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 is working on concrete actions to support the European Green Deal with a long-term vision of cutting-edge research reaching far beyond 2030.

A goal of BATTERY 2030+ is to develop a long-term roadmap for forward-looking battery research in Europe. This roadmap suggests research actions to radically transform the way we discover, develop, and design ultra-high-performance, durable, safe, sustainable, and affordable batteries for use in real applications. The purpose is to make a collective European research effort to support the urgent need to establish European battery cell manufacturing.

In the process of formulating this roadmap, stakeholders endorsing the BATTERY 2030+ initiative (at this stage more than 1300 individuals) were asked to provide written input to the first version of the roadmap published in July 2019. Representatives from the scientific community and battery industry have also been approached and asked to give their input. The Energy Materials Industrial Research Initiative (EMIRI) and European Automotive Research Partners Association (EARPA) organised a special workshop for their member organisations in September 2019. All the collected input was used to produce the second, more comprehensive draft published in November 2019 and discussed at the BATTERY 2030+ workshop on 20 November 2019 with more than 200 participants from research and industry.

We are very 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. The battery field is developing quickly and this roadmap is a living document that will be updated as the research needs change and the battery field progresses.

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1 Executive summary

Climate change is the biggest challenge facing the world today. Europe is committed to achieving a climate-neutral society by 2050, as stated in the European Green Deal.[1] 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.[2]

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.

BATTERY 2030+ will contribute to creating 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 roadmap, BATTERY 2030+ advocates research directions based on a chemistry-neutral approach that will allow Europe to reach or even surpass its ambitious battery performance targets set in the European Strategic Energy Technology Plan (SET Plan)[3] 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 and 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.[4] 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.

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 initative 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.

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

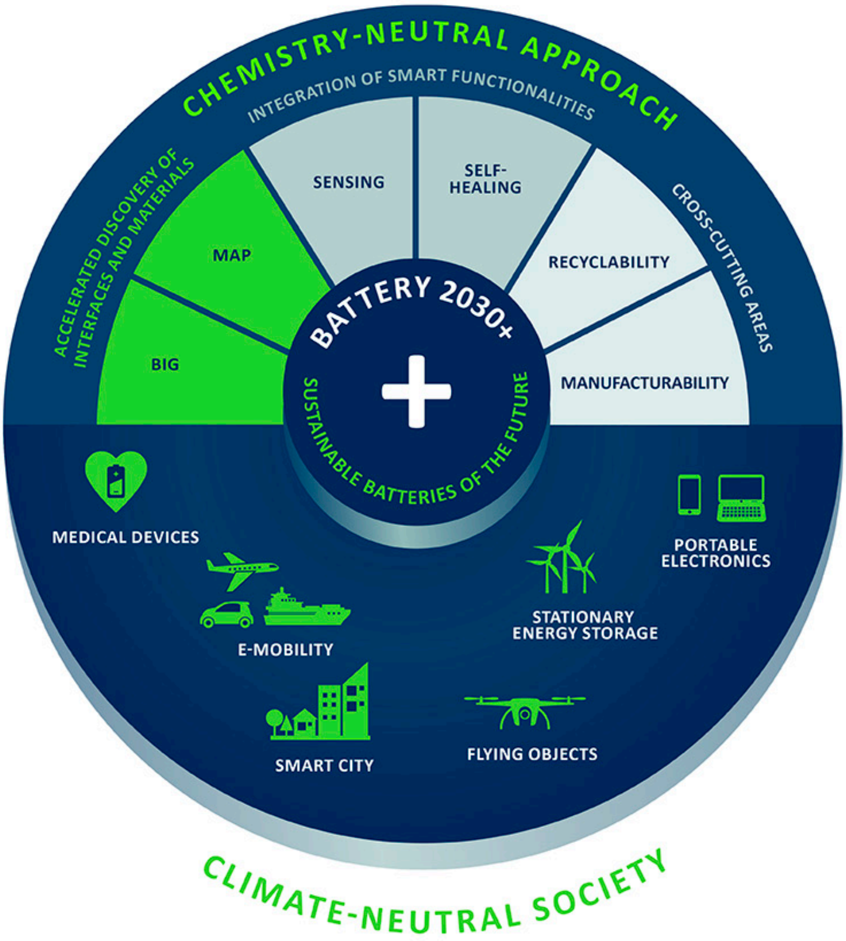


FIGURE 1. BATTERY 2030+: a holistic approach.

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’s total carbon footprint in 2017 was equal to 7.2 tons of CO_2 per person, according to Eurostat.[5] 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[1] 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.[2]

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.[6] 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).

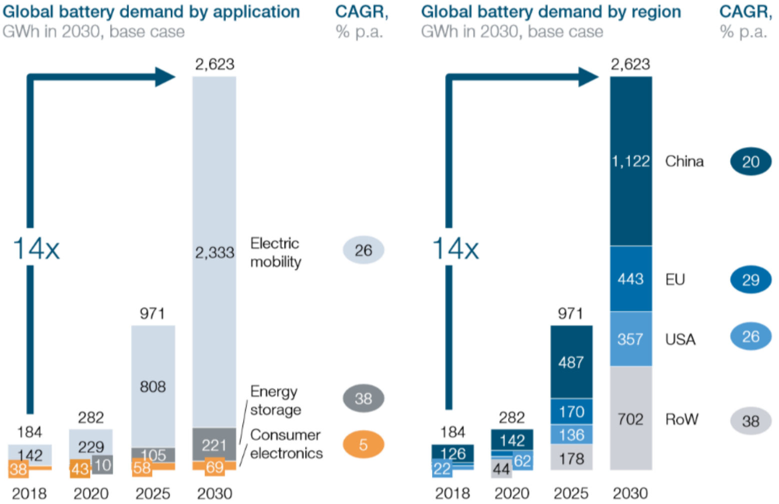


FIGURE 2. Expected growth in global battery demand by application (left) and region (right).[6]

The market for high-energy-density rechargeable batteries is currently dominated by the lithium-ion (Li-ion) chemistries, which performs well in most applications. However, current generation Li-ion batteries (LIBs) are approaching their performance limits. Without major breakthroughs, battery performance and production will not keep up with the developments necessary to build a 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 eco-system 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[7] 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.[8] 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.[9]

One action in the Strategic Action Plan[8] 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[9] and the SET Plan.[3]

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+ will follow a chemistry-neutral approach to facilitate the invention of the batteries of the future. BATTERY 2030+ is not about developing a specific battery chemistry, but about creating a **generic toolbox for transforming the way we develop and design batteries**. Thanks to this chemistry-neutral approach, BATTERY 2030+ will have an impact not only on current lithium-based battery chemistries, but also on post-lithium 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.

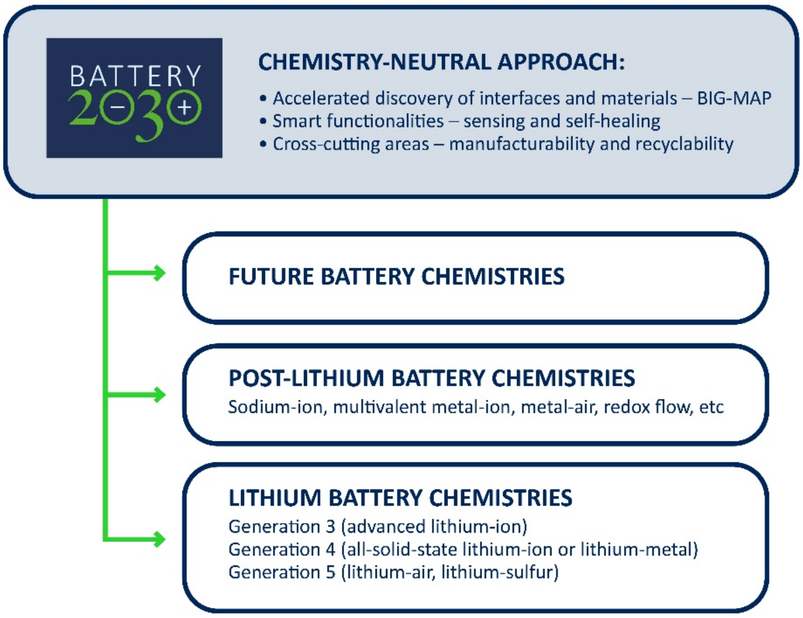


FIGURE 3. The BATTERY 2030+ chemistry-neutral approach will have an impact on both current state-of-the-art and future, as yet unknown battery technologies.

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:

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)**[10] to reinvent the way we perform battery materials research today. 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 AI and machine learning.

Interfaces in batteries are arguably the least understood aspect of the battery, even though most of the critical battery reactions occurs there, such as 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. 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 durability, enhancing lifetime, 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 batteries 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, state of safety, and usage history. Smart functionalities will enable better acceptance of used cells in primary and secondary applications.

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

4.3 Theme III: Cross-cutting areas

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

The battery of the future will be designed based on virtual representation taking into account sustainability and circular economy concepts including LCA.[11] 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[4] and digitalisation. The power of modelling and the use of AI should be exploited to deliver “digital twins”[12] 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

The six research areas that BATTERY 2030+ advocates as having major impacts on inventing the battery of the future are BIG, MAP, Sensing, Self-healing, Manufacturability, and Recylability. 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).

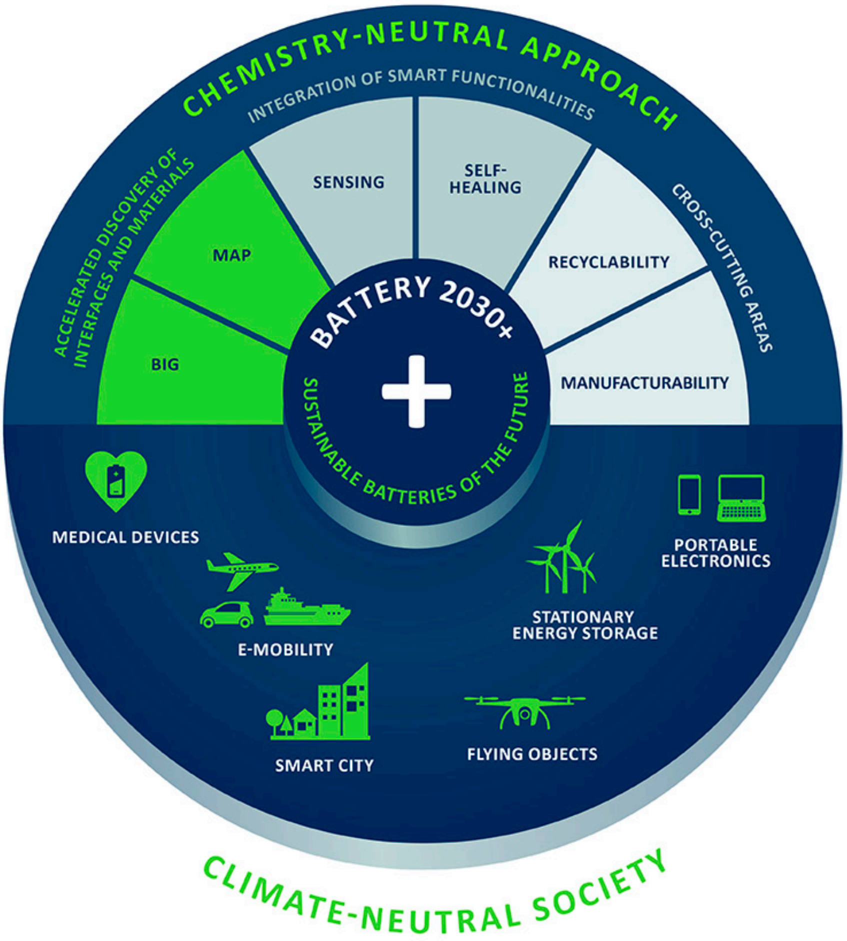


FIGURE 4. The BATTERY 2030+ vision is to invent the sustainable batteries of the future through a chemistry-neutral approach that will deliver ultra-high-performance batteries optimised for their intended applications, such as electro-mobility, stationary storage, medical devices, and robotics. BATTERY 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**.**

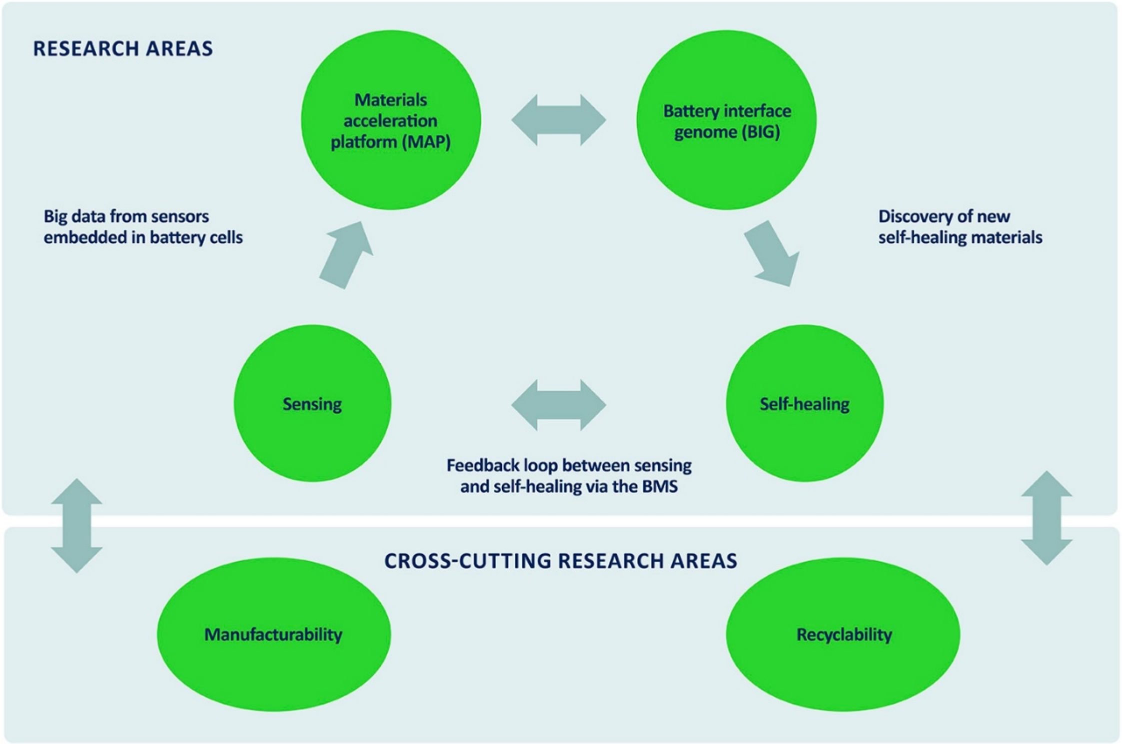


FIGURE 5. Interactions between the different BATTERY 2030+ research areas.

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 it will be possible to efficiently manufacture and recycle next-generation battery cells incorporating new materials, engineered interfaces, sensors, and self-healing functionalities.

For each research area, short-, medium-, and long-term goals have been identified and are here presented in Table 1.

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).		
	Deploy sensors capable of detecting various relevant phenomena (e.g., interface dynamics, electrolyte degradation, dendritic growth, metals dissolution, and materials structure change).	Deliver proof of concept of higher quality, reliability, and lifetime on the cell and module levels.	
Self-healing	Establising 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	Improving simulation tools, such as multiphysics models for reducing the computational burden of the manufacturing process.	Proof of concept of a digital-twin of a cell design (based on Li-ion chemistries).	An AI-driven methodology established for manufacturing, by integrating cell design sub loops that converge in a fully autonomous prototype system nourishing from BIG-MAP. The new concept is deployed to the industry and academia.
	Demonstrating the implementation of current AI technologies through deep learning and machine learning methods for cell design (for Li-ion chemistries).	Proof of concept of a digital twin of a cell manufacturing process (based on Li-ion chemistries).	
	Implementation of the AI-driven methodology for manufacturing (Li-ion chemistries) – including digitalisation.	Input from BIG, MAP, sensing, self-healing, recycling and other innovation areas integrated into the design and manufacturing process.	This methodology, which will help found a new commoditised state of the art, will be progressively deployed in industry and academia.
	Improving and scaling-up of new manufacturing processes (3D printing, dry processing).	Digital twin methodology adapted to the manufacturability of new battery technologies and innovative new manufacturing processes.	
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.	
	Establising a European system for data collection and analysis.	Significantly improve, relative to current processes, the recovery rate of critical raw materials	
	Demonstration of new technologies for battery packs/modules sorting and re-use/re-purposing.	Testing of recovered materials in battery applications.	
	Developing automated disassembly of battery cells.	Develop prediction and modelling tools for the reuse of materials in secondary applications	

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 at 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 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.

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 with close interactions with and support from other European initiatives, industry stakeholders, and networks that either are part of BATTERY 2030+, or who will be engaged early on.

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 over time.

Figure 6 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 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) engineering concepts have not been developed and tailored for efficient cell production; and (c) performance degradation remains an issue. The themes and research areas of BATTERY 2030+ will address these issues as shown in Figure 6.

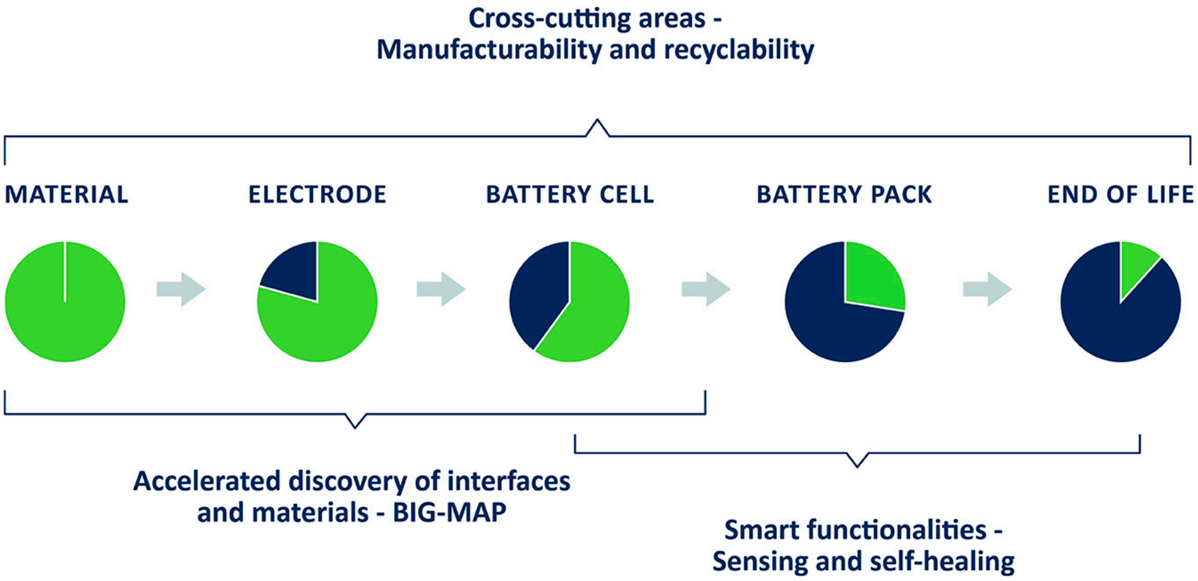


FIGURE 6. The decrease in total capacity as more inactive material is added when going from the material to the complete battery pack. The identified research areas will address these losses throughout the battery value chain. End of life represents the additional capacity loss due to degradation.

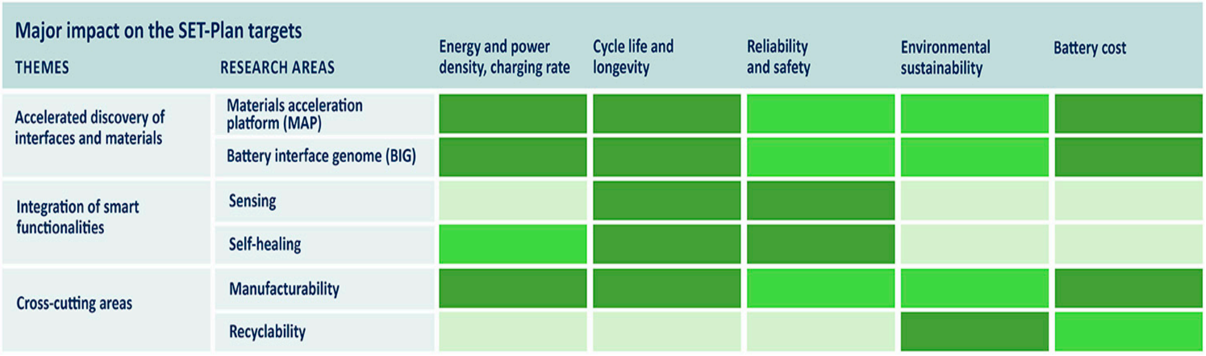
5.3 Impact on the European SET Plan targets for batteries

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[3] 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).



Major impact on the SET-Plan targets

THEMES	RESEARCH AREAS	Energy and power density, charging rate	Cycle life and longevity	Reliability and safety	Environmental sustainability	Battery cost
Accelerated discovery of interfaces and materials	Materials acceleration platform (MAP)	■	■	▨	▨	■
	Battery interface genome (BIG)	■	■	▨	▨	■
Integration of smart functionalities	Sensing	□	■	■	□	□
	Self-healing	▨	■	■	□	□
Cross-cutting areas	Manufacturability	■	■	▨	▨	■
	Recyclability	□	□	□	■	▨

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, but nickel-cadmium and nickel-metal hydride batteries as well as some non-rechargeable chemistries are also produced in Europe. There are also strong efforts to develop vanadium redox flow batteries, mainly for stationary energy storage solutions.

The first commercial LIBs came on the market in the 1990s. Since then, the energy density of LIBs has more than doubled while the cost has dropped by a factor of 15. 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.

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. Lead-acid batteries are still being developed for several of these markets due to their robust performance over a wide temperature range, high recycling percentage, and low cost. Advanced lead-acid batteries are expected to gain an increased market share over the next ten years. They cannot compete, however, for use in electric vehicles due to their considerably lower energy density. The development of redox flow batteries is mainly targeting large-scale energy storage applications, for which they have technical advantages such as scalability and nearly unlimited life. However, redox flow batteries have a very large environmental footprint and depend on the flow of large quantities of corrosive materials; as such, they are most suitable for stationary industrial applications.

The status of current commercial batteries and possible future chemistries is summarised in Figure 7, 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.[13–16]

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 hasards are regulated in the Battery Directive and in the upcoming Ecodesign Directive for Batteries. In its roadmap, the European Council for Automotive R&D EUCAR[17] 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.[18] The expected cost will decline to well below USD 100/kWh by 2024,[18] 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 increase in energy performances is levelling off for LIBs, so new solutions and ideas are sorely needed. It will be difficult or even impossible to satisfy future requirements for electrochemical energy storage using solutions based on current technologies.

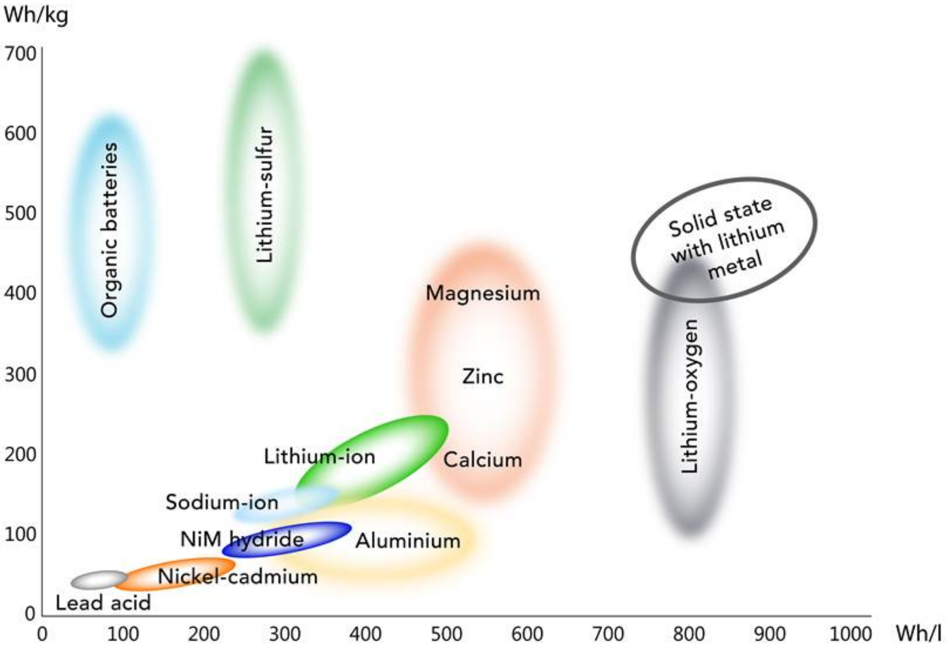


FIGURE 7. Current commercial batteries and targeted performance of future possible chemistries. The post lithium batteries chemistries are given as names indicating all kinds of metal-type batteries in respective category. There is a large uncertainty of their respective position in the graph. NiM hydride refers to nickel metal hydride.

BATTERY 2030+ is intended to move from the current state of the art for energy content to embrace the multiple possible future battery chemistries shown in Figure 6. 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 8 compares the European goals (shown in green), based on the development of different generations of batteries, with those of China, Japan, and the USA.

Several associations and countries have published roadmaps for batteries or for energy storage including batteries. Some recent roadmaps are from: EASE[19], EMIRI[20], EUCAR[17], implementation of the SET Plan Action 7[9], JRC [21–24], China[25], Finland[26], India[27,28], Japan[29,30], and the USA.[31]

Some international targets for automotive batteries expected up to 2035 are shown in Figure 8.[32] The green line represents the different generations of LIBs and when they are expected on the market, according to the SET Plan. The most ambitious target is that of USA Battery 500, which foresees that solid-state batteries will be available as early as 2022–2023. 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.

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.

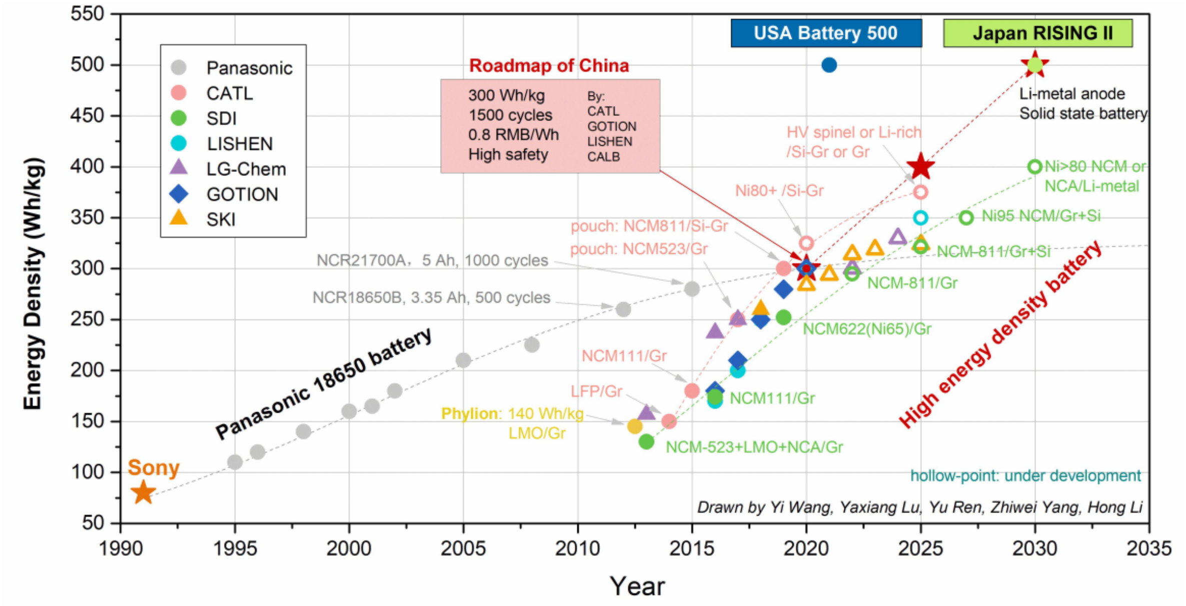
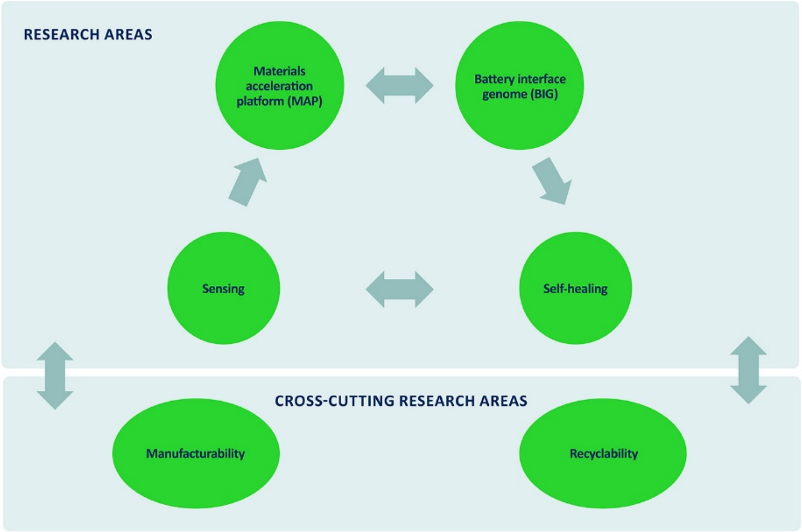


FIGURE 8. Comparison of the gravimetric performance of different batteries for automotive applications. The targets from the SET Plan coincide with the green line (different NCM-based generations of lithium-ion batteries). Japanese Rising II follows targets similar to those of the SET Plan, while China's 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.

7 Research areas



Battery research occurs throughout the value chain of battery development. Battery research can be oriented towards battery cells, based on competences in chemistry, physics, materials science, modelling, characterisation, etc. It can also be oriented towards systems where the battery cells are integrated into packs, to be used in different applications. Here, the field relies on knowledge of electronics, electrical engineering, systems control, modelling at the system level, AI, and machine learning – to mention but a few. Also, research in recycling has become more important and again relies on chemistry, metallurgy, physics, and materials science linked to the use of new efficient characterisation tools.

The European research infrastructure 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 LEAPS and LENS, which are resources with potential to enable the BIG–MAP initiative.

The areas of research advocated by BATTERY 2030+ rely on these cross- and multidisciplinary approaches with a strong wish also to integrate other areas of research to enable cross-fertilisation. In this section, detailed descriptions of the research areas proposed in this roadmap are given. Each section describes the current status in the field, the challenges and expected progress in realising the vision, and the overall objectives of BATTERY 2030+.

7.1 Materials Acceleration Platform (MAP)

Materials discovery and development crosscuts the entire clean energy technology portfolio, ranging from energy generation 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), which will be integrated with the **Battery Interface Genome** (BIG) described below. The proposed BIG–MAP infrastructure is modular and highly versatile, in order to accommodate all emerging battery chemistries, material compositions, structures, and interfaces. Following the format of Mission Innovation: Clean Energy Materials (Innovation Challenge 6) MAP Roadmap,[10] MAP utilises AI to integrate and orchestrate data acquisition and utilisation from a number of complementary approaches and technologies, which are discussed in the sections below.

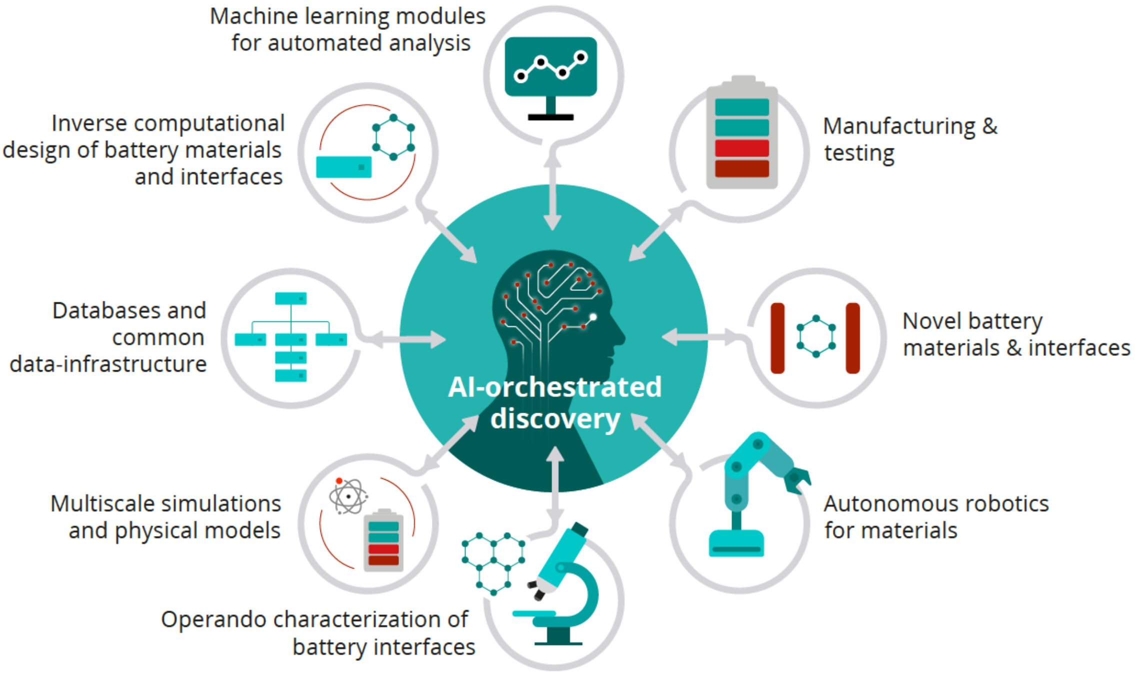


FIGURE 9. Key components of establishing a battery MAP.

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.

Successful integration of computational materials design, AI, modular and autonomous synthesis, robotics, and advanced characterisation will lay the foundation for dramatically accelerating the traditional materials discovery process. The creation of autonomous, "self-driving" laboratories 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 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).

In recent years, several examples have emerged in which the close integration of virtual (typically atomic-scale) computational material design and in operando characterisation[33] techniques in a circular design loop can accelerate the discovery cycle of next-generation battery technologies, such as high-capacity Li-ion cathodes[34] and materials for secondary metal–air batteries,[35] 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.

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 data from multiple sources, 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[36,37] for experimental data. Additionally, computational infrastructures such as PRACE and EuroHPC, and platforms such as UNICORE,[38,39] SimStack,[40] AiiDA,[41] and Materials Cloud[42] facilitate efficient and reliable high-throughput calculations. To fully exploit these data, extensive efforts, for example, by the European Materials Modelling Council (EMMC),[43] 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 ontology 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.

Multiscale modelling: Battery performance and lifetime are determined by many processes that occur on vastly different time and length scales.[44] 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. To address this challenge, single-scale models must be combined to form multi-scale workflows, for example, through deep learning models. Multi-scale modelling techniques are currently being developed, for example, to optimise real and virtual electrode microstructures[45] and to study the effects of the fabrication process on cell performance[46] and electrode surface film growth.[47]

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. 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. 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, 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,[48,49] and examples are also emerging in the development of solids and thin-film materials.[50,51] 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.[48]

Experimental and computational high-throughput screening of large compound libraries for activity in the accelerated formulation of relevant battery materials via the use of automation, miniaturised assays, and large-scale data analysis can accelerate materials discovery by up to one order of magnitude.[52,53] Several examples of fully automated high-throughput screening (HTS) systems for electrolyte formulation, cell assembly, and selected relevant electrochemical measurements are now available,[54] for example, at the MEET Battery Research Center in Germany.

AI in materials discovery offers great prospects,[55] 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. 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[56] and Phoenics[57] 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 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[58] of battery materials and interfaces requires the integration of high-fidelity data covering complementary aspects of the material and device characteristics. Currently, such datasets are sparse and cover only a fraction of the required data space.

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.[59]

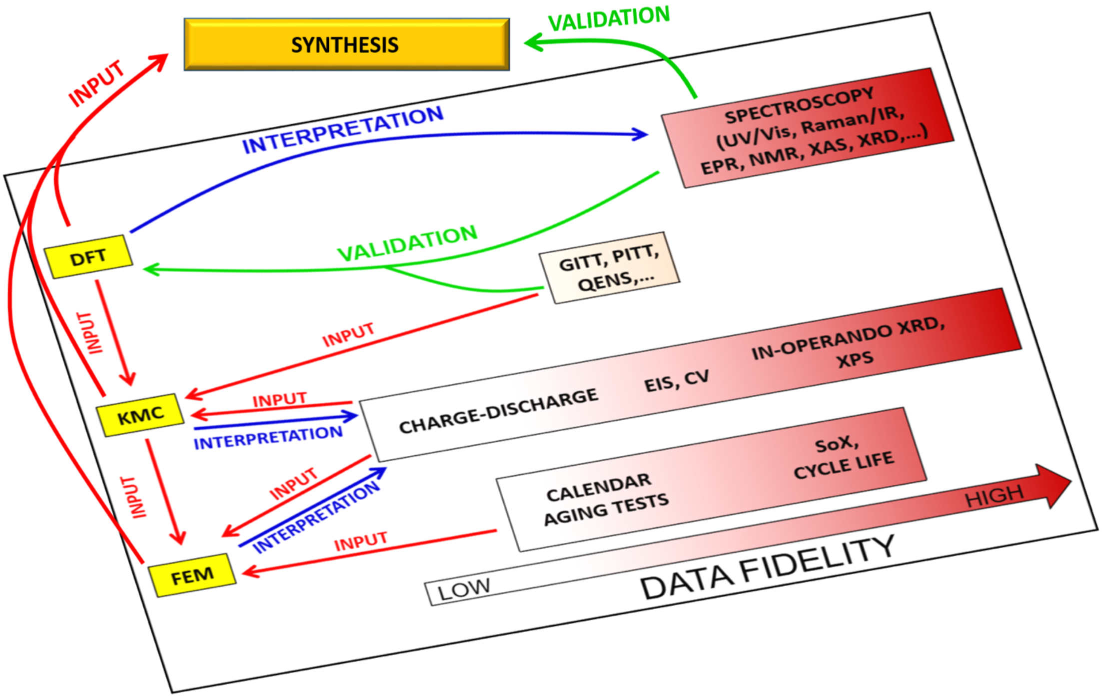


FIGURE 10. Illustration of the data flow between representative experimental and theoretical methods for studying battery interfaces. The fidelity of each method is generally proportional to its cost, but the fidelity–cost relationship can be optimised by acquiring data only when the given method/data is most valuable (adapted from[59]).

While machine learning could potentially massively accelerate the screening and identification of, for example, the structure–property relationships of inorganic energy materials,[60] a key challenge in the discovery of battery materials and interfaces is the development of autonomous workflows for extracting fundamental relations and knowledge from sparse datasets[61] 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[51] 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.[62,63] 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 with automated synthesis and characterisation technologies will enable the envisioned breakthroughs required for the implementation of fully autonomous materials discovery.[59]

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 consortium 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. 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,[64–66] many electrochemical tests do not work on short time scales; in particular, cycling experiments can take days to months or even years.[67] 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. 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.[49,68] 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. 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: The root of the multi-scale challenge is that it is not known how best to couple models at different scales in a efficient and robust way. The large gain in time with and accessible size of larger-scale models generally entail the sacrifice of detail and resolution. Releasing the full potential of inverse multi-scale modelling to support new materials and device design requires radically new approaches to link scales beyond the state of the art that can be achieved by isolated research communities in individual countries.[69] Machine learning techniques and other physics-guided, data-driven models can be used to identify the most important parameters, features, and fingerprints.[70] MAP will exploit European computational infrastructures, such as those offered by PRACE and EuroHPC, 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.

AI exploitation: AI-based generative models,[71] 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,[72] and in operando characterisation. These data must span all aspects of battery materials from synthesis to cell-level testing.[73]

Unification of protocols: MAP will offer a unique opportunity to leverage the size of this effort in the interest of standardising data 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.[74] 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. Interface-specific performance metrics at different time and length scales can be achieved, while retaining 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 data from all domains of the battery discovery and development cycle. Use automated 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.

In the medium term: Implement BIG in the MAP platform (BIG–MAP), capable of integrating computational modelling, autonomous synthesis robotics, and materials characterisation. Succesfully demonstrate the inverse design of battery materials. Directly integrate data from embedded sensors in the discovery and prediction process, for example, to orchestrate proactive self-healing. Demonstrate transferability of the BIG–MAP approach to novel battery chemistries and interfaces.

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. Implement and validate a digital twin of ultra-high-throughput testing on the cell level.

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. 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. 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 integrating dynamic events at multiple scales, for example, from the atomic to the micron scales. Therefore, BIG aims at establishing the fundamental "genomic" knowledge of battery interfaces and interphases through time, space, and chemistries.

The Battery Interface Genome – BIG – can be related to the concept of descriptors in catalyst design,[75] 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.

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.

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 provide an efficient SEI. Accelerated development of such an SEI would greatly benefit from high-throughput techniques and the AI-assisted rationalisation outlined here.

To derive valuable insights into the spatio–temporal evolution of interfaces and interphases, interoperability and scale coupling are necessary. The complexity of electrochemical systems usually forces the simplification of simulations such that they only qualitatively mimic the real situation in the battery. Therefore, even if the proper theory for performing the necessary statistical averages is derived, the obtained parameters/descriptors may deviate considerably from the parameters of the materials in their more complicated electrochemical environment. A coupling with physics-aware data-driven methods would strongly enhance the quality of the determination of interface descriptors, features, and parameters by enriching the physical simulation with validated correlations between idealised physics/chemistry-based simulations and data on real materials.

A complete and closed mathematical description of the whole reaction mechanism is enormously challenging, since coupled ionic and electronic transfer reactions in an electrochemically relevant environment include usually coupled multistep reactions.[76,77] These multistep reactions are often either tremendously simplified or the reaction steps are modeled in ideal environments.[78] In specific cases, it is possible to combine DFT methods with classical approaches to improve the description of surface reactions,[79] but generic approaches remain limited and an efficient and systematic coupling is still lacking.

7.2.2 Challenges

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 have been made in recent years to uncover the complexity of the interface dynamics and to control their reactivity and functionality, generating an enormous dataset whose depth remains largely under exploited. Hence, a complete paradigm shift is needed in order to address this fundamental challenge. For that, data must be collected, handled, and analysed in a more systematic and automated/autonomous manner, for example, 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, the simulations and models should become more realistic and closer to experimental conditions. Similarly, the experimental conditions should be made as ideal as possible to decouple the different effects and reactions, especially for the initial training of the hybrid physics-aware models discussed previously.

In this regard, key challenges include the development of new multi-scale modelling concepts (including physics-aware data-driven hybrid models to identify interphase descriptors), the development of new characterisation techniques, and the standardisation of experimental data and observables as inputs to physical models to make the link between observables and descriptors.

Building a fundamental understanding is the first step in controlling the complex and dynamic processes at the interfaces in emerging battery technologies, and thus holds the key to developing ultra-high-performance, sustainable, and smart batteries, fully exploiting the potential of the electrode and electrolyte materials. 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. 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.[80] It can be envisioned that mature battery interface/interphase characterisation techniques could provide high-throughput experimental input about battery interfaces during operation. Today, the analysis of experimental results is often too time consuming. One of the key challenges in establishing BIG is to automate the acquisition, curation, and analysis of the enormous datasets that will be generated. These data will feed the physics-aware data-driven hybrid models that will help us better understand and predict interface and interphase properties.

This will only be possible if datasets are created 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 battery interface genom[81,82] for interface/interphase development and dynamics. This has the potential to lay the foundation for the inverse design of battery interfaces/interphases[73], for example, using region-based active learning algorithms.[83]

Understanding and tracking different types of uncertainties in the experimental and simulation methods, as well as in the machine learning framework of, for example, generative deep learning models,[84] 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,[85] 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

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. 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, 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. 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.

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.[86,87] 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. At the high-throughput level, characterisation techniques should be organised to allow investigation of a large number of samples by providing the necessary meta-data. This requires workflows that can generate and analyse large amounts of data in an automated/autonomous manner, representing a major advance toward defining a new methodology for acquiring data about interfaces.

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, a checklist of good practices should be defined, becoming the project’s characterisation quality label. BIG thus represents a unique opportunity to design a common European strategy in which cycling 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.

Rather than a single physical property, a multi-scale/multi-feature approach combining different computational tools will certainly be necessary to grasp the dynamics of the interface at different scales.[44] 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 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. Merging advanced multi-scale modelling and data analytics will master the complex coupling of relevant length and time scales, which are so relevant to batteries. 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/interphases, it is necessary to incorporate the relevant physical understanding, and the model should be 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 11).

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.

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. 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).

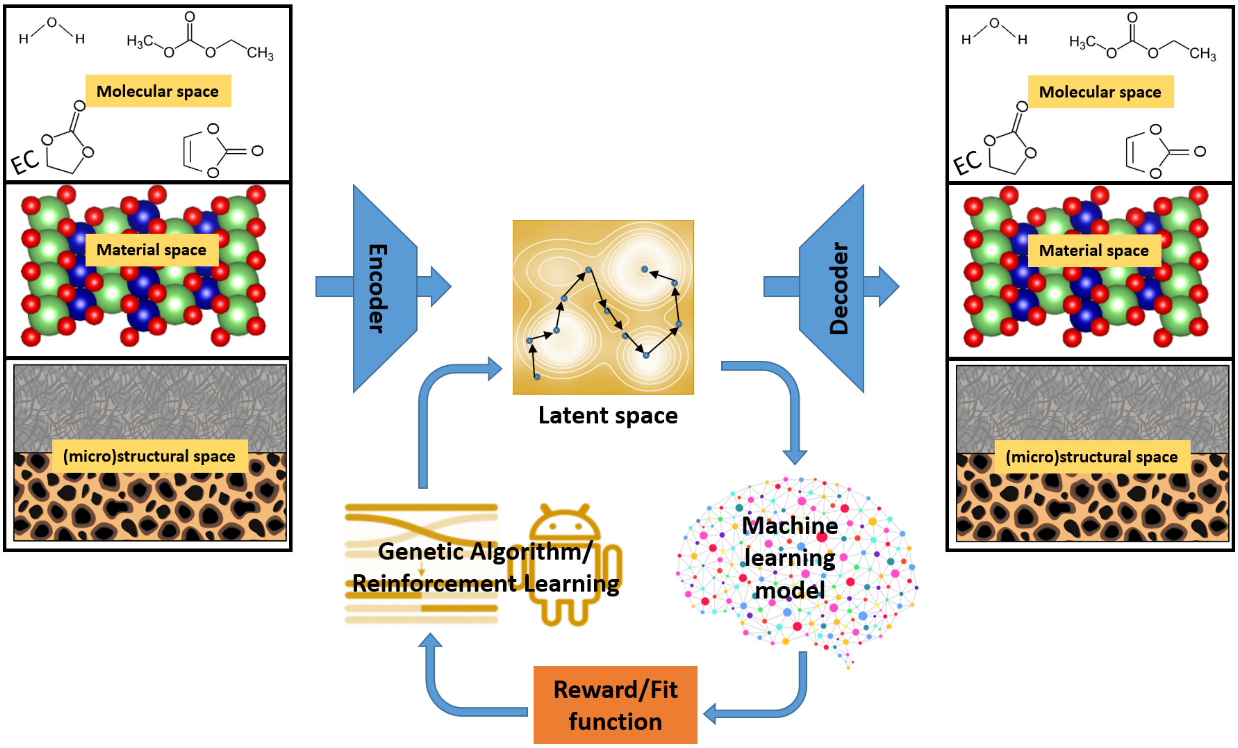


FIGURE 11. Generative model of interphase design. Variational auto encoder (VAE)-based encoding and decoding of chemical and structural information on a battery interphase into latent space, to enable generative battery interphase design through the use of, e.g., genetic algorithms or reinforcement-learning–based exploration [73].

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 characterisation and testing data using AI and simulations. Develop interoperable high-throughput and high-fidelity interface characterisation 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).[88] 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 electric vehicle (EV) applications. 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 (Figure 12).

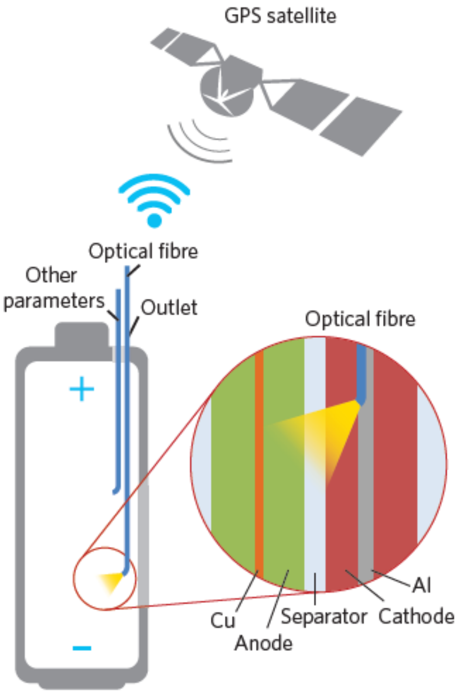


FIGURE 12. A future battery with an output analyser connected to sensor (optical fibres, wires, etc.) in addition to the classical positive and negative electrodes.

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, with wireless 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.[89] 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.[90] 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 13). 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).[91] 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.

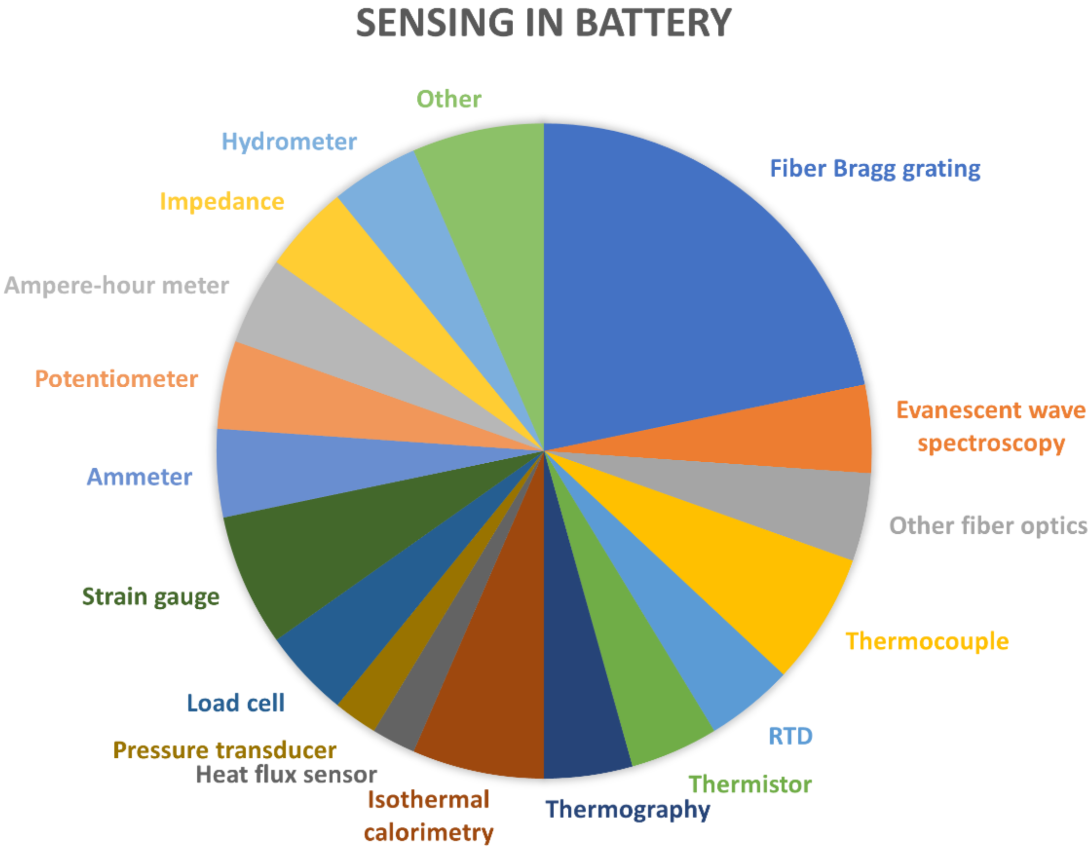


Figure 13. A glance at available sensing technologies for battery modules and systems.

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 13). 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.

7.3.2 Challenges

Numerous sensing technologies for battery modules and systems have been tried (Figure 13), 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.

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 (100µm) RTDs[92]. 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 25Ah 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 [93]. 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 [94]. 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[95]. 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.[96] 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.[97] 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.

A solution to this decoupling issue has been provided by the arrival **of microstructured optical fibres (MOFs),** also known as photonic crystal fibres (PCFs)[98]. 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 nano scale, such as SEI growth, lithium intercalation/deintercalation, and local ion concentration variations[99]. However, making such sensors requires the deposit of a metallic plasmonic nano structure 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 behavior 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[100]. **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.

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 communication between sensors and BMS systems. The communication interfaces must be viewed as an integral part of the sensor, and must be taken into account during the co-design of sensor and cell. One challenge concerns having the sensor information provoke autonomous reaction by the BMS, which is based on proven cell and battery models and may even be AI based. To realise the potential of this fascinating field, advances in both hardware 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.

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 wireless sensing to bypass the connectivity issues associated with implementing today’s sensors, whatever they are, provided that the noisy environment of the battery can permit wireless communications. It must be realised that adding wires to the cell could make manufacturing so expensive that it would outweigh sensor benefits. A first step 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 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.

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 define the proper transfer functions from sensing to BMS. Increase the operational temperature window by >10% through on-the-fly sensing.

In the medium term**:** Miniaturise and integrate the identified (electro)chemically stable and **multifunction** sensing technologies at the cell level but also in real battery modules, in a cost-effective way compatible with **industrial manufacturing** processes. 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 wireless communication between sensors and an advanced BMS relying on new AI protocols 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.

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.[101–108] Besides the absolute need to develop sustainable batteries, our increasing dependency on batteries calls for great efforts to ensure their reliability.[109,110] Detection of irreversible changes (sensing) is a first step towards better reliability. However, to really ensure reliability, the cell should be able to automatically sense damage and also to reinstate the virgin configuration together with its entire functionality.[89] A self-healing programme must thus be developed hand in hand with the sensing one. The ability to repair damage spontaneously is an important survival feature in nature, as it increases the lifetime of most living organisms. So a burning question is raised: Can we try to mimic natural healing mechanisms to fabricate smart and long-life batteries?[111] 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."[112]

As in the medical field, which heavily relies on the vectorisation of drugs for the treatment of diseases,[113,114] 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 14).[115–119] 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 QRL, 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 (Figure 14), enabling Europe to take worldwide leadership in BSH.

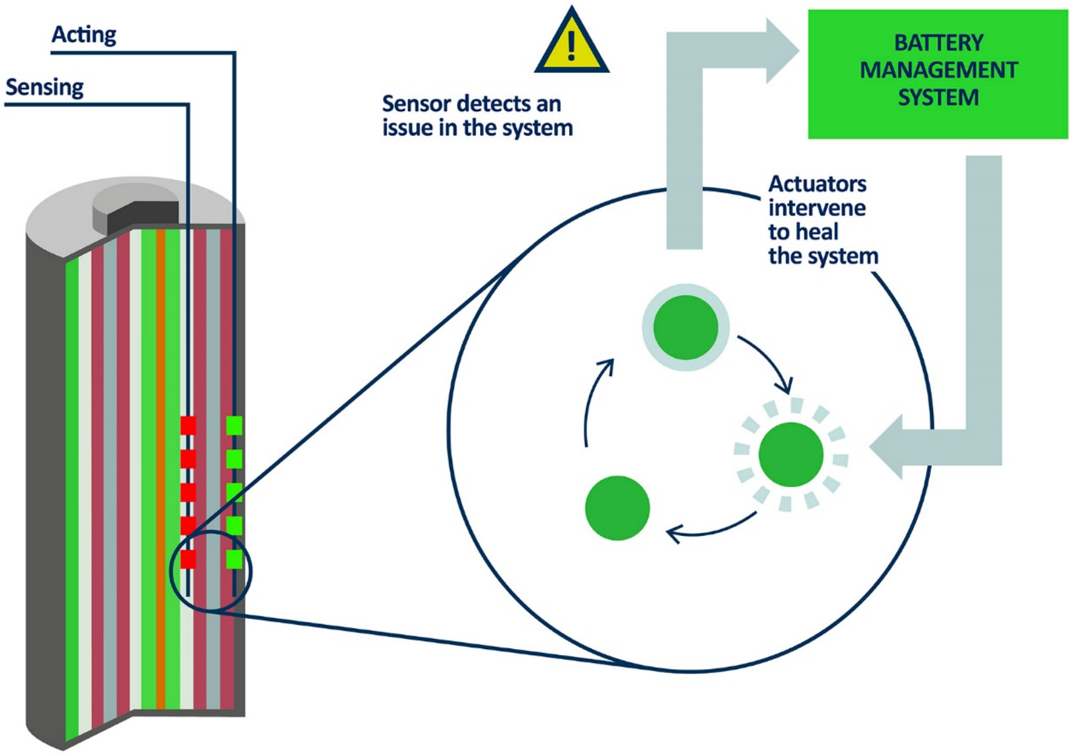


FIGURE 14. The synergy between sensing, BMS, and self-healing.

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.

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[120]. 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[121,122]. Functionalised and flexible polymers that are chemically compatible with battery components have been developed, 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[123]. A plethora of self-assembling materials [124–127] 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.

To protect batteries from thermal mismanagement (the most common failure mode), different approaches have been pursued that include thermo-switchable polymers with thermal self-protection integrated into the electrolytes and current collectors.[128–130]

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.

In conclusion, the field of BSH is rapidly gaining momentum, as shown in Figure 15.

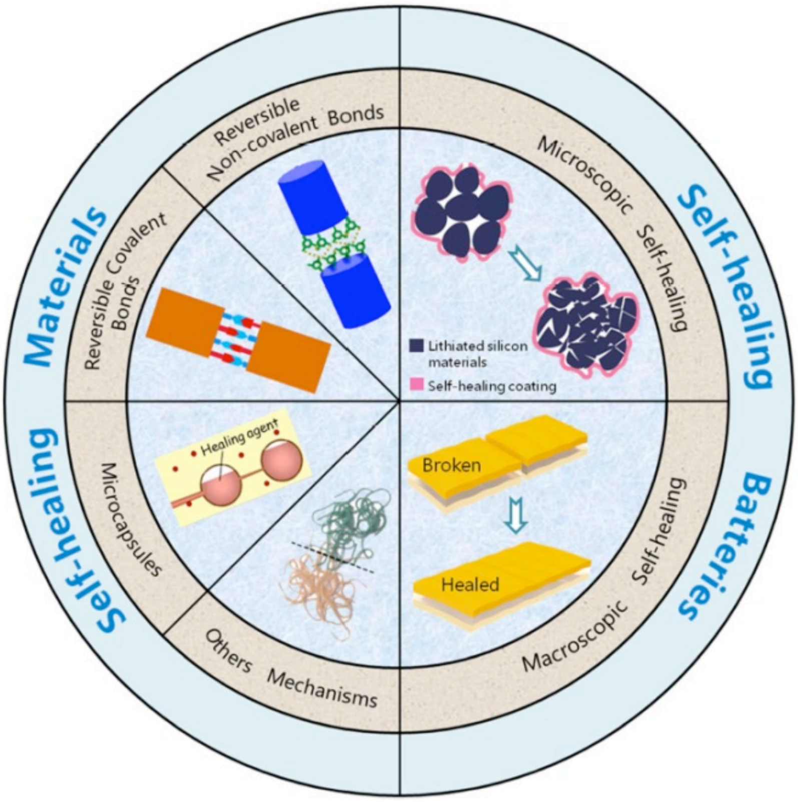


FIGURE 15. Schematic of self-healing mechanisms in battery material.[121]

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 (Figure 16)[123,131].

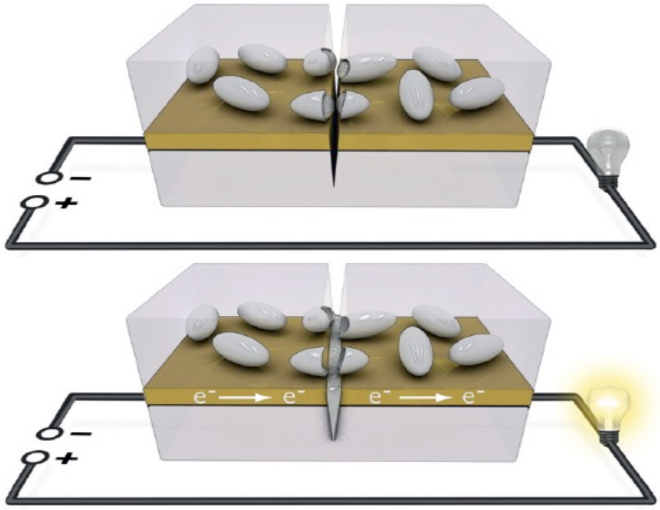


Figure 16. Testing self-healing of the gold line after damage [29].

Other conductive chemical systems, such as carbon-black (CB) dispersions, were similarly encapsulated and tested.[132,133] 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 (Figure 17) that exhibited high stretch ability and sustained the mechanical self-healing repeatability that helped the Si anode withstand large volume expansion after many cycles.[122,134,135] 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.[136] Besides hydrogen-bonded polymers, self-healing binders based on several other supramolecular interactions have also been employed for Si anodes[137–141] and sulphur cathodes.[142] Long-term testing is sorely needed to fully evaluate the practicality of these approaches.

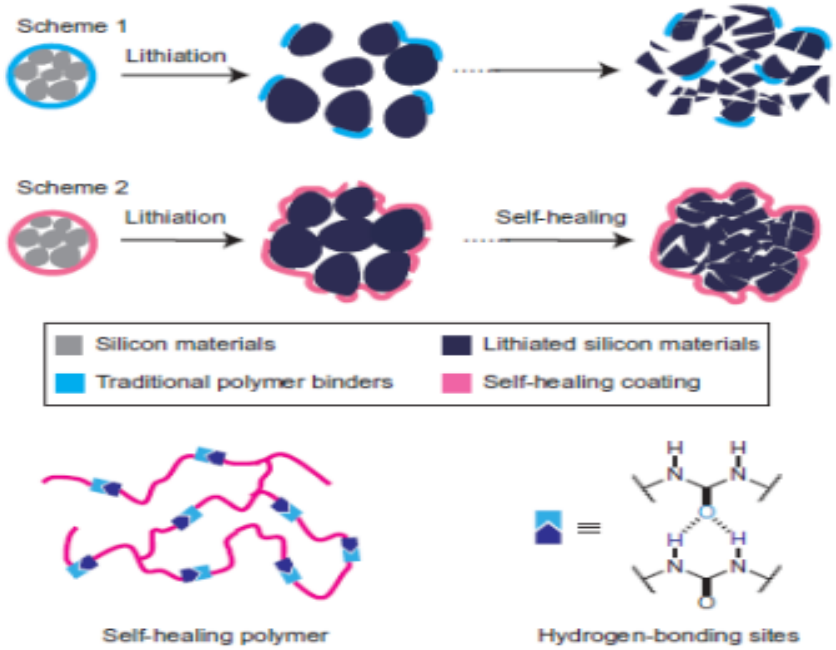


FIGURE 17. Design and structure of a self-healing silicon electrode.[131]

Another auto-repair concept developed by Deshpande et al.[143] 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[144] were shown to have excellent cycling performance (>4000 cycles) and a sustained capacity of 775 mAh/g at a rate of 200 mA/g. Self-healing alloys (Na-Sn) were also implemented by Mao et al.[145] 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.[146] Specific capacitances of 140 Fg^{-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,[126] 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[147]. 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 Li et al.;[148] briefly, 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 $mAhg^{-1}$ and high coulombic efficiency.[149] To further improve the efficiency of Li-S batteries, Zhang et al.[150] 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 [151]. 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.[152]. 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, Koratkar et al. succeeded in achieving substantial self-healing of the dendrites by using a high plating and stripping current (~ 9 mAh cm^{-2})[153]. 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 with which to synthesise the membrane, 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

Ultimately, we aim to develop a system for the on-demand delivery of molecules to solubilise a resistive deposit or to restore either a defective electrode/electrolyte interface in a battery or even the conductive networks within composite electrodes. Since separators are currently a "dead" component of the battery, we will use them as our toolbox for exploring the on-demand administration of healing agents. BATTERY 2030+ will not rely solely on autonomous self-healing tools (e.g., self-healing polymers and liquid–metal alloys). It will go beyond these and include the implementation of 3D porous multifunctional material composites, capsules, supramolecular species, and polymers capable of receiving specific molecules and releasing them on demand in response to physical or chemical stimuli to repair the "tissue" constituting the electrode/electrolyte and particle/particle interfaces. The development and implementation of on-demand 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. We must be bold and open-minded to tackle these new challenges while constantly keeping in mind battery constraints in terms of the chemical environment and manufacturing.

There is now 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: Establising a new research community that includes a wide range of R&D disciplines to develop self-healing functionalities for batteries. Engineer functionalised separators and develop supramolecular assemblies relying on H-H bonding for reversible crosslinking to repair electrode–membrane fracturing while being compatible with the targeted battery chemistry. Develop different on-demand 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.

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 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. 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.

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.

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.[21] Other commercial battery technologies exist as well (e.g., lead acid), 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. Additionally, there are some incipient activities in which modelling is applied to cell design,[154,155] which will open up new opportunities to explore new cell formats and designs.

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. The three main phases are included: electrode production, cell assembly, and cell finishing. They all comprise several steps, such as mixing, coating, drying, slitting, calendering, vacuum drying, and electrolyte filling.[156]

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

- Accelerate new cell designs in terms of performance, efficiency, and sustainability. Couple multiphysics 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 18).
- 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 19).

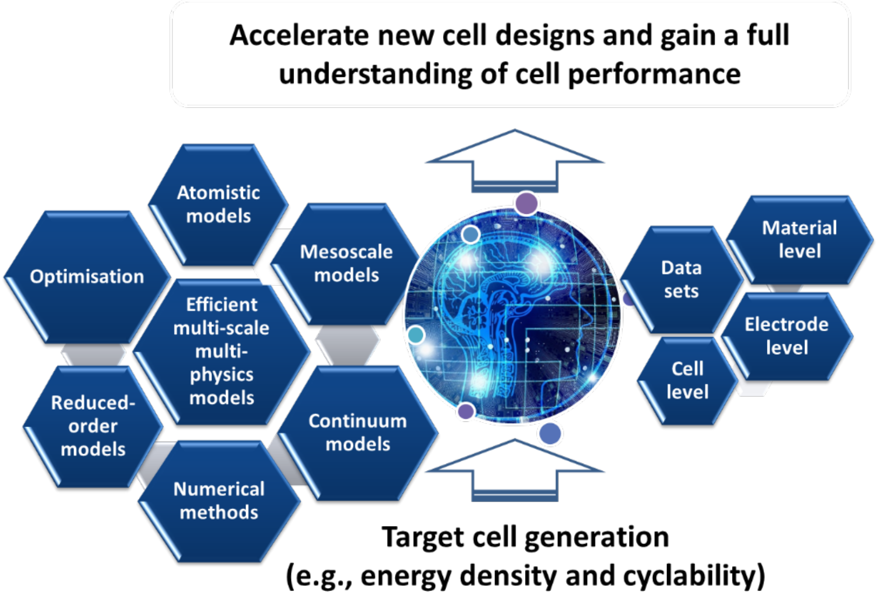


FIGURE 18. Inverse cell design based on digital twin of a cell.

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[157] 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.

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

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 for future cell design and manufacturing processes can be summarised as follows:

- new functionalities, such as self-healing materials/interfaces, sensors or other actuators, cell eco-design, and alternative cell designs
- a digital twin of inverse cell design, providing disruptive cell design capable of meeting performace targets (e.g., for energy, power, and cyclability)
- flexible and scalable manufacturing processes, as well as flexible, high-precision modelling tools for the optimisation of processing conditions and machine parameters, to minimise human labour, trial and error, and waste products; real-time models for the processing of electrode pastes and their performance in the cell (i.e., a digital twin of cell manufacturing)
- validated multiphysics and multi-scale models coupled to AI algorithms of cell manufacturing processes capable of providing an accurate understanding of each step of the process
- a digital twin of the manufacturing process: manipulating the complete virtual representation can actuate the physical world, improving the control of manufacturing facilities and processes

7.5.4 Forward vision

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 19). 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.

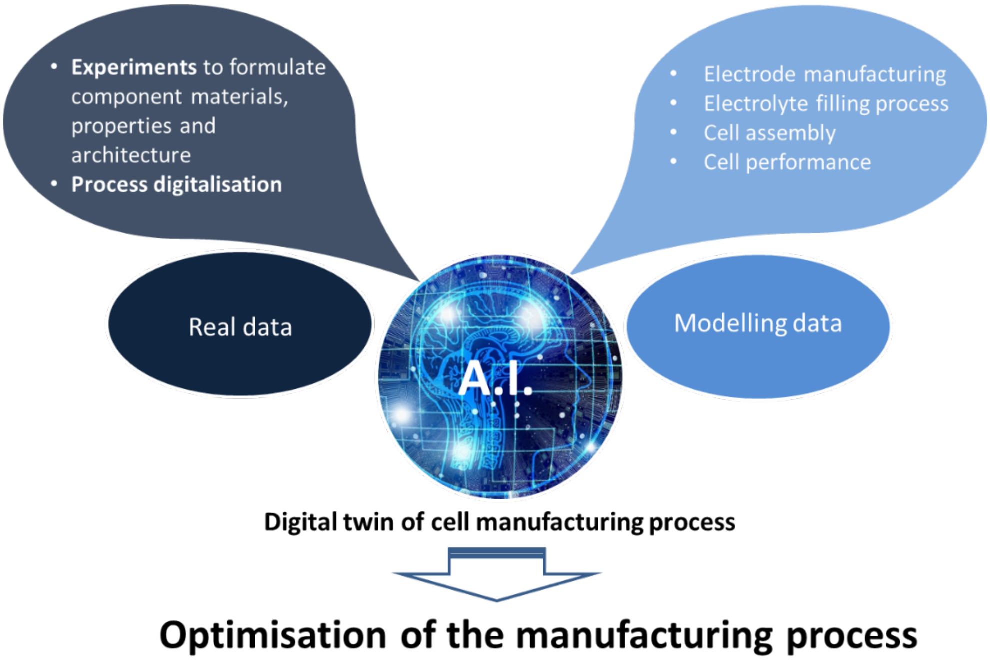


FIGURE 19. Digital twin of cell manufacturing processes.

The main benefits of this approach are as follows:

- gives new optimised cell designs for specific applications/cell chemistries
- develops new manufacturing methodologies
- develops models that calculate ultimate manufacturing parameters
- improves battery performance (e.g., power and energy density) through advanced design
- speeds up processing – rapid manufacturing and prototyping
- improves quality control
- provides an appropriate link to cell design, materials, and BIG–MAP, for example, by adding manufacturability and recyclability aspects as input parameters for the autonomous materials search, while MAP proposes design rules for manufacturing

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 physical 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, interfacing with BIG–MAP to form a fully autonomous system (using AI) (see Figure 20). Based on the BIG–MAP output, an automated cell prototype will be generated. In addition, 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.

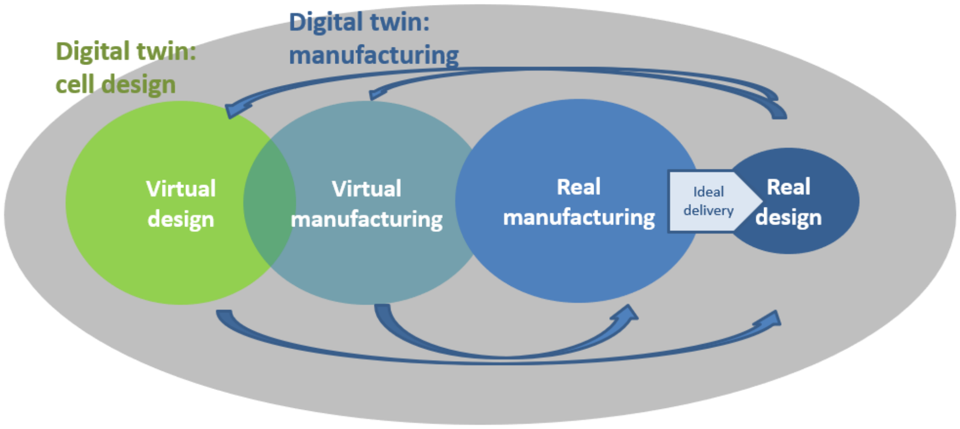


FIGURE 20. AI-driven design and manufacturing methodologies linked together as a whole.

Potential impacts of this approach:

- Accelerates the discovery of new cell designs and manufacturing processes; reduces the development time and cost for new battery cells; reduce battery research and innovation (R&I) cost.

Potential challenges of this approach:

- Data management (usable, accessible, integrated, and curated); data harmonisation (standards); intellectual property management (data ownership); the proposed methodology will not outperform SoA in some aspects, the outcome will work but may not be accepted by industry because it is too complicated/expensive (see the very conservative/price sensitive lead-acid battery industry)

***In the short term*:** The approach will be implemented starting from state-of-the-art information, and the focus will be the battery cell design methodology. This will include the improvement of simulation tools, such as multiphysics models, with the goals of reducing the computational burden and implementing current AI techniques through deep learning and machine learning methods for cell design. Start launching and implementing current AI-driven methodology to the LiB manufacturing steps. Additionally, the improvement and up-scaling of new manufacturing processes (e.g., 3D printing and dry processing) are foreseen.

***In the medium term*:** A proof of concept of a digital twin of a LIB cell design as well as proof of concept of a cell manufacturing process are expected. Input from other research areas BIG–MAP, sensing, self-healing and recycling will be integrated into the process. The methodology will be adapted to the manufacturability of new battery technologies, with the launch and implementation of the AI-driven methodology for manufacturing after developments in cell-level design and in new innovative manufacturing processes. Develop scalable battery chemistries, for example, multi-valent and organic chemistries. Demonstrate the transferability of the established BIG–MAP concept to alternative battery concepts (e.g., flow batteries).

***In the long term*:** The overall AI-driven methodology will reach full maturity and implementation by integrating cell design sub-loops that converge in holistic prototype development, forming a fully autonomous system supported by BIG–MAP. This methodology, which will contribute to the foundation of a new state of the art, developed as a commodity, will be progressively deployed to industry and academia.

7.6 Cross-cutting area: Recyclability

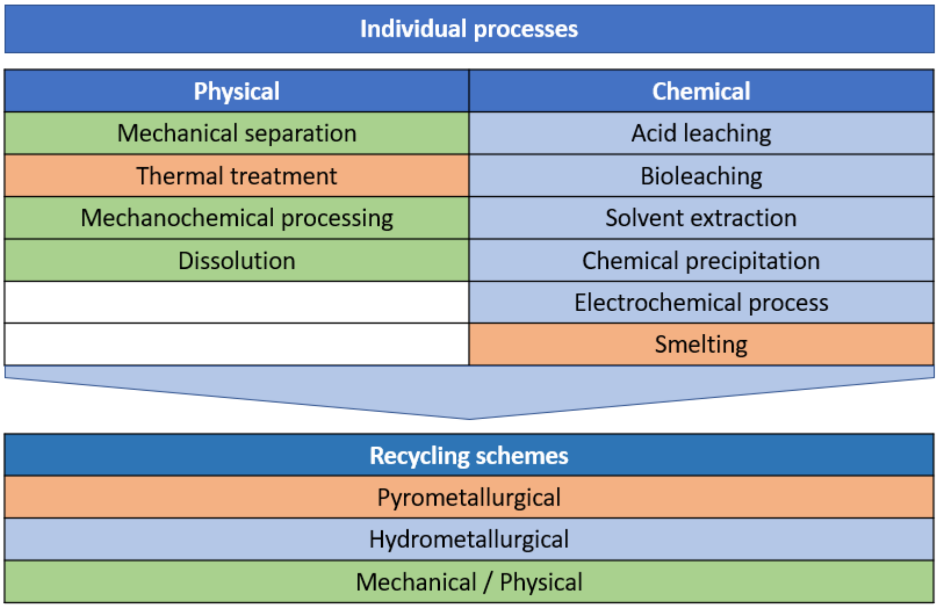
The development of battery dismantling and recycling technologies with high efficiencies going well beyond the EU Battery Directive 2006/66/EC target of 50% for most battery technologies is essential to ensure the long-term sustainability of the battery economy by 2030. This calls for new, innovative, simple, and low-cost processes targeting a very high recycling rate, small carbon footprint, economic viability, as well as logistics and business incentives. One technical approach will be the direct recovery of the active materials and single, instead of multistep 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 and economic and social aspects considering the whole life cycle), (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).

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), 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 this 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.** A revision of the Battery Directive is expected to be published by 2020 with updated categories and recycling efficiencies.

Currently, pyrometallurgy is the most applied method. 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 21.



Individual processes

Physical	Chemical
Mechanical separation	Acid leaching
Thermal treatment	Bioleaching
Mechanochemical processing	Solvent extraction
Dissolution	Chemical precipitation
	Electrochemical process
	Smelting

Recycling schemes
Pyrometallurgical
Hydrometallurgical
Mechanical / Physical

Figure 21. 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 to be developed 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. Closed loops will also decrease the dependency of the EU on critical metal imports.

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.

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 the 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. The quality of the recycled products could be a hurdle to closed-loop recycling.

- 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 notion of **"direct" recycling** processes, though 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 out-dated when recycled, (e. g. $LiCoO_2$ or NCM-111 cathode powders introduced ten years ago). These reconditioning processes are not currently available.
- Methodological challenges: the economic, ecological, and social impacts of emerging battery technologies must be analysed and estimated in a prospective manner. All material, component, and cell developers as well as recyclers and other stakeholders need to work together in an interdisciplinary way, to reach shared visions of 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. Standards for identification are important on the battery and cell levels in order to overcome these challenges.
- New battery technologies seem likely to enter future markets, for example, solid-state, lithium-sulphur, redox flow, and metal-air batteries in mobility and stationary applications. Proposed new recycling processes to cope with all these chemistries (and related BMS) will create new process challenges; for example, the presence of Li metal will affect safety aspects of the recycling processes. Recycling processes may have to be redesigned, for example, to use an inert gas atmosphere, depending on the battery type.
- 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 standardisation cannot be expected given the multiple applications on the market, so chemistry identification and quality sorting will become even more challenging. The required level of expertise can only be expected if advanced AI development 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 possibly at the local level with information from sensors. Processes to handle information from these innovations during the recycling phases will have to be developed and standardised. 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 decentralised collection and recycling processes/units need to be established, and societal acceptance issues need to be considered.
- A legislative framework must be established to foster/safeguard sustainable design, including design for recycling.

Tentative longer-term challenges

- 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 (decentralised, 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 maximise the number of individualised 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 “Eco-design preparatory study for Batteries” has the goal of providing 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). Sustainability is addressed within this description, but social aspects are not considered. Moreover, the outcome of the study considered only a limited number of chemistries and application fields.

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 minimise 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 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 summarised as shown in Figure 22.

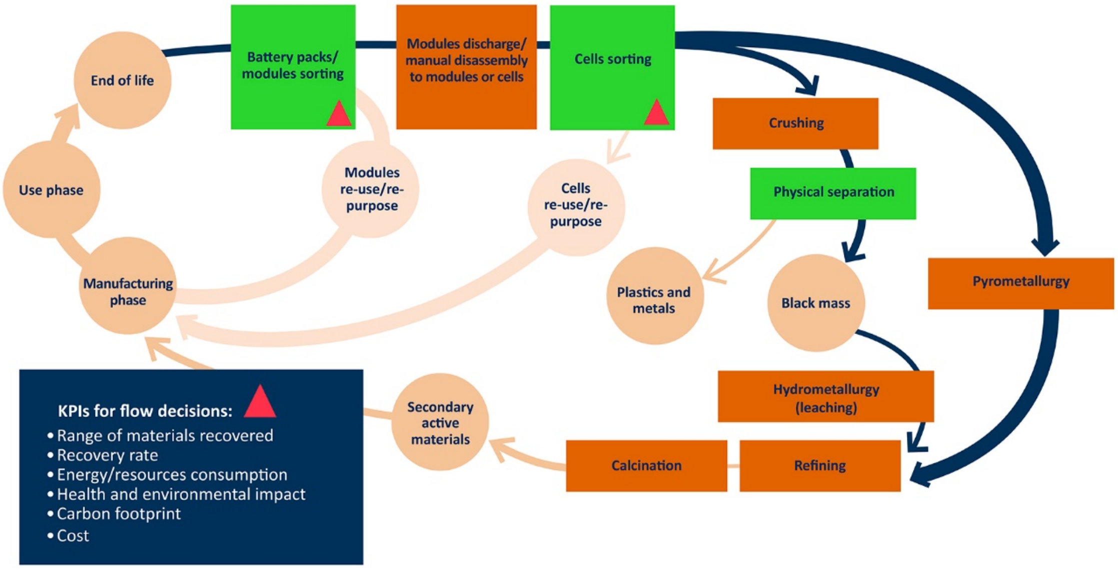


FIGURE 22. Present recycling process.

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* (Figure 23) based on:

- data collection and analysis (e.g., from labels, BMS, and sensors)
- modern small-carbon-footprint logistics concepts, including decentralised processing
- automated pack disassembly to the cell level
- investigating reuse and repurposing wherever possible
- automated cell disassembly to maximise 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
- international collaboration to be stimulated and developed

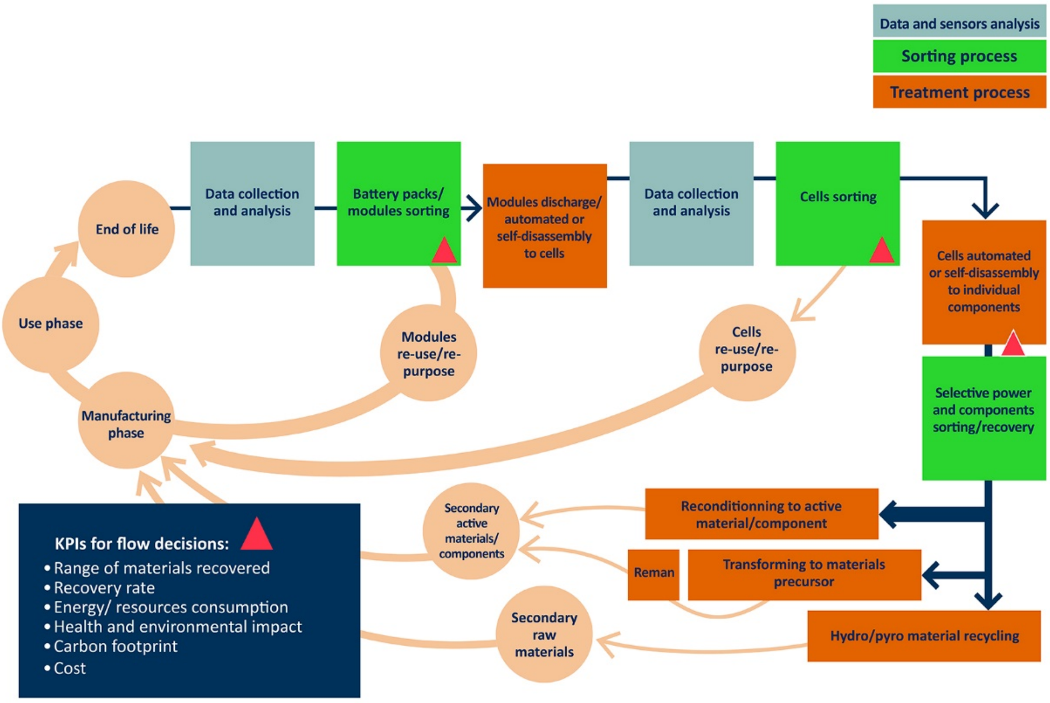


FIGURE 23. Future recycling process: direct recycling fully integrated with reuse.

In conclusion, over a time frame of ten years, we will develop a circular model incorporating specific R&I actions, such as preparing a battery design for maximum longevity as well as considering recalibration, refurbishing, and suitability for second-life applications and multiple usage. Integrated sensing and possibly self-healing concepts can be used to identify damaged/aged components and prepare for their reuse. The model will also include the development of concepts for traceability (e. g. via material finger prints and blockchain technologies, etc.), especially of critical raw materials (CRMs) throughout the cell life, automated cell sorting and evaluation, and the development of efficient, single-step, cheap, and sustainable processes for recovering valuable and critical materials. AI and sorting equipment will need to be applied in selective recycling processes, but versatile processes applicable to any battery technology will also be sought. The same approach to maximally recover battery components will be targeted even in the case of metal-air and other chemistries.

7.6.4 Forward vision

The new recyclability process 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 24).

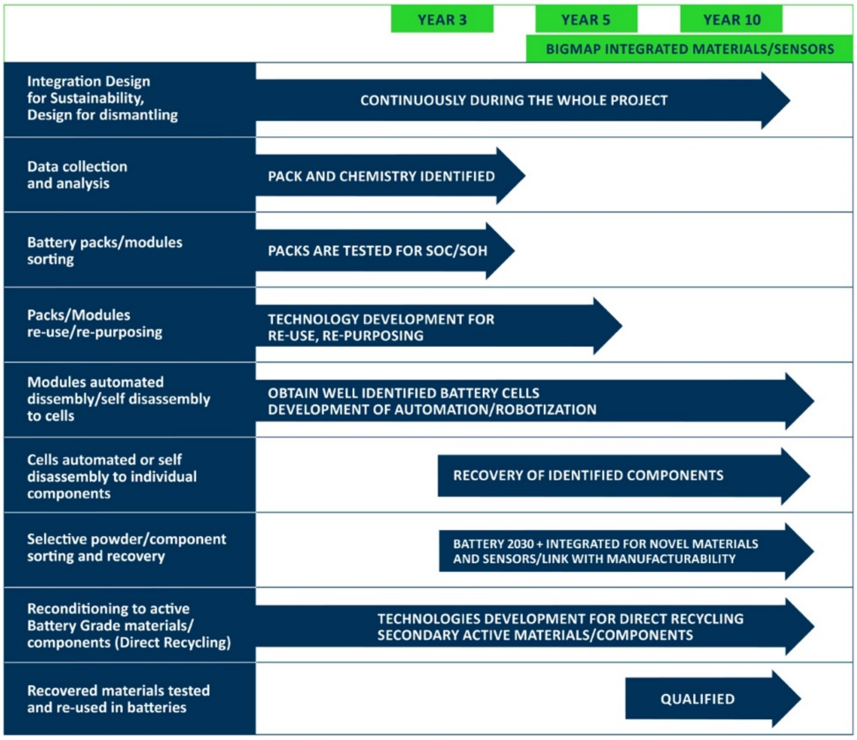


FIGURE 24. The ten-year roadmap for recyclability within BATTERY 2030+.

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.

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.

Impact

The recycling of LIBs from vehicles is still a developing business, with large volumes expected to be recycled as we approach 2030. Since current small volumes make recycling a cost-intensive industry, recyclers struggle to find the best balance between economics and meeting recovery targets, resulting in the industry not yet focussing enough on high efficiency and low emissions. The BATTERY 2030+ recycling programme will help prepare industry to treat the expected large future volumes in a responsible, sustainable, and economically viable way.

8 Abbreviations and glossary

AI	Artificial intelligence
AIMD	Ab initio molecular 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
CNT	Carbon nanotube
DoE	Department of Energy, USA
EARPA	European Automotive Research Partners Association
EASE	European Association for Storage of Energy
EBA	European Battery Alliance
EMIRI	Energy Materials Industrial Research Initiative
EMMC	European Materials Modelling Council
Energy density	Energy per unit volume (Wh/l)
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
LCA	Life cycle assessment
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
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
PCF	Photonic crystal fiber

QRL	Quality, reliability, and lifetime
RE	Reference electrode
SEI	Solid electrolyte interphase
SET PLAN	Strategic Energy Technology Plan
Specific Energy	Energy stored gravimetically, Wh/kg
SWCNT	Single-walled carbon nanotubes
SoC	State of charge
SoH	State of health
SP	Sensor plasmonics
Specific energy	Energy per unit mass (Wh/kg)
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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