Commodity TopNews
Fakten ● Analysen ● Wirtschaftliche Hintergrundinformationen
67

RECYCLING OF LITHIUM-ION BATTERIES IN GERMANY AND EUROPE

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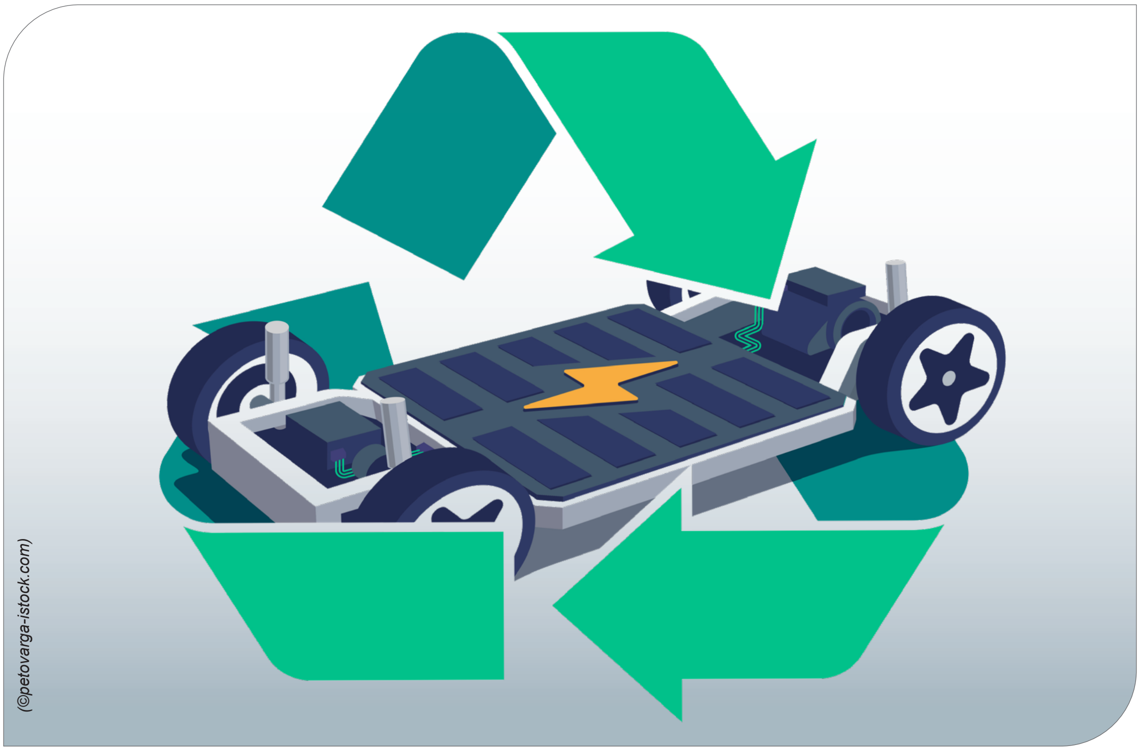

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Electric car chassis with battery pack

INTRODUCTION

With its Green Deal, the EU is aiming for climate neutrality by 2050 (EU 2019). A first step by 2030 will be to reduce greenhouse gas emissions by at least 55 % compared to the 1990 level. The structural changes required for this decarbonisation will affect all industries, above all the energy and transport sectors. Road transport generates around a fifth of all greenhouse gas emissions in the EU (DESTATIS 2022). In order to meet its emission target, the EU Commission is planning a ban on the sale of new cars with internal combustion engine (ICEs) from 2035. The electrification of mobility, i.e. replacing technologies and systems running on fossil fuels with alternative, low-emission forms of mobility such as battery electric vehicles, is very much in the focus of the EU. Lithium-ion batteries (LIBs) are currently the major battery type commercially available for applications such as consumer electronics, electric mobility and energy storage systems in combination with solar parks or wind farms. LIBs have many advantages over, for instance, lead-acid batteries: they have a high energy density, can be quickly charged, provide a consistent capacity (having a low memory effect) and high efficiency (Batterie-Forum Deutschland 2022). With the implementation of the EU's Green Deal, LIB production will rise, particularly for electric mobility (EU 2019). Most battery raw materials such as lithium (Li), cobalt (Co), nickel (Ni), graphite and manganese (Mn) originate from primary resources, whose production has a considerable carbon footprint. Attention is therefore increasingly focused on the recycling of LIBs and the secondary raw materials or recycled waste materials that can be gained from them.

MARKET DEVELOPMENT OF LITHIUM-ION BATTERIES

Since the first LIBs for mobile electrical devices were launched in 1991, they have been continuously developed and rolled out to a wide range of applications such as power tools, electric cars and energy storage systems (Table 1, Fig. 1).

In view of the ramp-up of electric mobility forecast, this market will continue to be the main application for LIBs (Fig. 1). In 2019, the electric mobility industry recorded considerably higher growth in Europe than in other regions (EV Volumes 2022). The Northern European countries and the Netherlands were leading in this development: in Norway, electric mobility had a 56 % market share, and two of the ten best-selling cars in the Netherlands were battery electric vehicles (BEVs) (VDA 2021). The United Kingdom and other countries recorded a 3-digit percentage growth in that area for the year. The main drivers of this development were government policies, generous subsidies, tax incentives for BEV, and a shift in consumer attitudes, mainly due to growing concern about climate change.

***Tabelle** 1: Main areas of application for LIBs and annual growth rates between 2006 and 2021 (Avicenne Energy 2021).*

Application	**Growth rate in % (2006 – 2021)**
Consumer electronics	8,2
Power tools	23,4
Electric cars	88,0 (since 2010)
Stationary energy storage systems (ESS)	75,3 (since 2008)
Electric buses (China)	82,7 (since 2012)

Actions to combat climate change have been prioritised by a number of European governments. Many countries have already changed their target date for a ban on the registration of new cars with ICEs. The United Kingdom has committed to carbon neutrality from 2050 and proposed a ban on the sale of all vehicles with ICEs from 2030. Norway is planning implementation of this goal earlier, in 2025. According to the German Environment Agency's climate balance, emissions in Germany from the transport sector amounted to 146 mt of CO_2 in 2020.

Germany is aiming for a reduction to no more than 85 mt of CO_2 by 2030. The Federal Government is expecting seven to ten million new registrations of electric vehicles in Germany by 2030 (Bundesregierung 2022). Despite the growth in 2019, there have been obstacles to the introduction of electric vehicles on a wider scale: the availability of only a limited number of models on the European market, and consumer perception of an inadequate charging infrastructure in some regions.

The continued strong growth in the global electric vehicle fleet will nevertheless lead to an increase in demand for battery raw materials, with the issue of recyclability of batteries gaining in importance as a result.

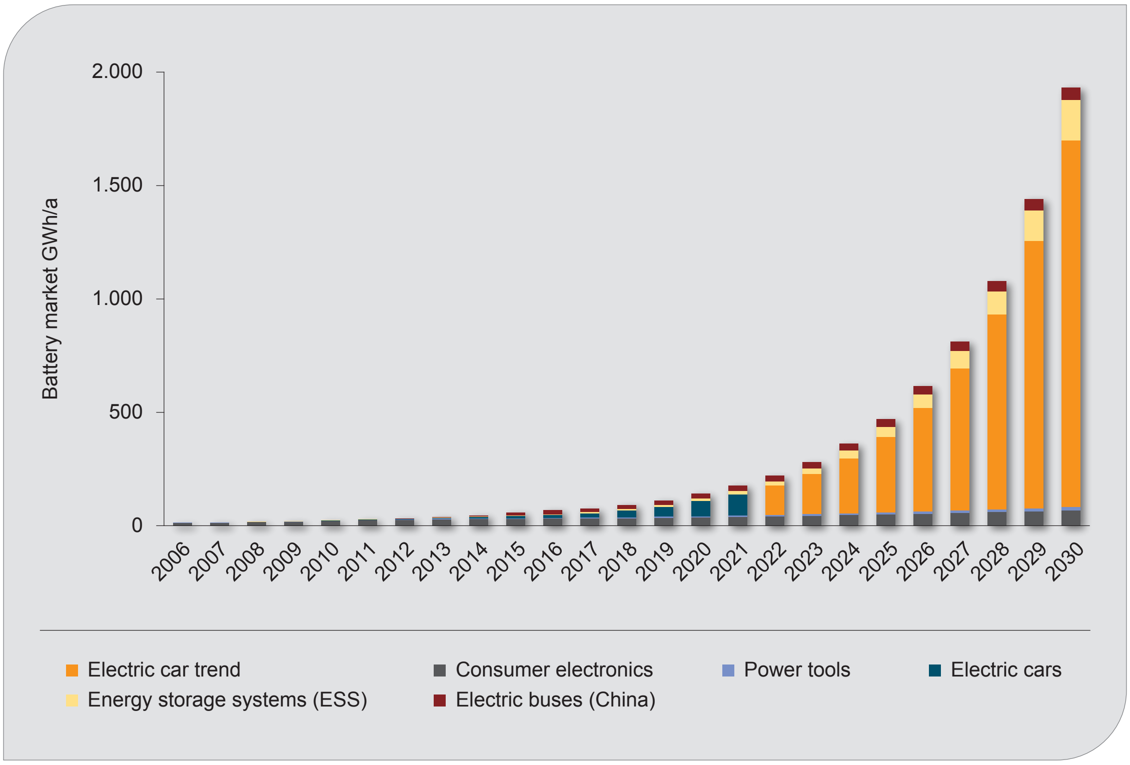
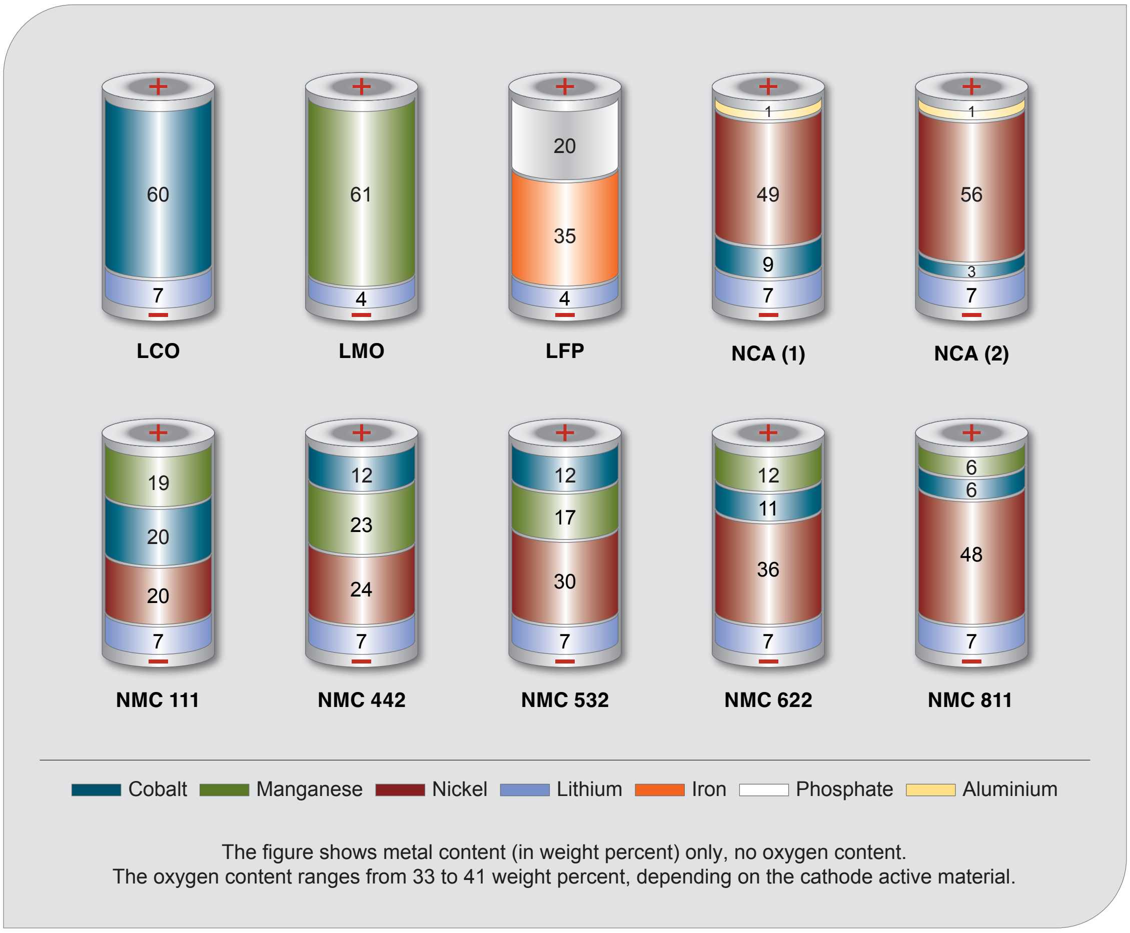


Fig. 1: *Global trend of the lithium-ion battery market by application (source: Avicenne Energy 2021). The future trend for electric cars until 2030 is highlighted in orange.*

TYPES OF LITHIUM-ION BATTERIES

Lithium-ion batteries have become the battery system of choice for many manufacturers of electric vehicles. The term “lithium-ion battery” covers various types of LIB, which differ mainly in the chemical composition of the cathode (KLIB 2022). Depending on the cathode type, the main constituents of an LIB are lithium, cobalt, nickel, aluminium, iron, manganese and phosphate (Fig. 2).

The anode material is usually graphite. The main LIB types for electric vehicles are lithium nickel manganese cobalt oxide (NMC) and lithium nickel cobalt aluminium oxide (NCA). As technologies have evolved, the relative content of nickel, manganese and cobalt in the NMC and NCA types have changed over time. While in first-generation NMC types (NMC 111), the nickel, manganese and cobalt content were the same, this has changed over time to a ratio of 8:1:1 of nickel, manganese and cobalt (NMC 811). An increasingly important type, particularly for low-cost BEVs, are lithium iron phosphate cells (LFP).



The figure shows metal content (in weight percent) only, no oxygen content.
The oxygen content ranges from 33 to 41 weight percent, depending on the cathode active material.

Fig. 2: *Lithium-ion battery types and composition of their cathode active material (DERA calculations).*

BATTERY RECYCLING

It would be possible to improve the CO_2 balance by increasing the recycled share of battery raw materials. The recycling of LIBs is a challenging task, because it is a technically complex process that is associated with safety risks, particularly the risk of explosion and fire. The use, disposal and recycling of LIBs are governed by the EU's Batteries Directive.

Legal framework, strategies and incentives

The EU's 2006 Batteries Directive rose to the challenges of its time, monitoring a range of dangerous substances such as lead, cadmium and mercury in portable and industrial batteries (European Commission 2022). However, the Directive was drawn up at a time when supply chains had a different role and the exponential growth in large LIBs for electric vehicles was not conceivable. The proposal for a new Batteries Directive responds to the challenges of today. It specifies labelling rules, information obligations and supply-chain due diligence standards, and requires metal-specific recycling rates and the use of recycled content in batteries with a capacity above 2 kWh, most of which are used in electric vehicles. And for the first time, information on the carbon footprint of battery production has to be stated. The EU objectives that will be enacted in the new Batteries Directive concern the recycled content and recovery rates for certain metals, such as cobalt, nickel and lithium. Proposed in 2019, the new Directive may take effect in 2023 (Fig. 3).

The European Battery Alliance was launched in 2017 with the aim of creating a complete, globally competitive and sustainable value chain for batteries in the EU. Together, the Commission, the member states, the European Investment Bank and stakeholders from industry and research determine objectives and actions, and act as a catalyst for the rapid development of a battery ecosystem. Only recently, in February 2022, the European Battery Alliance set up the European Battery Academy, whose aim it is to foster skills for the fast-growing battery ecosystem in Europe.

The EU's Batteries Directive has been enacted into German law by the Batteries Act, which governs the placing on the market, collection and environmentally compatible disposal of batteries and accumulators. 2021 saw a major amendment of the Batteries Act (BattG2). Following the new EU Batteries Directive, further changes can be expected from about 2022.

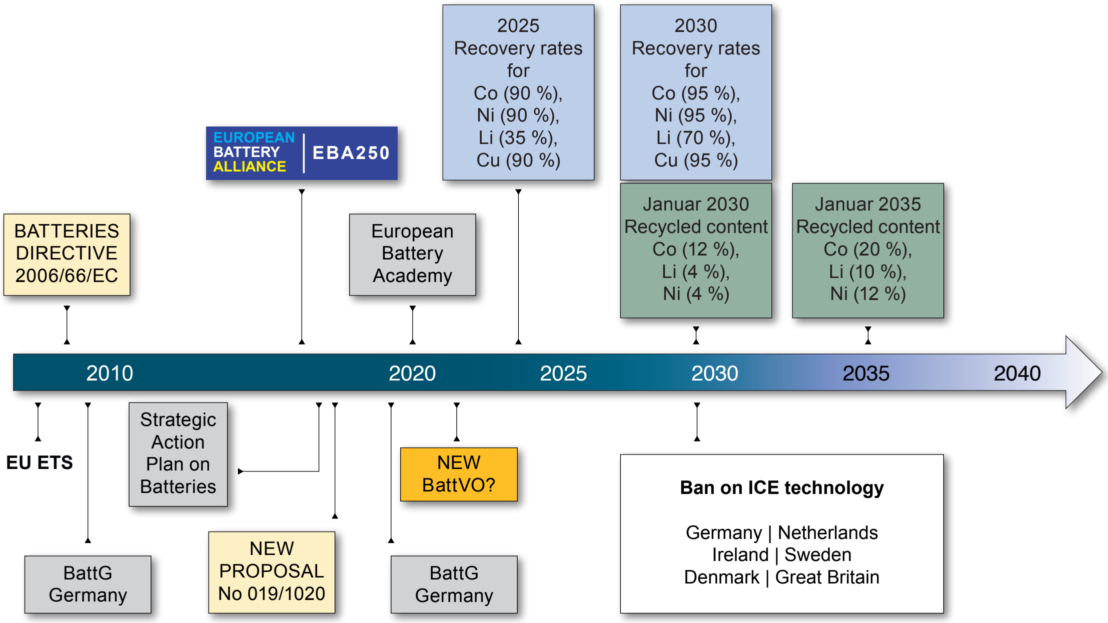


Fig. 3: *Development of legal frameworks since the introduction of the EU's Batteries Directive in 2006.*

An additional legal framework that could in future have an impact on the recycling of LIBs is the European Union's Emissions Trading System. At present, it has little impact, but that could grow as carbon prices rise. It is, however, not yet clear what price level would have a significant incentive effect on recycling; what recycling processes have a competitive advantage; and what impact the carbon price will have on the competitiveness of the European LIB recycling industry. The European Commission is considering applying the "battery approach" also to end-of-life vehicles, and to waste electrical and electronic equipment, for which there are a large number of recycling companies.

The Federal Ministry of Education and Research (BMBF) has been funding various battery recycling-related projects in Germany since 2014. Current research focuses on the processing of raw materials and the digitalisation of recycling processes (BMBF 2021). As part of the Recycling & Green Battery competence cluster, 16 projects at 34 German university and research institutes are studying battery recycling.

Recycling facilities in Europe

Despite the rapid growth in consumption and the increasing prevalence of BEVs, recycling is very limited, since the quantity of recoverable material depends not on the current annual battery production level, but on past years of production, the product types and the product life spans.

Generally speaking, the process for the recycling of LIBs comprises these steps:

- *Registration/collection of end-of-life batteries*
- *Preparation: sorting, disassembly, discharging*
- *Thermal and/or mechanical pre-treatment*
- *Main processes: pyrometallurgy and/or hydrometallurgy to produce marketable refined metals or metal salts*

The European market for LIB recycling is at an early stage with a large potential for growth. Among the established industrial recycling facilities in Germany and Europe with an annual capacity of > 2,000 t of batteries are Umicore, Accurec, Nickelhütte Aue, AkkuSer and Duesenfeld. Pilot plants with an annual recycling capacity of < 2,000 t include TEM, SNAM, Volkswagen and Primobius. Lithium-ion batteries are a difficult feedstock for recycling plants, particularly with regard to corrosion, slag properties, and their energy and mass balances. The recovery of lithium is currently still a challenge, because lithium can corrode the fireproof materials in standard furnaces for cobalt, copper and nickel. During the processing of LIBs, e.g. lithium forms part of the slag and can be recovered using hydrometallurgical processes. The lithium content of these slags is comparable to that of spodumene concentrates. According to information in the patent originating from the LiBRi research project, a total lithium yield of around 90 % from slag could be achieved, which is comparable to the yield from spodumene concentrates (Brückner et al. 2020).

The processes used by Umicore, Accurec and Duesenfeld are described below.

Umicore

Umicore’s industrial-scale pilot recycling plant in Hoboken, Belgium has been operational since 2011. It has a total permitted annual capacity of 7,000 t of LIBs. The Umicore process combines pyro- and hydrometallurgical steps but does not include mechanical pre-treatment. Only large industrial batteries have to be disassembled to the module or cell level. The pyrometallurgically generated metal alloy is treated (further) using a hydrometallurgical process to recover cobalt, nickel and copper. Lithium can be recovered from slag containing lithium and manganese (Sojka et al. 2020; Umicore 2022). Umicore have announced introduction of the latest generation of their recycling technology some time in 2022. The aim is to achieve yields of more than 95 % of cobalt, nickel and copper for a wide range of battery chemistries. The technology is also expected to permit recovery of most of the lithium (UMICORE press release 2022).

Accurec

The Accurec process recycles portable and industrial LIBs, including LIBs from the automotive industry. The company’s recycling plant at Krefeld, Germany, operational since 2016, has a total input capacity of around 6,000 t annually. LIBs are sorted as required, dismantled and discharged. They then undergo thermal pre-treatment (pyrolysis) and are mechanically separated in a multi-level internal separation plant. The resulting products are a steel fraction, a copper-aluminium fraction, and an electrode powder that is rich in cobalt and nickel. This undergoes pyro- and then hydrometallurgical treatment to permit recovery of the cobalt and nickel salts and metals. Accurec’s future approach includes the recovery of lithium and graphite (Sojka et al. 2020).

Duesenfeld

Duesenfeld’s recycling plant in Wendeburg, Germany, has been running since 2018, specialising in mechanical separation. Following discharging and disassembly of the LIBs – in some cases to cell level – they are shredded in a multi-level process. Among the end products are firstly black mass, and also steel, aluminium, plastics and electrolyte material. The black mass is treated hydrometallurgically, currently on a laboratory scale, to recover lithium, nickel, cobalt, manganese and graphite (Sojka et al. 2020; Duesenfeld 2022).

Recycling capacities in Europe

Many battery recycling projects have been announced across Europe. Of particular note are the plants of the SMS Group, including the Primobius plant, which promises effective recycling of LIBs. Solvay and Veolia are stepping up their battery recycling partnership and have announced their intention to build a demonstration plant for the recycling of battery materials in France. Stena Recycling have plans for a new battery recycling facility in Sweden with an annual capacity of 10,000 t from 2023. In Central Europe, Volkswagen have recently opened a pilot plant in Salzgitter, Germany, and the recycling company Elemental Holding has announced plans for a new plant for the processing of batteries in Poland. Endesa and Urbaser in Spain have said they will start up their own battery recycling plant in the city of Cubillos del Sil in 2023, with an annual capacity of 8,000 t of batteries. More plans for the building of recycling plants have been announced, for instance, by Northvolt, who are planning to commission a plant in Norway with a capacity of 125,000 t per year by 2030. BASF are proposing to build a plant in Germany, while another, by the Glencore and Britishvolt joint venture, will be located in Britain. Both of these are to be commissioned in 2023 and 2024, respectively. Europe currently has an annual recycling capacity of around 100,000 t, which could be increased to 220,000 t by 2023 with the new recycling plants announced. In Germany, LIBs are at present recycled at nine sites, with an annual capacity of circa 49,000 t (Fig. 4).

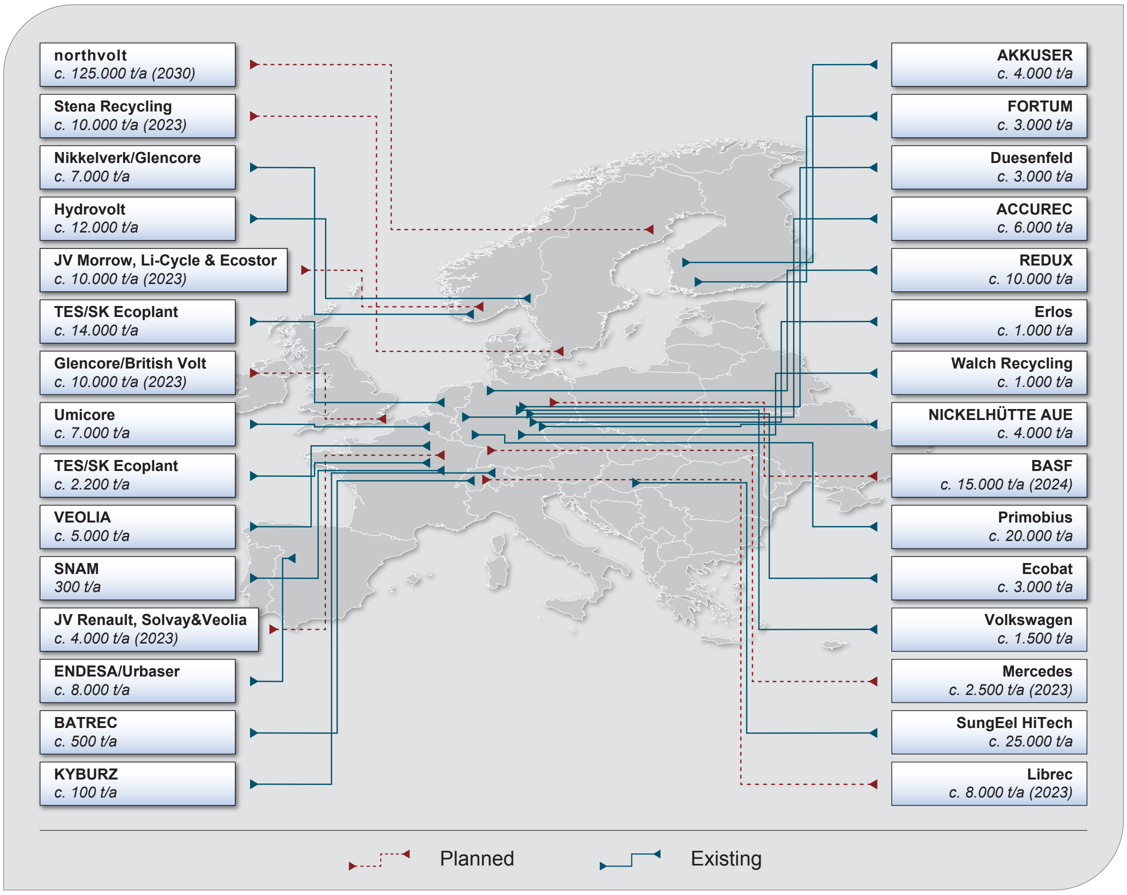


Fig. 4: *Overview of recycling capacities in Europe. Existing and planned (dotted lines) plants (based on data from the respective companies' press releases).*

Assessment of recycling potential of lithium-ion batteries

In order to quantify the recycling quantity of LIBs from the electric mobility sector, we made the following assumptions regarding return rates: 50 % return rate for LIBs after eight years' use; 60 % return rate for LIBs after ten years' use; 90 % return rate for LIBs after twelve years' use; loss of the remaining 10 % through process waste and export losses. Continued use as a second-life battery was not taken into account. This would make the return period for LIBs significantly longer. Recycling rates for nickel and cobalt in established industrial pyrometallurgical and hydrometallurgical processes are between 90 and 95 % (Brückner et al. 2020). A potential recycling rate of 50 % was assumed for lithium, as it is hard to recover in a metallurgical process and economic recovery is not yet possible. For economic reasons, there is as yet no industrial-scale recovery of manganese and graphite either. However, in view of technological innovations, future recycling rates of 20 % are assumed for both manganese and graphite. Based on these assumptions, around 18,000 t of lithium, 169,000 t of nickel, 49,000 t of cobalt, 9,000 t of manganese and 21,000 t of graphite could be made available from the recycling of LIBs in 2030 (Fig. 5). As percentages of 2030 demand, secondary raw materials would theoretically make up 9 % for lithium, 16 % for nickel, 26 % for cobalt, and 3 % for manganese. Breakdowns and full details of these simplified estimates based on statistical uncertainties are provided as part of the BMBF-funded BatMix project.

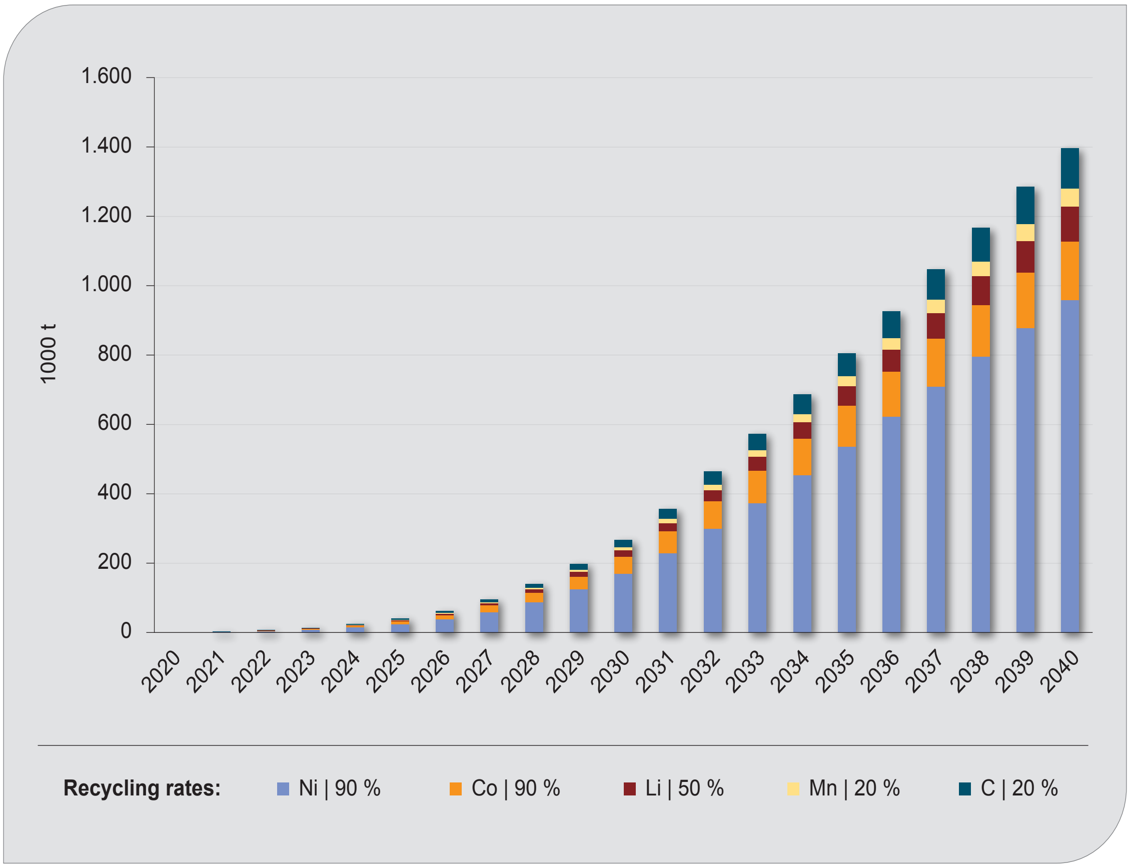


***Fig. 5:** Recycling quantities of battery raw materials nickel, cobalt, lithium, manganese and graphite (C) until 2040. Calculations based on raw material demand for electric mobility 2020 – 2030 (Marscheider-Weidemann et al. 2021) and assumptions regarding return and recycling rates.*

Data for the European electric mobility sector show raw material demand in 2030 for around

97,000 t of lithium, 449,000 t of nickel, 65,000 t of cobalt and 100,000 t of manganese. This is based on a total production capacity of 1,000 GWh per year for electric cars.

An assessment of raw material quantities has to take into account that no single LIB type has so far emerged as dominant and it is not yet clear which type will prevail in the future. Estimates of the recycling share and the economic operation of current recycling plants and investments in new plants are therefore relatively uncertain. Potential recycling processes will have to be suitable for all current and future cell types. They will also need to be scalable, to permit economic operation.

FUTURE BATTERY TYPES

Forecasts regarding future market shares of the various battery types are still uncertain, as new materials enter the market. It can take up to 30 years from first reports to commercial sales, depending on research advances and material demand in connection with current energy sources. The first mention of lithium-air and lithium-sulphur batteries, for instance, was in the 1960s and 70s, but these types are still at the laboratory stage. With the LCO and NMC types, on the other hand, it took only ten to 15 years from the patent to commercial market entry. Refining well-established battery chemicals can take up to eleven years, as the example of the NCA type has shown. From 2025 onwards, the launch of new battery types, such as sodium-based, is expected. But it is still not clear which will be the battery of the future, since many crucial factors affect the establishment of new battery types (Fig. 6).

Battery development

Sony Corporation launched its first lithium-ion battery in 1991, an LCO type, this had been preceded by work on early LIBs. The Goodenough laboratory had discovered back in 1980 the capability of lithiated transition metal oxides with a $NaFeO_2$ structure to reversibly store lithium ions at relatively high potentials (Mizushima et al. 1981). Elements such as nickel or cobalt, also in mixtures with manganese, aluminium, iron etc., showed the same capability (Blomgren 2017). This basic idea led to the discovery of three classes of oxide cathode in the 1980s: 1) layered oxides ($LiCoO_2$), 2) spinel oxides ($LiMn_2O_4$) and 3) polyanion oxides (such as today's LFP cathode). Johnson et al. (1998) were the first to describe the lithium-rich NMC cathode material in 1998 and had it paten-

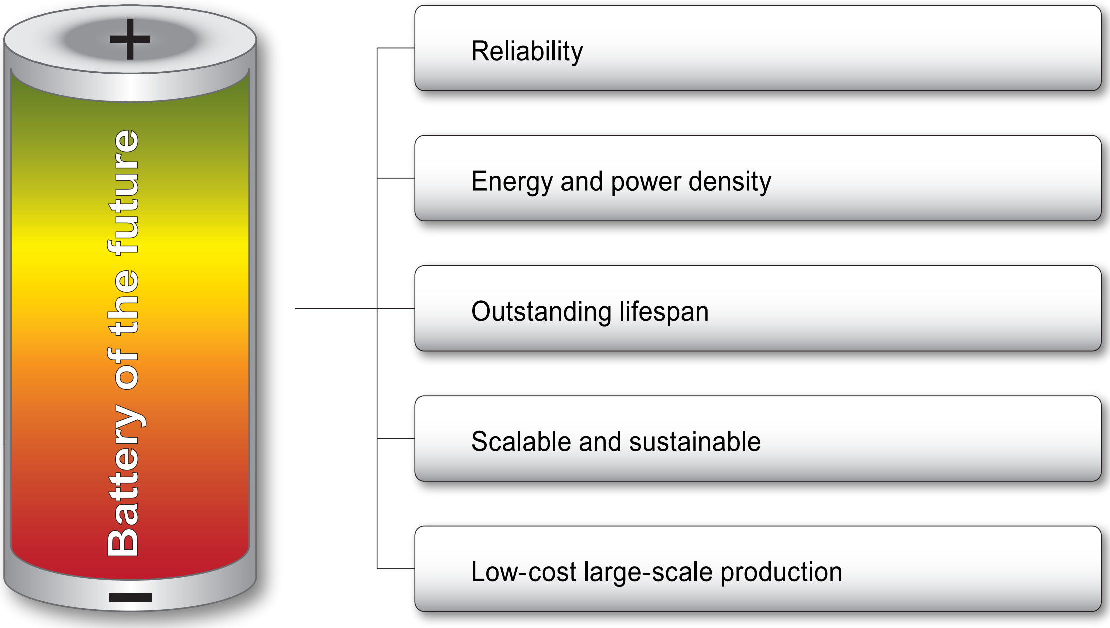


Fig. 6: *Criteria for the battery of the future.*

ted in 2001 (Fig. 7). With the development of new systems like alternative metal anodes such as sodium-ion batteries, and gaseous or liquid cathode systems, at least some existing LIB types may be replaced in specific applications. Reasons for developing new materials have been to improve safety, through the use of thermally more stable materials, and to lower costs by replacing, for instance, cobalt with less expensive materials such as manganese.

Another challenge is to increase the energy density of LIBs. Research in this area is carried out mainly on lithium-sulphur and lithium-air batteries. The first lithium-sulphur battery was proposed in the 1960s by Herbert & Ulam (1962) and patented, but a major breakthrough in development was achieved only in 2009 by Nazar and colleagues (Nazar et al. 2009). The lithium-air battery was proposed in 1996 by Abraham and Jiang (Imanishi & Yamamoto 2019). This battery type captured worldwide attention as a potential energy store for electric vehicles in 2009.

Recently, solid-state batteries have attracted much interest as energy storage systems for electric vehicles. The use of lithium metal as the anode material could increase energy density by 20 % compared to carbon, which is currently used (Ulvestad 2018). Based on electric vehicle manufacturers' technology roadmaps and technological advances, Yole Développement believes that the commercial launch of solid-state batteries will start in 2025. The mass production of vehicles with solid-state batteries, however, is expected no earlier than 2030 (Yole Développement 2020).

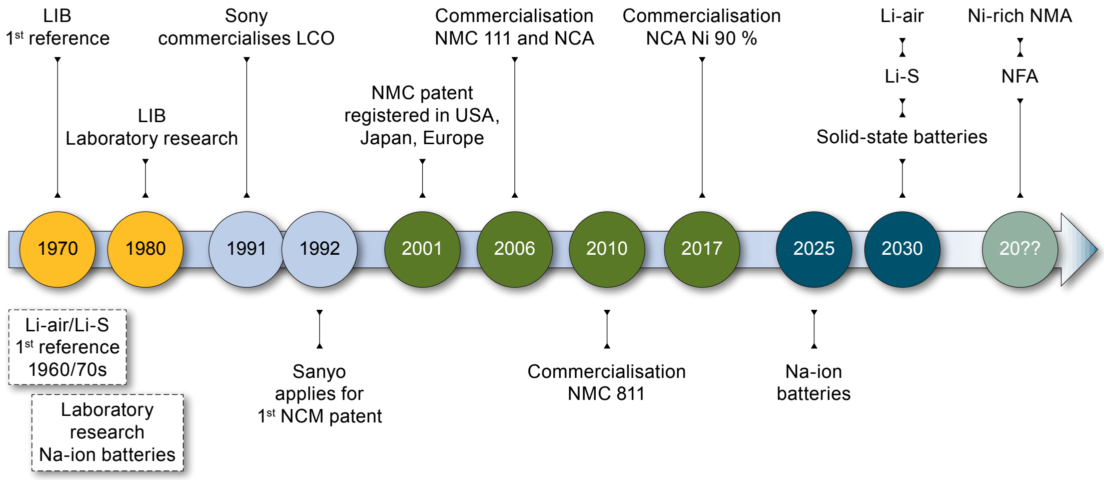


Fig. 7: *Innovation analysis on lithium-ion battery types and alternatives. High-Ni NMA – nickel-manganese-aluminium type, NFA – nickel-iron-aluminium type.*

CONCLUSION

At present, the industrial recycling of LIBs uses two processes: 1) pyrometallurgy, with downstream hydrometallurgical treatment for recovery of the refined metals or salts; and 2) hydrometallurgy, requiring upstream mechanical shredding of the cells or modules. Each process has its own specific advantages and disadvantages. Because of the different physical and chemical properties of the raw materials contained in LIBs, maximum output is not yet possible, which is why the focus is still on valuable metals such as nickel and cobalt. Higher value shares of lithium in batteries could also help intensify recovery efforts.

Thanks to intensive research in the field of LIB recycling, many research projects in Europe have progressed to the pilot plant stage in the past ten to 15 years. Commercially operational plants have annual capacities of several thousand tons. As the market penetration of electric vehicles progresses, the number of LIBs available for recycling will significantly and steadily increase. Recycling capacities in Europe can therefore be expected to expand considerably to meet the growing demand. The legal framework and the requirement to use recycled content resulting from the amendment of the Batteries Directive will have an impact on the recycling sector in the EU.

Recycling facilities in Europe could have annual capacities of 380,000 t and more by 2030. Although the main process routes have already been defined, some issues still need to be resolved regarding the upgrade of existing recycling plants and the next generation of plants. In the future, combined processes and the breakthrough of additional innovative LIB types and alternative competitive battery types will play a major role. The development of LIBs and LIB recycling is highly dynamic, and more advances in many areas are therefore expected in the next years.

One major development focus in this context has to be the “design for recycling” of future LIBs, which takes into account the suitability of product design for recycling and the recyclability of all types of battery materials. Another key challenge remains the widespread registration of end-of-life LIBs and their channelling into high-quality recycling processes. The “battery passport” required in the new Batteries Directive could play a key role in this respect.

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