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Conference paper

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From Volta's pile to lithium ion battery: 200 years of energy

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Abstract: The aim of this paper is to provide a historical and scientific overview of the battery world, from the disrupting discovery of Alessandro Volta to the latest advances in lithium ion batteries. For sake of clarity, it has been divided into three parts: the past, the present and the future. The first part illustrates the historical path which led to the lithium ion batteries. The second part reports the main challenges to the Li ion batteries that have been and still must be faced for increasing their performance and their sustainability. In the last part, considerations about the future of lithium ion batteries are discussed, with a special focus on sustainability.

Keywords: Alessandro Volta's heritage; battery timeline; Li-ion batteries; sustainability.

The past

The first changes in human energy consumption arose with the ramping up of the evolution of steam engines in the 17th and 18th centuries and the breakthrough of Thomas Newcomen and James Watt in the mid-1700s that gave rise to the modern steam engine. In less than one century, coal was used for heating, powering steam engines, and generating electricity. With the low-cost automobile and the spread of electricity, our society's energy needs proliferated, doubling every 10 years. After World War II, the employment of unsustainable energy sources rose out of control until the Great Oil Crisis in 1973 when the Arab oil-producing nations instituted an oil embargo [1, 2]. This event highlighted the world's continued dependency on fossil fuels for the first time.

In addition, the uncontrolled exploitation of fossil fuels dramatically increased greenhouses gas emission and climatic change issues. The consumption of fossil fuels, in fact, exposes the atmosphere to huge quantities of carbon dioxide and other greenhouse gasses, leading to global warming, *i.e.* the increase in global average temperature. Human activities' consequences have been induced a global warming that reached approximately 1 °C above pre-industrial levels in 2017 and is increasing by 0.2 °C per decade [3, 4]. For this reason, the Paris agreement (entered into force on November 4th, 2016) aimed to hold the global average temperature increase less than 2 °C above pre-industrial levels and pursue efforts to limit it to 1.5 °C [5, 6]. Nevertheless, a global energy transition away from fossil fuels is urgently needed. However, renewable power sources cannot be fully exploited due to their intermittent nature if paired with large-scale energy storage. In this scenario, energy storage systems (ESSs) are essential to overcome the mismatch between the generation and the demand for electric energy [6] (Fig. 1).

In 2018, primary energy consumption in Europe was 635 million tonnes of oil equivalent (Mtoe) spread over a range of different energy sources, with renewable energy sources contributing more than one-third (34.2 %) of the

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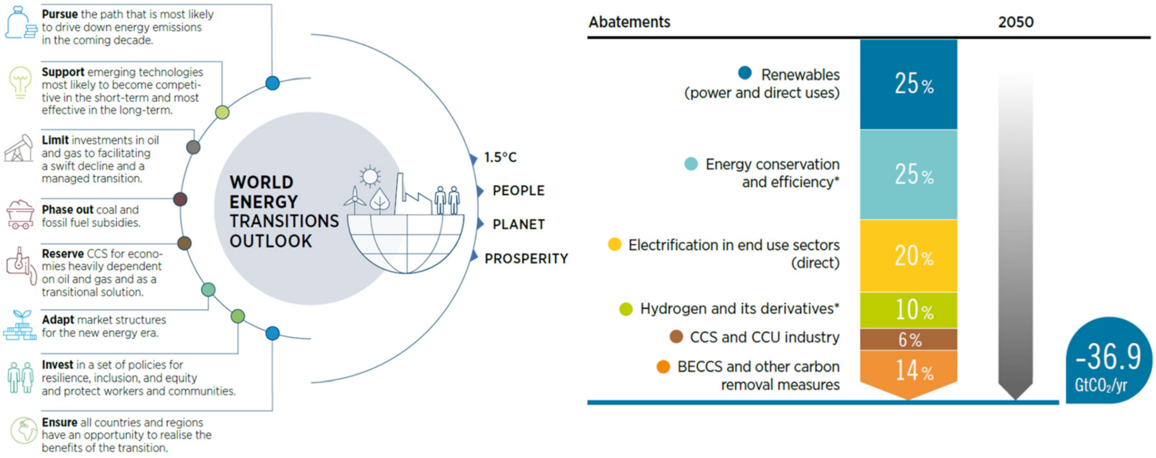


Fig. 1: Guiding framework of World Energy Transitions Outlook (WETO) theory of change and carbon emissions abatements under the 1.5 °C scenario (%) [6]. CCS, carbon capture and storage; CCU, carbon capture and utilization; BECCS, bio-energy with carbon capture and storage.

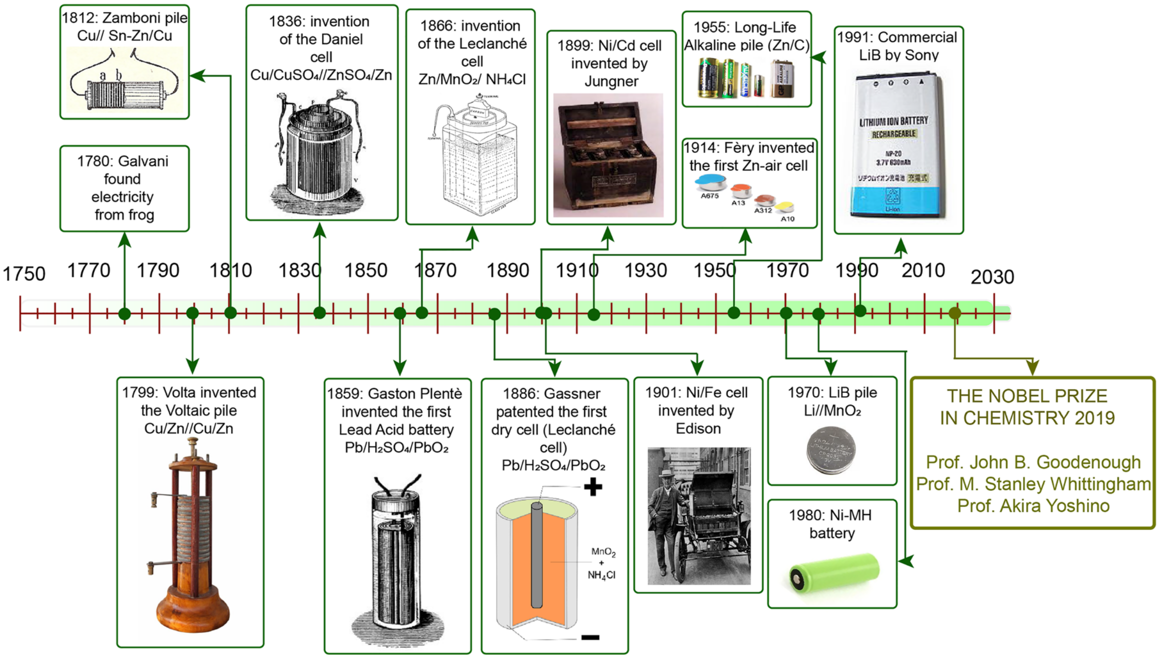


Fig. 2: Timeline of battery development from 1750 to 2020.

EU's total production. This energy fraction has increased significantly in the last few decades in combination with increased energy demand [2, 6].

In this context, lithium ion batteries (LiBs) have been one of the most used electrochemical ESSs since 1991. However, there has been a long way to go for their development (Figs. 2 and 3). The starting point can be found in Italy in the 1800s, with the studies of Luigi Galvani (University of Bologna) on animal electricity [7] and Alessandro Volta (University of Pavia) who first revealed his 'electric battery' to Napoleon's court in France [8, 9]. Volta's work had an enormous impact on the progress of electrochemical science, catalysing the rapid evolution of battery history with the cumulative discovery of many important electrochemical systems. In less than two centuries, John Daniel solved the problem of hydrogen evolution by dividing the electrodes in two separate compartments,

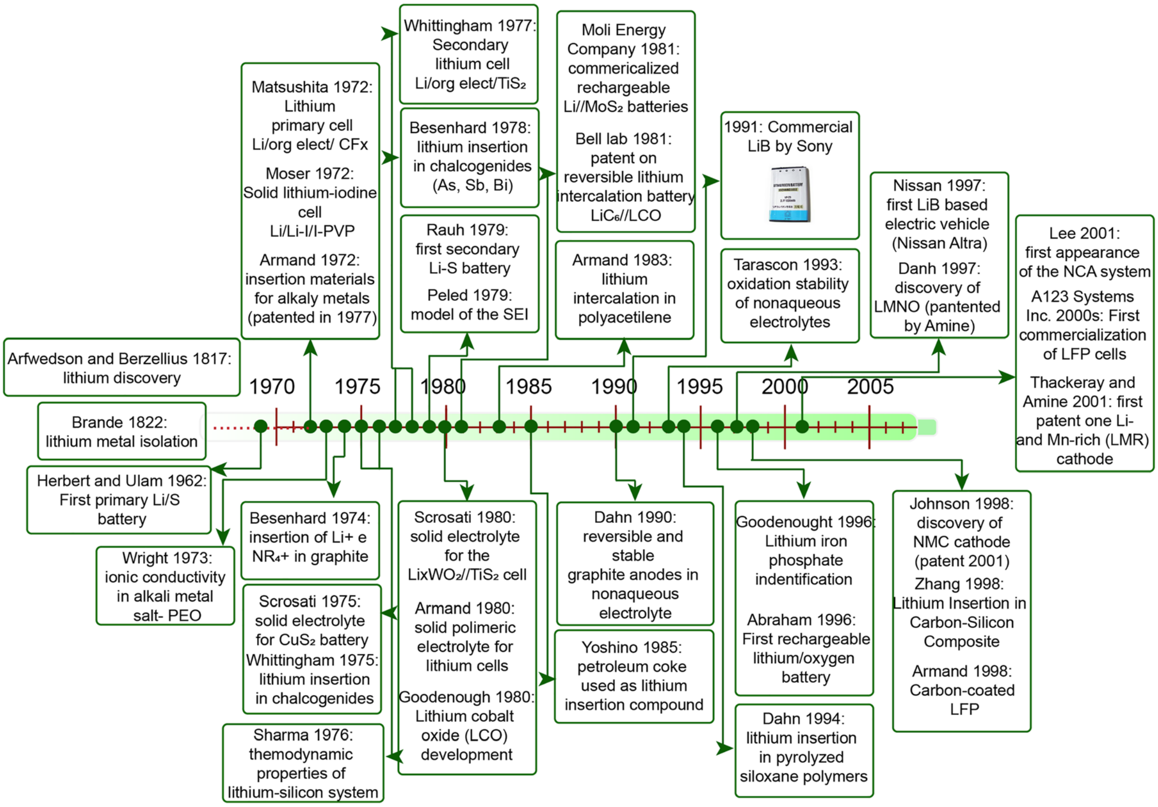


Fig. 3: Timeline of lithium-ion battery and related technologies development from 1970 to 2005. PVP, polyvinylpyrrolidone; PEO, poly(ethylene oxide); NMC, $LiNi_xMn_yCo_{1-x-y}O_2$; LCO, $LiCoO_2$; LFP, $LiFePO_4$; SEI, solid electrolyte interphase; LNMO, $LiNi_{0.5}Mn_{1.5}O_4$; LMR, lithium-and-manganese-rich oxides; NCA, $Li(Ni_{0.8}Co_{0.15}Al_{0.05})O_2$.

and Giuseppe Zamboni (1812) introduced the first example of a dry cell constituted of silver, zinc, and paper discs. Further developments were presented by Georges-Lionel Leclanché (1866) and Gaston Planté (1859), who set the bases for the modern dry cell and the lead-acid battery, respectively. Although with some important variations from the original design, it is remarkable to note that these systems are still in use today with different applications. Just think of lead-acid and the ignition of car engines, or the Leclanché cells that paved the way to alkaline batteries [10].

The first effective electric cars started to circulate in 1896, with a sale peak in 1912, even if they were quite expensive and their driving range was not very high. After only a decade, several factors started to mark the end of the first age of electric vehicles and the rise of gasoline cars to the top of the market, especially in the US: the development of streets and highways that favour connections among faraway cities, the discovery of oil reserves and the decrease of gasoline car prices after Ford introduced the assembly lines, making these gasoline vehicles more reliable and comfortable [11].

The first waves of interest in lithium-based batteries emerged in the early 1960s, fuelled primarily by governmental and military interests in high energy density storage chemistries. As a result, most literature from this era exists in the form of technical reports submitted to governments. In the sequent years, many research groups were working to build lithium-based systems [12]. The breakthrough came in 70' with the contribution of J. O. Besenhard and S. Whittingam with the development of so-called 'insertion' or 'intercalation' electrodes [13, 14]. These are typically based on compounds that can reversibly host and release lithium ions inside and outside their open structure.

The use of lithium, the most electropositive metal on the periodic table and the lightest alkali metal, led to the development of a cell with a practical specific energy around 40–45 Wh kg^{-1} [15–17]. Several companies tried to commercialize such high-energy systems despite the Li metal anode's possible risks. A notable Canadian company (Moli Energy) introduced the first rechargeable Li metal battery with a MoS_2 cathode with an emphasis on AA cells, creating a first-generation technology in 1985. Indeed, by 1988, they began selling an AA cell with more than 2 Wh under the name Molicel, but this market operation failed because of several fire incidents [16]. These were due to the lithium-metal electrodes' instability when in contact with liquid electrolytes. The successful cathode came from Goodenough and co-workers, with studies started in 1980. They correctly predicted that transition metal oxides, rather than sulfides, would be more stable at high oxidation states, exhibiting higher potentials and, thus, higher cell voltage and energy density. Several layered structure materials based on $LiMO_2$ (M = V, Cr, Co, Ni) were synthesized and investigated, but $LiCoO_2$ (LCO) was the most convincing [18].

Most of the problems of the lithium cells were associated with the presence of lithium metal. The very high reactivity with the electrolyte and irregularities on the surface can lead to non-uniform deposition of lithium during charging, forming dendrites that eventually grow, short-circuiting the cell. At that time, the conditions were not ripe for the employment of lithium metal [12]. Solid electrolytes have been studied to solve the problematic coexistence of lithium metal and liquid electrolytes. After P. V. Wright's discovered ionic conductivity in alkali metal salt complexes of poly(ethylene oxide) (PEO) in 1973 [19], polymer electrolytes were highly studied and proposed for batteries since 1980. The number of contributions to this field has grown significantly, reflecting progress in understanding molecular and supramolecular architecture, which is necessary for fast ion transport in polymers [20, 21].

In order to guarantee cycle life and safety, Dahn, Armand, and Yoshino groups investigated less aggressive anodic materials replacing the lithium metal, studying lithium intercalation in graphite, polyacetylene, and petroleum coke, respectively [22–24]. Therefore, the "rocking chair" strategy is the most promising, in term of safety, in which a lithium-ion source substitutes lithium metal. Lithium ions are "rocked" between lithium-carbon and lithium-metal oxide intercalation compounds, which serve as the electrode couple during such battery cells' charge and discharge processes [25–27].

The combination of these efforts led to the development of the first lithium-ion batteries (LiBs) by Sony in 1991, employing graphite and LCO electrodes. For what concerns the positive electrode, the relatively low practical specific charge (140 mAh g^{-1}), as well as drawbacks in terms of stability and toxicity, have encouraged the development of alternative active materials. In addition, cobalt prices have increased rapidly because of the criticality of cobalt reserves. Since the beginning, $LiNiO_2$ has been a target compound because it is structurally identical to LCO, *i.e.* a layered metal oxide, is less expensive, and has a higher energy density than LCO [28]. However, despite the extensive research, $LiNiO_2$ is not commercially available in its pure form due to preparation and security issues. $LiNiO_2$ has inadequate thermal stability in the charged state, which can result in thermal runaway of the cell. A lot of work is being done to partially replace nickel with some other elements in order to solve the safety issue. In $LiNiO_2$, aluminium is specifically used as a co-dopant with cobalt. The most well-known derivative is $Li(Ni_{0.8}Co_{0.15}Al_{0.05})O_2$ (NCA), which is applied commercially on a small scale. The fast reaction with moisture, producing Li_2CO_3 and LiOH on the surface of these high Ni content materials, like NCA, represents the major concern [26]. Lithium iron phosphate ($LiFePO_4$) was proposed as a positive electrode material by Goodenough et al. in 1996 [29], but its poor electronic conductivity prevented its widespread use. Armand et al. [30] suggestion to coat the $LiFePO_4$ particles with carbon represented a significant step forward for the material's application, and as a result. Lithium nickel manganese cobalt oxide ($LiNi_xMn_yCo_{1-x-y}O_2$ – NMC) materials were patented by Thackeray et al. in 2000 [31], and due to their high specific capacity (compared to LCO) and high voltage (compared to $LiFePO_4$), they are of interest for the developing EV market. It must be considered that NMC still contains cobalt, but to a much lower extent with respect to LCO.

It was clear from the beginning that a compromise between performance, cost and safety should be found. $LiMn_2O_4$, a naturally abundant, affordable, and environmentally friendly cathode material, was considered an interesting cathode material despite the specific charge smaller than LCO and capacity fading issues and was commercialized in 1996 [32]. To improve the electrode's performance, the manganese was partially substituted with other metal ions. $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO), first reported in 1997, displays the best overall electrochemical performance of these derivates [33]. The voltage was thus increased from 4.1 V to 4.7 V in comparison to $LiMn_2O_4$ spinel, and the specific capacity was 140 mAh g^{-1}.

To improve the specific energy and cycle life of LIBs further, in-depth studies on electrolytes and electrolyte/electrode interphases were performed. The ground-breaking research by Dahn et al. [34] demonstrated the co-intercalation of propylene carbonate (PC) into graphite and discovered that the addition of ethylene carbonate (EC) as a co-solvent significantly reduced PC decomposition. This result is attributed to the preferred decomposition of EC, which prevents the cointercalation of PC and forms a favourable solid-electrolyte interphase (SEI) layer on graphite. An electrolyte based on $LiPF_6$ in EC/dimethyl carbonate (DMC) was proposed by Guyomard and Tarascon [35] in 1993, and it later became the standardized electrolyte formulation for today's LIBs. LCO – based LIBs has reached 250 Wh kg^{-1} and 600 Wh L^{-1}, which is nearly three times higher than the initial LIB produced by Sony thanks to the $LiPF_6$ – EC/DMC electrolyte and some additives [36].

The present

LiBs are widespread and present in our daily life. As an example, the sale of smartphones has increased more than tenfold in the 10 years from 2007 to 2017, from 120 million units to 1500 million units [37], and 80 % of the LiB waste (around 190 kton) available for recycling in 2019 came from portable electronics [38].

When we talk of lead-acid or Ni-metal hydride batteries, we know exactly what chemistry is inside. On the contrary, the term 'lithium ion batteries' is very general and includes several chemistries, mainly working with the same 'rocking-chair' concept of Li ion transport between the electrodes. The driving force for the development has been, and still is, the need for high specific and volumetric energy and power with an eye to the safety. More recently, the sustainability and the idea of a battery value chain have moved the research, specifically at European level, toward Co-free (or Co-less) batteries, with electrodes processed in aqueous or solvent-free environment. Figure 4a shows the huge advances, over the last three decades, in terms of energy increase and cost decrease of LIBs [39, 40]. These advances have been achieved because of the important research developments, supported by the continuous and increasing demand that, specifically, allowed the decrease of the production cost. In terms of energy, the discovery of new materials, the amelioration of the old ones, the improvement in the assembly and the reduction in weight and volume of inactive materials have brought energy and power increase. From the materials point of view, the cathode and anode active materials are the leading actors, and great differences in the cell energy and power are mainly due to the kind of selected electrode materials that can span in a capacity range of 130–200 mAh g^{-1} for the cathode to 175–4000 mAh g^{-1} for the anode, with an operating voltage from 3.4 to 4.8 V for the former and 1.75 to 0.05 V for the latter. Figure 4b shows the specific capacity and the potential of the different cathode and anode materials. Of course, only few electrolytes based on ionic liquid are stable in such a wide potential range. The formation of a solid electrolyte interphase (SEI) on the low-potential anodes, and of a cathode electrolyte interphase (CEI) on the high-potential cathodes is what makes LIBs able to work far from the thermodynamic stability of the electrolyte [41–43].

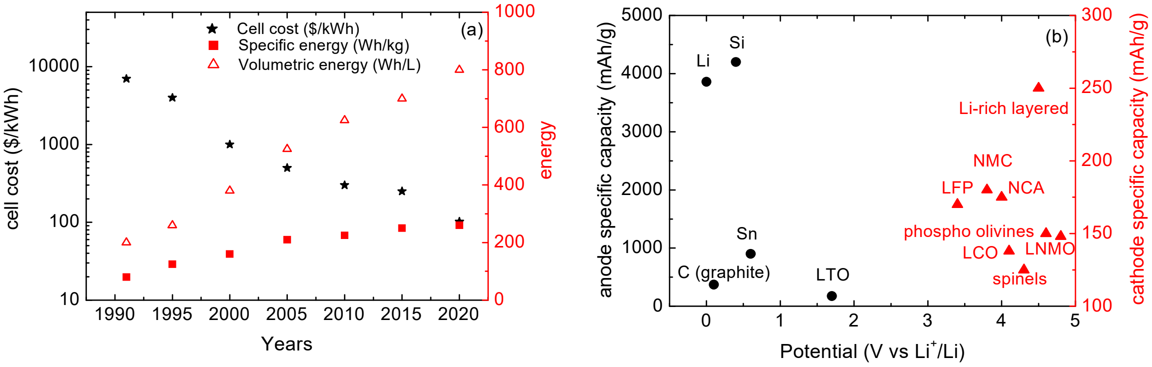


Fig. 4: Li-ion batteries cost and performance: (a) cost and energy performance variation over time; (b) anode and cathode specific capacity and their potential. NMC, $LiNi_xMn_yCo_{1-x-y}O_2$; LFP, $LiFePO_4$; NCA, $Li(Ni_{0.8}Co_{0.15}Al_{0.05})O_2$; LCO, $LiCoO_2$; LNMO, $LiNi_{0.5}Mn_{1.5}O_4$; LTO, $Li_4Ti_5O_{12}$.

At present, one of the most used cathode materials is NMC, *i.e.*, a layered material containing Ni, Mn, and Co. With the aim to lower cobalt content, several stoichiometries have been used, with increased capacity and potential by increasing Ni content. However, the thermal stability is inversely proportional to Ni content. Another material widely studied is the high-voltage spinel LNMO, *i.e.*, $LiNi_{0.5}Mn_{1.5}O_4$, a very promising Co-free cathode material.

The battery technology development depends not only on material improvements, but also on achieving safer batteries and more sustainable processes. As an example, solid electrolytes are less flammable than liquid electrolytes. Advanced solid electrolytes or some binders possess self-healing capability by means of hydrogen bond formation when a damage occurs. The possibility of repairing damage guarantees the cell's safety and a longer cycle life. Si-based anodes that suffer of high volumetric variation upon the electrochemical processes of lithiation/delithiation, can fulfil the need of boosting volumetric energy and long cycle life by taking advantage of the self-healing properties of the binder or of the SEI [44].

Even the so-called inactive materials, *i.e.*, binder, conducting additives, current collectors, and separators, are crucial for the good and effective battery operation. The synergy between binder, conducting additive, and current collector is responsible for a good electric contact, resulting in low resistance and high cell power. A tailored separator, too, may improve the contact with the electrodes, thus increasing the interfacial contact and improving the ionic transport of Li^+ from the electrolyte to the electrodes.

Sustainable processes are desirable, and, among others, electrode preparation can be improved in terms of sustainability. The organic solvent usually used for electrode casting on current collectors is *N*-methyl pyrrolidone, a toxic and flammable organic solvent with a high boiling point. It can be substituted by water with the goal of use a less toxic solvent and less energy to evaporate and recover it. Water can be evaporated in air, without spending energy and money on a condenser system. In addition, if the energy used does not come from renewable sources, a smaller CO_2 emission is involved in a shorter and less energy consuming casting system [45].

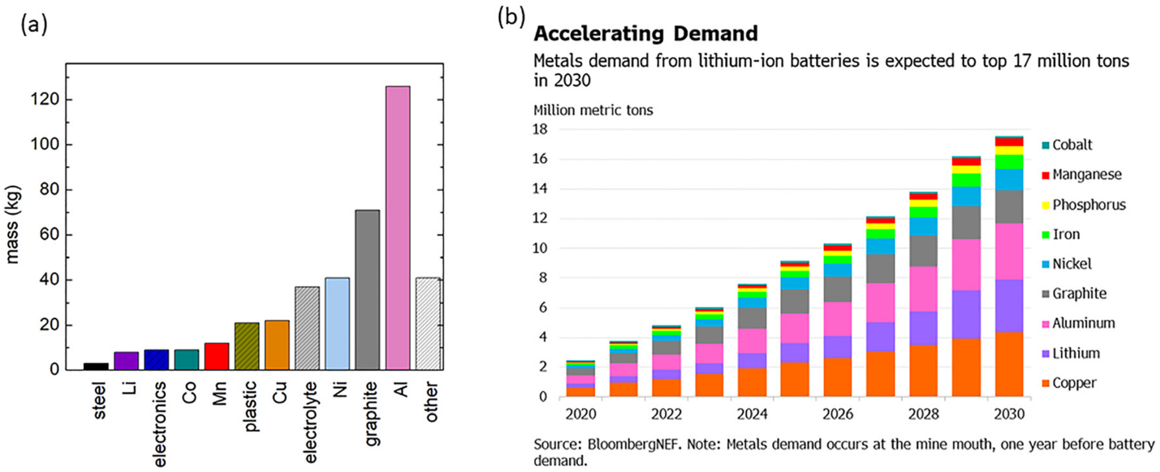
The future

LIBs will also cover the energy need in the future for those applications requiring both energy and power requirements. However, most of the study on other systems started several decades ago began to give significant results. Some examples are the renewed interest in Li metal batteries, where the graphite anode is substituted with a Li metal anode, still maintaining the cathode insertion material, using a solid electrolyte. Advances in polymeric and inorganic solid electrolytes are the driving force behind safe and high-performance batteries. The solid-state technology will have an impact on the electrification process that will be more relevant in some sectors, like transport [6].

The increase of volumetric energy can also be attained by using Si–C based anodes with percentages of Si near 50 %, with suitable morphology and nanostructure, and with the use of self-healing binders [17, 46].

In the sector of stationary storage, sodium-ion batteries (NIBs) may be used, together with LIBs or replacing LIBs, as a more sustainable alternative energy electrochemical storage system [47]. Sodium is cheap, abundant (1000 times more than Li) and widespread worldwide. NIBs history began with that of LIBs [48, 49]. However, the best performance of the latter made NIBs neglected until the growing interest in topics related to Li, like availability, cost and geo-politic issues, renewed the interests toward them. Strong of the research findings on LIBs, the studies on NIBs saw an acceleration in the last few years, and what seemed possible but not so advantageous in terms of performance, is now a real system able to be applied also in the most challenging applications like electric vehicles. The first generation of mass-produced sodium-ion batteries has been installed in a production electric car for the first time in 2023. The model is part of Volkswagen's joint venture with the Chinese JAC (Sehol E10X) and can cover 250 km driving range on a single charge. The world's biggest electric vehicle (EV) battery makers, the Chinese CATL and BYD, are also on the way of the mass production, with BYD expected to start this year. This is only the first generation of NIBs in EVs, and their progress will be much quicker thanks to the industry experience gained in LIBs over the last decades [50].

Another point to be addressed in the future concerns the recycling and reuse of batteries [51]. Materials demand is expecting to tenfold in 10 years and approach 18 million tons in 2030 and the key question is whether there is sufficient supply to meet booming demand [52] (Fig. 5).



Accelerating Demand

Metals demand from lithium-ion batteries is expected to top 17 million tons in 2030

Source: BloombergNEF. Note: Metals demand occurs at the mine mouth, one year before battery demand.

Fig. 5: Materials demand for Li-ion batteries: (a) mass of materials used in a 400-kg EV pack; (b) projection of materials and metals demand for Li-ion batteries [52].

In addition, as demand for EV batteries grows, countries are struggling to build their own supply chains. The supply chain necessarily has to include the recycling/reuse step for limiting any dependence from third countries, even if not being totally self-sufficient. Europe is working in this direction, with the development of regulations to control the end of the life of these batteries with special emphasis on the efficiency levels of recycling and recovery of materials. The objective is not only to limit the extraction of new raw materials, but also to protect the environment by minimizing the negative impact of battery wastes. In Europe there is a proliferation of recycling plants and projects involving industries, research centres and academies, as well as initiatives aiming at recycling. Among the latter, Eucobat [53], the European association of national battery collection systems, and EuRIC [54], which acts as an interface between the industry and the European Union for the exchange of best practices in all matters related to recycling, are worth mentioning.

Perspectives

The electrification process necessary for facing the climate changes involves the efficient use of renewable energy sources, which imply their combination with energy storage systems. Electrochemical energy storage systems have been demonstrated as the most versatile for different purposes, from stationary to portable applications. Among them, LIBs soar over the other chemistry for their high gravimetric and volumetric energy and power. After more than 30 years from their commercialization, they are still at the cutting edge with continuous improvements in terms of performance, cost and safety. However, with an eye to the future, it is necessary to find also other systems that can be used as an alternative to LIBs when the performance requirements are not so stringent, and to pursue an effective policy for recycling and reuse of LIBs to preserve our planet from wastes and preserve raw materials.

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