (BMS) will be implemented.

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 will enable manufacturing on the existing equipment. With the variety of self-healing materials at hand and new self-healing materials yet to be developed within BIG-MAP, the existing manufacturing routes will have to be reconsidered to 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 which have to be explored. At last, a procedure for the manufacturing of the self-healing components (i.e. self-healing electrodes) in LIB cells will have to developed. Following the short-term goals, in the medium term the integration between manufacturability & recyclability criteria and the development of new self-healing components is to be demonstrated. 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 a full system for direct recycling is to be developed and qualified, 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. Altogether, sustainability and recyclability concepts will be considered in the design of the cell from the beginning. In the medium term, an initial proof of concept will be performed, showing the integration of manufacturability criteria in the recyclability goals (easy to dismantle, sort and reuse).

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

To make this vision possible a final piece has to set in place. This piece is using the same 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.

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

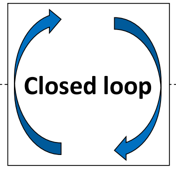
Cross-link	Short term (3 years)	Medium term (6 years)	Long term (10 years)
BIG-MAP & Sensing	Data from sensing and from operando *characterization* are correlated. Data of different sensor types regarding the output format is standardized and compatible. An ontologized data management is in place. Data is efficiently transferred from sensing to modelling, and from modelling to sensing.	On-the-fly analysis of multimodal data from sensing on instrumentalized batteries. Accelerate material characterization & discovery by on-the-fly utilization of sensing data in BIG-MAP. Multisensor input is transferred to the BMS. Preemptive & curative approaches are combined with an emphasiz on interfaces and forwarding ontologies.	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. 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.
BIG-MAP & Self-Healing	Monitoring and assessment of self-healing. A predictive model is established to predict failures in self-healing and estimate the end of self-healing properties to work. 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. A predictive model is established to predict failures in self-healing and preventive self-healing is triggered. Interfaces or materials (e.g. electrolytes) are designed to facilitate self-healing. Preemptive & curative approaches are combined.	
Sensing & Self-Healing	Frequency of sensing data from self-healing to interact with BMS is defined. Amount and location of the sensors regarding the self-healing functionalities is defined. Detection of whether self-healing is working.	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. Self-healing is triggered based on sensor data. Preemptive & curative approaches are combined.	
BIG-MAP & Cross-cutting areas	Exploration of new cell designs for the BIG-MAP disruptive materials, considering recyclability constraints. New manufacturing routes of the BIG-MAP components, based on the AI data-driven models.	A demonstration of the new and flexible manufacturing processes of the novel battery chemistries	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.
Sensing & Cross-cutting areas	A procedure for the automatic insertion at pilot scale of the benchmark sensors inside the LIB cells. Adaptation of internal interfaces and connections (communication pathways, electrical connections and power, etc.) to cell manufacturing tools and constraints under consideration of recyclability.	A demonstration of the integration between manufacturability & recyclability criteria and the development of new advanced sensors. 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. A procedure for adaptable manufacturing of the self-healing components (i.e., self-healing electrodes) in LIB cells. New cell design configurations including self-healing components to be explored. 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. Demonstration of spatial distribution of self-healing functionalities manufactured with roll to roll processes. Special cell design configurations to facilitate self-healing reactions New methodologies on multiscale modelling of manufacturing to be introduced and validated.	
Cross-cutting areas: Manufacturability & Recyclability	Integrated design for sustainability and recyclability concepts in the manufacturing routes. Implement design for sustainability and recyclability concepts in the AI data-driven models. Consider sustainability and recyclability concepts in the design of the cell.	An initial POC of the integration between manufacturability criteria and the recyclability goals (easy to dismantle, sort and reuse).	

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

Cross-link	Short term (3 years)	Medium term (6 years)	Long term (10 years)
BIG-MAP & Standards	Continue the development of the ontology eco system. Establish international collaborations. Realize a broad implementation of the **Battery 2030+ Electronic Lab Notebook** (ELN). Find attractive ways for researchers to use ontologies and standardization. Find ways to include new metadata and observations in otherwise standardized processes.	Go **from** the **Electronic Lab Notebook (ELN) to** the **Lab as a Service (LaaS)**. Utilize the ontologies and standards to make data fully FAIR. Have well-defined & standardized interfaces to enable reproducibility & interoperability.	Accelerate research by use of ontologies & standards.
Sensing & Standards	Ensure a transparent flow of information and enable the comparability of sensor results (sensor sensitivity and type, data postprocessing, environmental conditions etc.). Define how to determine data from measurements for each sensor type. Ensure the metrological traceability of sensors with regards to primary references in order to ensure comparable measurements and hence more meaningful experiments. 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).	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 connections.	Standardized communication with the BMS and generation of standardized sensor data for the **Battery 2030+ Electronic Lab Notebook (ELN)**. Automatized and standardized insertion of advanced sensors in the new generation cells.
Manufacturability, Recyclability & Standards	Standardization of metadata reports and data produced by digital tools (battery models, etc.) in use. Standardized protocols and reports in use. Inline quality control for common chemistries and processes in place. Find ways to handle sensitive data.	Process neutral and machine open standardization. Standardized & interoperable **Battery 2030+ Electronic Lab Notebook (ELN)** in use. Standardization in validation of digital tools.	Be able to hand over full battery history (battery passport: chemistry, manufacturing protocols, SoH,...) to recyclability. Standardized interoperable automated data acquisition for the **Battery 2030+ Electronic Lab Notebook (ELN)**.

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 or 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 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
BEV	Battery electric vehicle
BIG	Battery Interface Genome
BIG–MAP	Battery Interface Genome–Materials Acceleration Platform
BMS	Battery management system
BSH	Battery self-healing
CEI	Cathode–electrolyte interface
CFD	Computational Fluid Dynamics
CNT	Carbon nanotube
DEM	Discrete Element Method
DoE	Department of Energy, USA
EARPA	European Automotive Research Partners Association
EASE	European Association for Storage of Energy
EBA	European Battery Alliance
EDS	Electrostatic spray deposition
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
FBG	Fibre Bragg grating
FOEWS	Fiber optic evanescent wave spectroscopy
HPC	High-performance computing
HTS	High-throughput screening
JRC	Joint Research Centre, the European Commissions
KMC	Kinetic Monte Carlo
LBM	Lattice Boltzman Method
LCA	Life cycle assessment
LCOS	Levelized cost of storage
LEAPS	League of European Accelerator-based Photon Sources
LENS	League of Advanced Neutron Sources
LFP	Lithium iron phosphate (cathode material) – $LiFePO_4$
LIB	Lithium ion battery
Li-ion	Lithium ion battery
LM	Liquid metal
LMO	Lithium manganese oxide (cathode material) – $LiMn_2O_4$
MAP	Material Acceleration Platform
MC	Monte Carlo
ML	Machine learning

MOF	Microstructural optical fibers
NCA	Lithium nickel cobalt aluminium oxide (cathode material)
NMC	Lithium nickel manganese cobalt oxide – $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$
NMC 532	Lithium nickel manganese cobalt oxide – $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$
NMC 622	Lithium nickel manganese cobalt oxide – $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$
NMR	Nuclear magnetic resonance
NPS	Nano-plasmonic sensing
P2D	Pseudo two-dimensional
PCF	Photonic crystal fiber
POF	Proof of concept
QRL	Quality, reliability, and lifetime
RE	Reference electrode
RFB	Redox Flow Battery
ROM	Reduced order models
SEI	Solid electrolyte interphase
SET PLAN	Strategic Energy Technology Plan
Specific Energy	Energy stored gravimetrically, Wh kg^{-1}
SWCNT	Single-walled carbon nanotubes
SoC	State of charge
SoH	State of health
SP	Sensor plasmonics
Specific energy	Energy per unit mass (Wh kg^{-1})
TEM	Transmission electron microscopy
TRL	Technical readiness level
TBMS	Thermal battery management system
XAS	X-ray absorption spectroscopy
XRD	X-ray diffraction

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