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On the Current and Future Outlook of Battery Chemistries for Electric Vehicles—Mini Review

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Abstract: As the electrification of the transportation industry is accelerating, the energy storage markets are trying to secure more reliable and environmentally benign materials. Advanced materials are the key performance enablers of batteries as well as a key element determining the cost structure, environmental impact, and recyclability of battery cells. In this review, we analyzed the state-of-the-art cell chemistries and active electrode and electrolyte materials for electric vehicles batteries, which we believe will dominate the battery chemistry landscape in the next decade. We believe that major breakthroughs and innovations in electrode materials such as high-nickel cathodes and silicon and metallic lithium anodes, along with novel liquid electrolyte formulations and solid-state electrolytes, will significantly improve the specific capacity of lithium batteries and reduce their cost, leading to accelerated mass-market penetration of EVs.

Keywords: cathode materials; solid-state electrolytes; Li metal; silicon; EVs

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1. Introduction

Over the past five years, the global demand for electric vehicles (EV) has increased rapidly. In the next decade, we expect an exponential rise in EV sales [1]. Because of elevated demand, auto manufactures have been pressured to adjust their line-ups of models and associated supply chains (especially as they pertain to EV batteries and their subcomponents) to guarantee that they are the best set to capitalize on this heightened demand outlook [2]. We note that the various strategies used by automakers and governments to rectify these issues have led to the politicization and localization of EV battery supply chains [1]. The rise in EV demand has also spurred competition among EV and EV battery manufacturers to secure access to the critical raw materials (CRMs) used in LiBs, most notably lithium, cobalt, and nickel. In 2021, Fitch Solutions analysis forecasted that global EV sales would reach over 26.7 million units in 2030, representing year-over-year growth of 379.0% from 2021. More importantly, in this series, the analysis forecasted that the annual global demand for EV batteries, in terms of total global capacity, would reach 1925 GWh by 2030, an increase of 688% from 244.7 GWh in 2021 [3].

Recently, CIC energiGUNE reviewed major announcements made around the battery industry in the North American region, where the United States of America has become a predominant player in the global future of gigafactories (Figure 1) [4]. However, unlike a year ago, Canada had become active in developing its industry. An example of this is the joined project announced by Stromvolt last October in building the first large-scale EV battery cell manufacturing plant (10 GWh) before 2030 in Quebec. Last March, another Canadian company (Lion Electric) planned to develop a production facility of up to 5 GWh by 2023 in Quebec. Moreover, a British company (Britshvolt, city, country) announced launching a 60 GWh facility in Quebec, which would make it one of the largest manufacturing plants on the continent.



Figure 1. North American battery initiatives.

One of the latest announcements was a new battery factory in Ontario made by Stellantis and LG Energy Solution, with production to initiate in early 2024. Canada's efforts are gradually enabling North America to close the gap with Asia and Europe, the two main poles in the race to dominate this sector in the future. Based on a BloombergNEF (BNEF) analysis, the average cost of a Li-ion EV battery pack dropped by 89% from $1200/ kWh in 2010 to $132/kWh in 2021 [5]. This represented a 6% fall from $140/kWh in 2020. EV battery packs are made up of numerous interconnected modules that consist of *tens to hundreds* of rechargeable Li-ion cells. Collectively, these cells were assessed at ~$101/kWh, or approximately 77% of a battery pack's overall cost (Figure 2b). BNEF's 2021 Battery Price Survey predicted that the overall pack price would fall below $100/kWh in 2024.

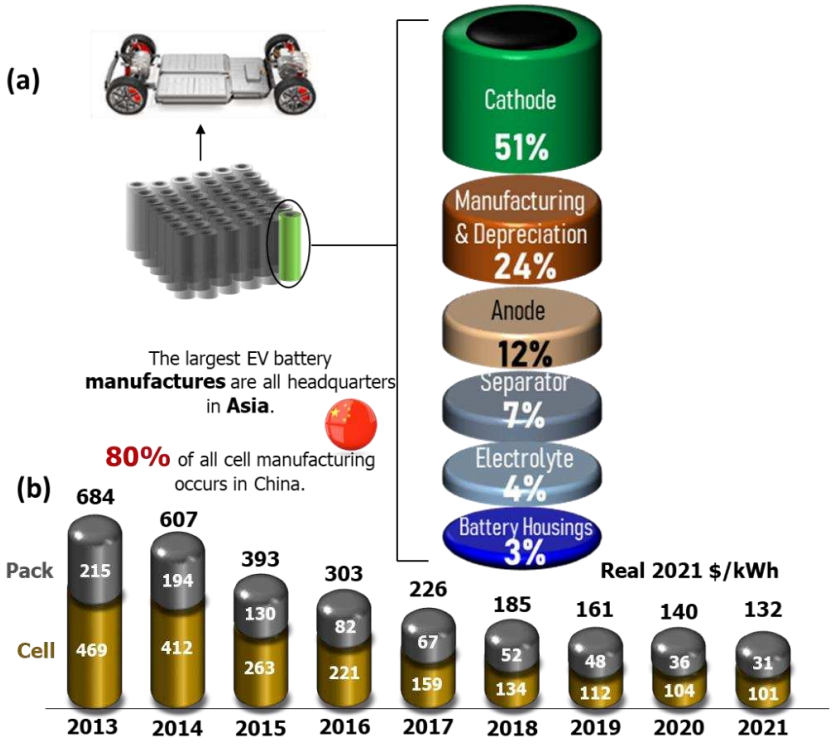


Figure 2. (**a**) Schematic breaking down the cost of an EV battery cell [5]. (**b**) Volume-weighted average pack and cell price split. Adapted with permission from Ref. [6]. Copyright 2022 Copyright BloombergNEF.

Around this price, EVs will be comparable with internal combustion engine vehicles (ICEs) in some markets, as the current rates will fluctuate depending on various strategies such as geography and manufacturers. However, in the near future, if the raw material prices rise, the average pack cost might increase to $135/kWh in 2022 in nominal value. In the absence of other enhancements that can alleviate this effect, this might mean that the above date by which rates would drop under $100/kWh might be delayed by at least two years. This would affect EV manufacturers' margins or affordability and might damage the economics of energy storage developments [6]. In Figure 3a, Mauler el. calculated and evaluated raw material markets (lithium, manganese, cobalt, nickel, and graphite) in regard to future cell development [7]. Their roadmap study integrated both cost restrictions and material price expectations throughout 2030. Based on their analysis, the highest single raw material that would impact the cell cost in 2030 was nickel (+4.2–19.3 $ kW h^{-1}), followed by graphite, cobalt, lithium, and manganese (+1.4–8.0 $ kW h^{-1}, +1.0–3.7 $ kW h^{-1}, +0.3–3.2 $ kW h^{-1}, and <0.1 $ kW h^{-1}, respectively). Because the nickel intensity of Li-ion batteries is increasing with the use of high-nickel cathodes (NMC 811 and NCA), a nickel spike would have severe consequences on the LIB technology roadmap. Therefore, securing supply is a prudent strategy.

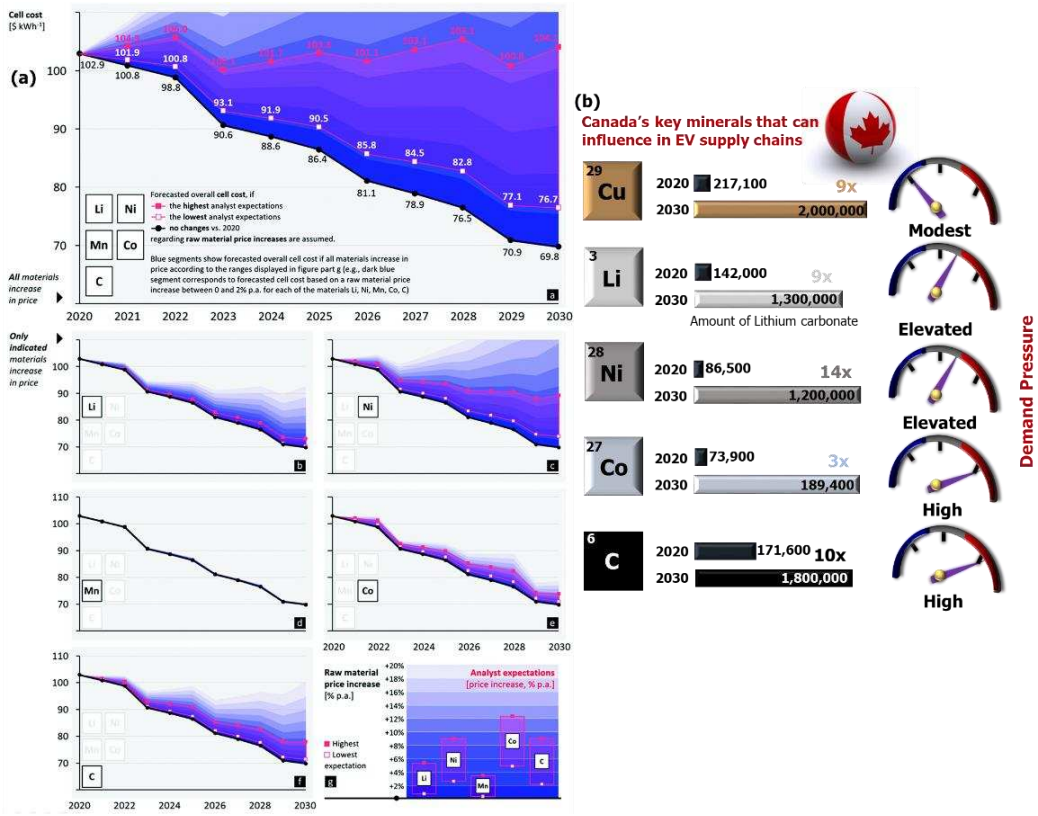


Figure 3. (**a**) Impact of increasing raw material prices on cell cost throughout 2030 for 5 materials alongside analyst expectations for raw material price developments, reprinted with permission from [7]. Copyright 2022, The Royal Society of Chemistry. (**b**) Summary of the key minerals that could help Canada gain a major influence in the EV supply chain, along with demand pressure on the right side [8].

Canada aims for zero-emission vehicles to account for 30% of annual passenger vehicle sales by 2030 (announced by NRCan). By 2030, the global battery and plug-in hybrid electric vehicle fleet should exceed 200 million, according to estimates (done by IEA, Bloomberg). This transformation will require developing significant amounts of new materials. Figure 3b illustrates the key minerals that could help Canada a gain major influence in EV supply chains. Canada has a long history of generating 31 minerals, including the five key minerals

for EVs mentioned in Figure 3b, and has the possibility to produce more (based on NRCan reports). These critical minerals are (i) required for Canada's economic security, (ii) essential for Canada's transition to a sustainable, carbon-neutral economy, and (iii) a sustainable source of critical minerals for Canada's partners, positioning Canada as one of the leading mining nations in the near future. Other relevant domestic materials with increased demand include iron (steel), aluminum, and rare-earth elements. The criticality of minerals and demand for them from low-carbon technologies, especially EVs and energy storage, will put significant pressure on supply chains [7]. Lithium, cobalt, and graphite carry additional risks, since future demand for them will be disproportionately greater than their current production capacity. To improve the specific capacity, reduce pack costs, and promote safety aspects, LIB technology keeps advancing. Advanced materials (cathode/electrolyte/anode) are the key performance enablers of batteries as well as a key element determining the cost structure, environmental impact, and recyclability of battery cells. Our objective was to focus on a limited set of battery chemistries that will dominate the battery chemistry landscape well into the 2030s and beyond. The following sections provide an overview of advanced battery technologies. This effort was completed on the basis of various publications and industrial reports (Canada Research Interfaces, Arthur D. Little, Sila, Lux Research, CIC energiGUNE, BloombergNEF, and Fitch Solutions), not unlike other review papers.

2. Automotive Batteries: State-of-the-Art Li-Ion Cathodes

LIBs consist of various types of cathode materials, with each combination having distinct pros and cons in terms of costs, safety, performance, and other parameters. The structure and voltage representative profile for each cathode material is illustrated in Figure 4. Currently, lithium cobalt oxide (LCO), known as a mature cathode chemistry, is the most predominant battery technology for consumer electronics. However, because of its structural instability in terms of overdelithiation, this chemistry is not suitable for EV applications [9]. Other cathode chemistries such as lithium iron phosphate (LFP), lithium nickel manganese cobalt oxide (NMC), lithium nickel cobalt aluminum oxide (NCA), and spinal lithium manganese oxide (LMO) have become dominant battery materials for automotive applications compared with LCO because of their abundant resources, stable crystal structures, and low price.

From the technical point of view, competing Li-ion battery technology can be compared across five characteristics, costs, safety, lifespans, power, and energy densities, as illustrated in Figure 4b. Up to the present, various automakers, such as the Mercedes-Benz Group, Volkswagen, Tesla, BMW, Nissan, Chevrolet, and BYD, have adopted NMC, LFP, LMO, and NCA battery technologies in order to reduce greenhouse gas (GHG) emissions and mitigate oil consumption. The yields of the major four cathode materials have increased significantly over the years, reaching 180 Ktons in 2016 and expected to rise by 220% in 2025 (to 400 Ktons) [11]. Based on the global EV outlook (2017), the whole market share accounted for 83% of LCO, LFP, and NMC, which were, except for LCO, utilized mainly for EV applications. By 2025, the NMC battery technology is estimated to grow its market share by 15% because of its higher energy density than LFP, while LMO and NCA will retain stable occupancies of the total market [12]. Tesla uses NCA technology, whereas most other carmakers utilize NMC, LMO, LFP, or blended (LMO and NMC) technology, as shown in Table 1. In the foreseeable future, these mature battery technologies will still govern the automobile market before novel battery chemistries dominate EV applications.

To date, the highest volumetric and gravimetric energy densities at the cell level (18650-type cells) are 670 WhL^{-1} and 250 $Wh.kg^{-1}$, respectively [13]. The EV driving range has continuously grown to ~300 km with high current energy content (>100 kWh).

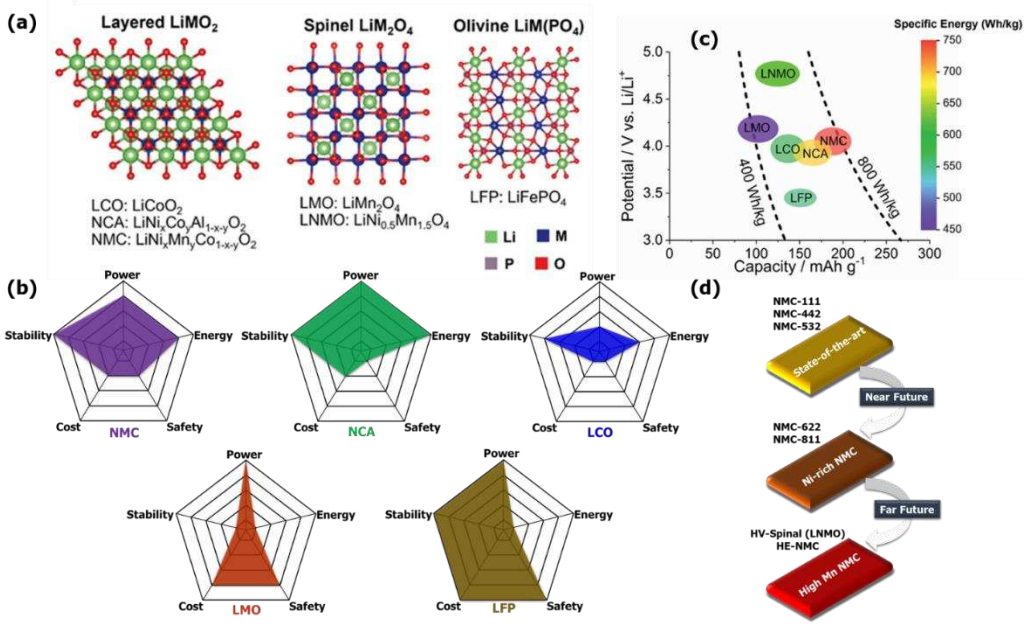


Figure 4. (**a**) Crystal structures of mainstream Li−ion cathodes such as layered (3Rm), spinel (Fd−3m), and olivine (Pnma) systems (viewed along the a−axis). (**b**) Radar summary of the features for the most common cathodes. (**c**) Operating potential, specific capacity, and specific energy of several such cathodes. (**d**) Schematic of the state-of-the-art NMC cathode materials, near-future NMC materials, and far-future NMC series materials. (**a**–**c**) reprinted with permission from [10]. Copyright 2022, John Wiley and Sons, Inc.

Table 1. Various lithium-ion battery chemistries for EVs. LTO: $Li_4Ti_5O_{12}$, C: graphite; Si: silicon; NMC: $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$; NCA: $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$; LFP: $LiFePO_4$; and LMO: $LiMn_2O_4$. (Data from 2017–2021).

Cell Chemistry	Type	Producer	Voltage (V)	Capacity (Ah)	Energy Density ($Wh.L^{-1}$)	Specific Energy ($Wh.kg^{-1}$)	Energy (kWh)	Driving Range (km)	EV Model
C/**NMC721**	Pouch	LG Chem	3.65	64.6	648	263	85	392	Audi e-tron GT
C/**NMC722**	Pouch	LG Chem	3.65	129.2	-	148	93	472	Audi e-tron GT
C/**NMC721**	Pouch	LG Chem	3.65	78	-	156	77	305	Audi Q4 e-tron-SUV
C/**NCM 622**	Prismatic	Samsung SDI	3.68	94	-	148	38	178	BMW i3
C/**NMC622**	Pouch	LG Chem	3.75	55	228	151	65	417	Chevrolet Bolt
LTO/**NMC**	Prismatic	Toshiba	2.30	20	200	89	20	130	Honda fit EV
C/**NMC 622**	Pouch	LG Chem (Umicore)	-	60	164	142	64	484	Hyundai KONA Electric
Lithium-ion polymer C/**NMC**	Pouch	LG Chem	-	-	-	-	77	488	Hyundai ioniq 5-LR AWD
Lithium ion polymer-C/**NMC811+**	Pouch	SK Innovation	3.56	180	-	250	64	370	Kia Niro
Lithium-ion polymer battery-C/**NMC622**	Pouch	SK Innovation	3.56	180	-	250	64	391	Kia Soul EV
C/**NMC811**	Pouch	LG Chem	-	11.2	230	163	78	303	Kia EV6-LR AWD
C/**NMC622**	Laminated	LG Chem	-	56.3	-	151	62	364	Nissan LEAF E Plus

Table 1. *Cont.*

Cell Chemistry	Type	Producer	Voltage (V)	Capacity (Ah)	Energy Density ($Wh.L^{-1}$)	Specific Energy ($Wh.kg^{-1}$)	Energy (kWh)	Driving Range (km)	EV Model
C/**NMC532**	Pouch	AESC	3.65	56.3	205	130	39	240	Nissan LEAF
C/**NMC721**	Pouch	LG Chem	2.08	130	166	160	52	395	Renault Zoe e-tech electrique
C/**NMC**	Pouch	LG chem	3.65	52	316	152	18	145	Smart fortwo Electric
C/**NMC532**	Pouch	LG Chem	-	-	-	-	78	292	Volvo XC40
C/**NMC111**	Prismatic	Samsung SDI	3.70	37	357	185	36	300	VW e-Golf
C/**NMC721**	Pouch	LG Chem	1.85	145	267	164	58	350–544	Volkswagen ID.3
C/**NCA**	Cylindrical	Panasonic	3.60	3.4	673	236	60–100	330–500	Tesla Model S
C or SiOC/**NCA**	Cylindrical	Panasonic	3.60	4.75	683	260	75–100	490–630	Tesla model 3
C or SiOC/**NCA**	Cylindrical	Panasonic	3.60	3.4	673	236	60–100	330–500	Tesla X
C/**NCA**	Cylindrical	Panasonic	-	-	-	-	42	182	Toyota rav4 EV
C/**LFP**	Prismatic	BYD	3.20	216	279	166–140	86	505	BYD Tang Electric
C/**LFP**	Pouch	LG Chem	3.75	27	144	90	19	132	Chevrolet Spark EV
C/**NMC622-LMO**	Prismatic	Samsung SDI	4.08	105	-	126	42	320	Fiat 500e
C/**LMO-NMC**	Pouch	LG Chem	3.70	16	-	-	36	160	Ford Focus Electric
C/**LMO-NMC**	Prismatic	Li Energy Japan	3.70	50	218	109	16	132	Mitsubishi iMiEV

2.1. Nonlayered Cathode Materials: LFP Technology

Lithium iron phosphate $LiFePO_4$ (LFP), a cathode with an olivine structure developed in 1996 by John B. Goodenough (a Nobel laureate in chemistry), exhibits excellent cycle life, enhanced safety, and elevated thermal and electrochemical stability owing to the strong bond energy of its PO_4 tetrahedral units [14]. These characteristics, accompanied by its abundant usage of iron and inherently inexpensive cost, make it an attractive cathode alternative to replace structurally unstable LCO. However, this material has been limited by its poor energy density (190 $Wh.kg^{-1}$-cell level) and low electronic conductivity ($\approx 10^{-9}$ S cm^{-1}), leading to relatively high cost per kWh despite low material costs [15]. In order to enhance the surface and structural conductivity, two efficient strategies were adopted, using either conducting agent (carbon) coating or metal doping onto the LFP electrode [16]. In addition, nanoscale materials are effective at improving the electronic conductivity by reducing the lithium-ion diffusion pathway. To date, various battery companies, including A123 and BYD, have successfully commercialized cheap carbon-coated nano-LFP materials using rotary kilns. Despite these solutions, LFP technology is within close range to its uttermost theoretical value (~170 $Wh.kg^{-1}$), having little room for further development to achieve the next-generation EV energy density (>250 $Wh.kg^{-1}$ at a cell level in 2025). Moreover, because of its low volumetric energy density (220 $Wh.L^{-1}$), high safety, and fast charging times ($\approx$2.5 h), LFP is more suitable in public transport and heavy-duty applications and in standard-range cars (e.g., the Chevrolet Spark, BYD (e6), and BMW Active Hybrid 3 and 5 series) [17]. Moreover, LFP still dominates in commercial EVs (23.8 GWh) and offers unmatched total energy throughput, capable of 600,000–800,000 km accumulated driving distance [18].

2.2. Nonlayered Cathode Materials: LMO Technology

In 1983, M. Thackeray and coworkers developed spinal lithium manganese oxide $LiMn_2O4$ (LMO), and after 13 years, it was commercialized by Moli Energy. LMO cathodes offer an additional opportunity to eliminate cobalt while also profiting from lithium wt.% reduction when compared with NCA and NMC (layered transition metal oxides) [19]. Their three-dimensional spinal structure provides a facile Li^+ diffusion pathway and thus the capability of higher rates than those of 2D frameworks [20]. LMO is similar to LFP, as it is inexpensive (<\$10 kg^{-1}) compared with other cathode materials and low in energy density, but it delivers high power, which is pivotal for EV applications. LMO's utilization is limited by its low capacity (theoretical capacity: 148 $mAh.g^{-1}$) and short lifetime owing to its Mn dissolution (i.e., structural instability) upon cycling in the electrolyte [21]. Thus, 70% NMC is blended with LMO-type cathodes to bring out the best in each system by prolonging the lifetime and enhancing the capacity of the NMC part alongside the low cost and high-rate capability of LMO-type cathodes. This LMO/NMC composite has been implemented in most EVs, such as the BMW i3, Chevy Volt, and Nissan Leaf. In the long term, the next-generation high-voltage $LiNi_{1/2}Mn_{3/2}O_4$ spinel (HV-LNMO) is the main focus of research development for EVs (Figure 4d). Nickel incorporation into LMO spinal allows for high energy density (~580 $Wh.kg^{-1}$) and high operating voltage (~4.75 V vs. Li/Li^+) through a two-electron $Ni2^+/4^+$ redox couple [22]. This enhancement in energy density led to material cost reduction, regardless of the addition of an expensive material (Ni), as less material was needed per kWh compared to LMO [10]. However, LNMO suffers from severe capacity fading (it still cannot achieve more than 200 cycles to 80% capacity) and incompatibility with commercial electrolyte, leading to electrolyte decomposition at high voltage (>4.5 V) [23]. Therefore, various strategies are still under investigation to improve such failure mechanisms, including high-voltage electrolytes, surface coatings, doping strategies, and particle morphology optimization [24].

2.3. Layered Cathode Materials: NCA Technology

Layered lithium nickel cobalt aluminum oxide ($LiNi_{1-x-y}Co_xAl_yO_2$, NCA) is a potential new archetypal cathode material to replace structurally unstable lithium nickel oxide ($LiNiO_2$, LNO). The current focus is to further increase the relative fraction of Ni in its composition, resulting in simultaneously reducing cobalt utilization and increasing energy density, thus lowering the cost/kWh in both ways. However, it is well known that increasing the Ni content in NMC cathodes results in the deterioration of both thermal stability and cycling performance. Meanwhile, the optimized NCA (NCA-80, $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) shares similar characteristics with the NMC811 by exhibiting a high specific energy (200 $mAh.g^{-1}$ at 4.3 V vs. Li/Li^+), long lifespan (>15 years), and reasonable specific power (>200 $Wh.kg^{-1}$ at the cell level), thus joining NMC cathodes as front runners within the next generation of hybrid EVs [25]. Moreover, the elimination of manganese in NCA-80, -81, and -82 materials leads to good capacity retention compared with NMC811, whereas the incorporation of aluminum ions (Al^{3+}) aims to minimize detrimental phase transition, enhance the thermal stability, and increase the operation voltage (via the weakening of the Ni–O bond by the stronger Al–O bond) while reducing the overall cost [26]. Although material costs for LFPs (Co- and Ni-free materials) are approximately 50% lower than those of NCAs, the reality is that the NCAs require higher voltage (3.6 V compared to 3.2 V vs. Li/Li^+), with limited quantities of active materials and ~30% higher capacity. This serves as a counterbalance to the concerning high expense [26]. At room temperature, the stability of the reversible structural evolution (hexagonal 1–2 phase) during long-term cycling is crucial. For this reason, the setup is kept at 60% depth of discharge to have good capacity retention. However, operating at elevated temperature or voltage initiates fast capacity fading owing to grain boundary breakage and the aggressive formation of a solid electrolyte interface (SEI) [27]. The main reason affecting power fade and capacity decay is the high interfacial resistance at the cathode, while the polarization and overpotential are caused mainly by the formation of NiO structures on the surface at elevated temperatures

(>60 °C). In a few scenarios, NCA is safer than other battery materials, primarily because of its high tolerance of overcharge, as it offers a slightly lower potential at full charge. The energy density for NCA technology is anticipated to reach 700 $Wh.L^{-1}$ and 300 $Wh.kg^{-1}$ at a cell level by 2025 [28]. In 2006, Tesla commercialized its first Roadster "long-range EVs" using Panasonic cylindrical 18,650-type Li-ion batteries with NCA cathodes. The NCA/C chemistry is mainly employed by Tesla products (the Model S-2010, Model X-2015, and Model 3-2017) and other models including the Mercedes-Benz S400, Toyota RAV4 Hybrid, and VW E-Golf. Recently, BASF launched a new high-energy-density cathode active material (HEDTEM NCA) for automotive drivetrains by increasing the Ni content from 80% to 90% [29]. Even though the detailed technology is restricted because of confidentiality, it is crucial to increase the mechanical strength of the particles as well as raising the electrode density to 3.6 g cm^{-3}. Developing single crystalline nickel-rich cathodes could be a direction with a bright future for improving EV power by following the successful story of LCO [30].

2.4. Layered Cathode Materials: NMC Technology

Layered $LiNi_{1-x-y}Mn_xCo_yO_2$ (NMC) cathodes are among the most widely researched lithium-ion systems. They combine nickel, LMO, and LCO. NMC has low internal resistance from manganese, high capacity from nickel, and low costs due to less cobalt content in the lattice [31], with specific capacity similar to or higher than that of LCO at similar operating potential. NMC's design is variable by modifying the stoichiometry of the metal elements. Specifically, by incorporating more Ni (redox Ni^{2+}/Ni^{4+}), higher energy density and capacity can be attained, albeit with poorer thermal stability and lower calendar life. Meanwhile, manganese has the advantage of forming a spinel structure to attain less internal resistance but results in poor specific energy. The presence of cobalt (Co) improves electric conductivity, which fosters high-rate capability. However, increasing the Co content substantially increases the material cost. Combining these three metal elements enhances their merits. Thus, various formulations were developed through low-cobalt NMC cathodes while exhibiting continuously increased capacity with higher Ni content, for instance, $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC-111, 160 $mAh.g^{-1}$), $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC-532, 170 $mAh.g^{-1}$), $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC-622, 180 $mAh.g^{-1}$), and $LiNi_{0.85}Mn_{0.1}Co_{0.1}O_2$ (NMC-811, 200 $mAh.g^{-1}$) at 4.3 V Li/Li^+ [32]. Furthermore, higher ionic diffusivity (10^{-8}–10^{-9} $cm^2.S^{-1}$) and electronic conductivity (2.8×10^{-5} S cm^{-1}) improve the rate capability of NMC-811 compared with NMC-111 (ionic diffusivity: 10^{-11}–10^{-12} $cm^2.S^{-1}$ and electronic conductivity: 5.2×10^{-8} S cm^{-1}) [33]. The most recent cutting-edge cathode materials for LIBs are NMC-111, NMC-442, and NMC-532 (Figure 4d). Within the near future, Ni-rich NMC cathodes (NMC-811 and NMC-622) will be highly implemented in the EV industry because of their low costs and high specific energy. Even though NMC-811 displays the best electrochemical performance amongst these NMC series materials, it also exhibits the most surface reactivity (similar to that of NCA) because of nickel ions' instability in the liquid organic electrolyte in the delithiated state [34]. The structural instability of the NMC-811 results in thermal safety challenges due to its high exothermic self-heating rate and low exothermic onset temperature (120 °C). Apart from NMC-811, the reactivity of other NMC cathodes with electrolytes is influenced by the upper cutoff potential, especially at 4.7 V, indicating a trade-off between safety and high energy density (e.g., NMC-811 charged to 4.4 V, ~234 $mAh.g^{-1}$, compared with 4.7 V, ~260 mAh g^{-1}). This prompts the need for additional cathode components to protect the surface to mitigate the undesirable parasitic reactions between the electrolyte and the electrode, such as electrode coatings, which represent a widely utilized technique. This posttreatment typically generates coverage and interface issues due to the incompatibility of the foreign material [33,35], resulting in segregation during electrochemical cycling and chemical synthesis. Beyond simple surface coatings, core–shell structures are preferred as a better approach to carry out uniform encapsulation of Ni-rich NMC materials. In these, Mn-rich sections occupy the surface (shell) to enhance the stability, while Ni-rich

sections dominate the particle core, providing desirable electrochemical performance [36]. The core–shell concept has been optimized at the primary particle level by creating a radial concentration gradient shell family with Ni-rich cores (> 80%) using a coprecipitation method in a continuously stirred tank reactor [37], resulting in better rate performance and enhanced thermal stability. These high-energy NMC (HE-NMC, $Li(Li_wNi_xCo_yMn_z)O_2$) layered–layered composite materials are the next generation battery cathode materials because of their better capacity retention and specific capacities (>270 $mAh.g^{-1}$) compared with conventional NMCs [37]. These HE-NMCs, however, suffer from various issues that limit their practical applications, such as (i) poor rate capability; (ii) voltage decay and large voltage hysteresis; (iii) huge, irreversible capacity fade during the initial cycle; (iv) slow kinetics and electrode damage that result from the anionic redox ($2O^{2-}/O^{2n-}$ [$n < 4$]); (v) structural instability and cation migration during charge–discharge; and (vi) electrolyte oxidation at higher potentials.

Various attempts have been made to overcome these shortcomings, including (i) cationic substitution, (ii) electrolyte additives, (iii) surface coating, (iv) anion/cation doping, and (v) acid treatment. Recently, disordered rock salt (DRX) $LiMO_2$ cathodes represented cobalt-free layered cathodes that required excess lithium and d0 metal species [38]. The addition of d0 species into $Li(MM')O_2$-type structures assisted in stabilizing the disordered arrangement. However, these materials are at an initial stage of research development. For sustainability reasons, titanium (Ti^{4+})-based oxides offer the advantage of being earth abundant and d0. The replacement of oxygen by fluoride anions led to high energy densities (~1000 $Wh.kg^{-1}$) and reversible capacities (>300 $mAh.g^{-1}$, 1.5-5 V vs. Li/Li^+) by preventing the occurrence of irreversible oxygen loss and/or O_2 redox reactions [38].

So far, cathodes with 60% Ni content have been fully commercialized with ~3.4 g cm^{-3} electrode density [22]. Toshiba commercialized an NMC/LTO prismatic cell for the Honda Fit EV (2013), while Panasonic/Sanyo and LG commercialized NMC/C prismatic cells for the VW e-Golf (2015) and Chevrolet Bolt (2016)/Renault Zoe (2017), respectively [39]. Even LG Chem invested in an NMC cell production facility for the Bolt and Chevy Volt in Michigan, United States. In 2017, the Bolt received three titles as the North American Car of the Year, the Motor Trend Car of the Year, and an Automobile Magazine 2017 All Star and was listed among the best 25 inventions of 2016 [40]. Several companies are actively manufacturing Ni-rich NMC and NCA cathode technologies to meet the EV market's demands, including SK Innovation, Li Energy Japan, BASF, General Motors, Li-Tec, and TODA [41]. Driven by the need for lower costs and higher energy density, all leading cathode manufacturers, battery makers, and automotive OEMs with BEV portfolios have invested heavily in commercializing nickel-rich cathodes. The year 2019 is set to be a watershed year for NMC 811, as CATL, LG Chem, and SK Innovation are all expected to start 811 production, and China's 2019 NEV subsidy policy includes aggressive incentives for 811 adoption.

3. Promising Anode Materials for High Energy Density

3.1. Silicon Technology for Near-Term EVs

Silicon (Si), known as an alloying anode, has been regarded as one the biggest near-term advanced Li-ion breakthroughs for replacing graphite. Si offers significantly higher theoretical capacity (~4200 $mAh.g^{-1}$), lower potential at ~0.4 V vs. Li/Li^+, and abundant resource availability on Earth [42]. Besides these advantages, silicon's superior capacity also causes one of its most challenging problems, as the material swells nearly 400% upon lithiation compared with the 7% swelling typical for graphite [43]. The large structural deformation causes significant structural strain and fracture/reformation of the Si particle, along with solid electrolyte interphase (SEI), during the initial few cycles, resulting in low Coulombic efficiency and capacity loss. Mitigating these effects requires engineering costly nanostructures, mixing silicon with carbon to make a composite (Figure 5), or using electrolyte additives to stabilize SEI formation.

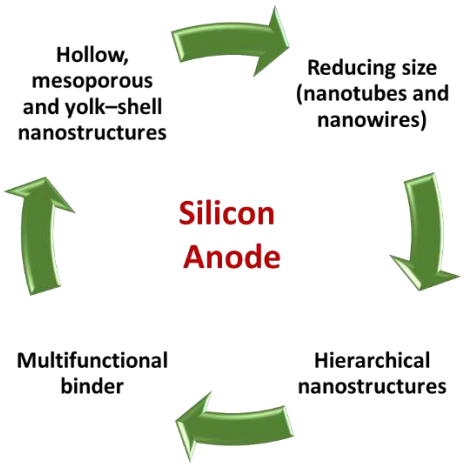


Figure 5. Schematics of several methods for enhancing the performance of Si anodes.

Although these methods can achieve high energy densities, manufacturing with them is prohibitively expensive. Therefore, completely replacing graphite with Si is not realistic at the moment; cheaper silicon–carbon composites, which today use less than 10% silicon, offer only minor performance improvements but have been commercialized in high-volume applications. Panasonic became one of the first cell manufacturers to use silicon–carbon composite anodes (Si or SiO_x) in the 2015 Tesla Model S (driving range: 330 km) and the 2017 Tesla 3 (driving range: >490 km); today, it leads all other companies in patent filings. Most other major cell manufacturers now offer a silicon-containing anode, albeit at lower volumes, and materials suppliers have also been able to innovate to retain their roles in the supply chain [43].

3.2. Li Metal Anode

Lithium metal anodes offer 10 times greater theoretical specific capacity than graphite anodes. With its unbeatable theoretical capacity (3860 $mAh.g^{-1}$), lithium metal is the ideal material for LIB anodes. Ideally, at the cell level, substituting a graphite electrode with lithium metal would result in 50% and 35% enhancements in energy density and specific energy, respectively (Figure 6a) [44]. Although metallic lithium anodes offer substantial improvements over graphite (but low cycle life), safety concerns have prevented their use because of the following aspects, as shown in Figure 6b: (i) metallic lithium is highly reactive and expands during ion uptake; (ii) the formation of hair-like lithium dendrites on the anode surface remains a significant problem, reducing capacity and limiting cycle life; and (iii) on repeated cycling, dendrites can unevenly grow through battery separators, causing short circuits and even battery fires [45]. Nonetheless, the strengths of lithium metal encourage its further development, and several manufacturers are initiating commercialization of batteries with various methods of sealing lithium in anodes [46] or applying protective layers and pairing the anodes with solid-state electrolytes. Moreover, it is recommended to use thinner lithium discs (<30 μm) rather than thicker Li to obtain more chemistry stability and the ability to notice soft short circuits [47]. This approach permits rapid detection of the issues of attaining dendrite-free cycling and acceleration of the process of commercialization. However, academic research will probably encounter newfound challenges during prototyping. Table 2 elucidates four different processing routes for highly reactive and adhesive Li metal anode, including vapor-based [48], melt [49], dry [50], and anodeless methodologies [51], and only two approaches for graphite or silicon–graphite composites [52,53].

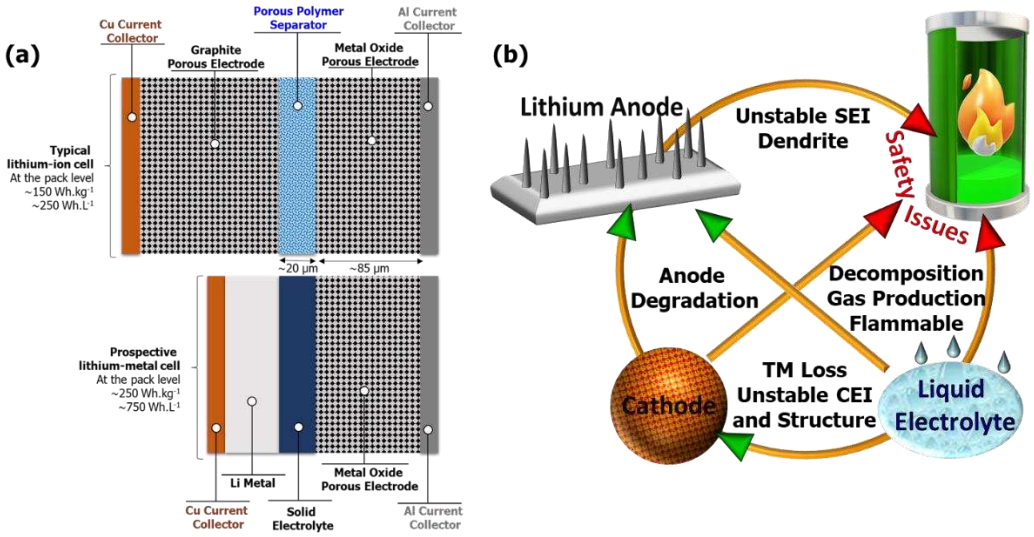


Figure 6. (**a**) A typical Li-ion cell (**top**) and a lithium metal cell (**bottom**) containing a solid separator and a dense layer of metallic lithium. (**b**) Failure mechanisms and safety hazards of Li-metal anodes using conventional liquid electrolytes.

In order to obtain high energy density, it is important to reduce the amount of inactive compounds, such as the solid electrolyte separator, binder, and conductive additive. The electrolyte, binder, and carbon additives provide ionic conductivity, mechanical stability, and electronic conductivity to the electrodes, respectively. Comparatively, liquid electrolytes offer good contact between the particles during cycling, while solid electrolytes still face many challenges. During cycling with SSE, the loss of interfacial contact between the particles can generate limitations for electronic conductivity, so it is essential to optimize the composition of these three elements.

Separators allow the passage of lithium ions while blocking electrons between electrodes. Some separators are not stable at high temperatures, which can cause the degradation of separators and internal short circuits. Furthermore, some separators are hydrophobic, which makes the passage of lithium ions difficult. Consequently, it is essential to make efforts to develop new-generation solid electrolytes.

Carbon additives are used in the electrode composition but can contribute to the decomposition reaction of solid electrolytes because they can generate interfacial resistance [54]. Conductive additives with low surface areas (vapor-grown carbon fibers, VGCF) have better diffusion pathways, limited parasitic reactions, and good performance compared with those with high surface areas [55].

3.3. Solid-State Electrolytes

EVs batteries on today's market are dominated by anodes of silicon, graphite, and their combinations with the conventional liquid electrolyte. Leading developer Sion Power claimed that its cells with conventional lithium foil anodes could achieve 470 Wh.kg^{-1} and that its cells using proprietary protected lithium anodes could achieve 550 Wh.kg^{-1} [43], values much higher than those of the best LIBs with graphite anodes (280 Wh.kg^{-1}). Start-ups in this space currently source anode materials from laboratory chemical providers, as there is not yet a strong supply chain for lithium metal foils. There is a need for the lithium industry to build the capacity to address this need within the next 10 years before solid-state batteries reach commercialization, as these batteries achieve higher capacities than incumbent Li-ion batteries only if they use metallic lithium anodes.

Table 2. Processing routes for anode materials, including their advantages and challenges.

Methodologies	Pros	Cons and Challenges
Graphite/Silicon		
Wet processing	▪ Well-established process ▪ High throughput	▪ Drying process of the film ▪ Critical for some water-based processes
Solvent-free (or-reduced) approach (e.g., electrostatic spraying, extrusion)	▪ Elimination of the solvent and drying process lead to cost and energy savings	▪ Film uniformity problems ▪ Challenging to scale up
Li metal		
Melt processing (liquid phase)	▪ Thin layers ▪ Applicable for porous structures	▪ Immature process ▪ Special safety precautions: handling Liquefied lithium and inert atmosphere/Vacuum required
Vapor-based processing (e.g., evaporation, sputtering)	▪ High-quality and thin films	▪ Expensive process ▪ Limited throughput and thickness ▪ Vacuum required
Plating	▪ No direct handling of metallic Li ▪ Thin layers ▪ No excess Li	▪ Homogeneity problems ▪ Large-format batteries not utilized yet ▪ Pretreatment (formation) necessary ▪ No excess Li (capacity fading due to SEI formation)
Dry processing (extrusion)	▪ Well-established process	▪ Processing of thin foils ▪ Defect rate ▪ Handling Li (adhesive as it sticks to rolls) ▪ Dry/inert atmosphere required

The next generation of batteries will be solid-state batteries (SSBs) in order to challenge incumbent Li-ion technology, as the conventional LIB has reached a bottleneck period of development. Minimal amounts of liquid or polymer electrolytes added to SSBs will lead to better reversibility capacity and greater cycle durability at the electrode/electrolyte interface. However, this approach is not well-defined, as some papers have used different terminology, ranging from "hybrid solid" to "quasisolid" to "solid". Therefore, Figure 7 classifies batteries according to their content to avoid confusion for new battery scientists. From a technical point of view, competing electrolyte technologies can be compared across seven characteristics to differentiate the resulting battery performance, namely safety, interfacial properties, electrochemical and thermal stabilities, withstanding dendrite formation, electrolyte decomposition, and conductivity, as shown in Figure 7b.

Solid electrolytes are promising candidates for improving safety and enabling high energy density when using Li-metal anodes. For most of the 1990s and 2000s, the solid-state battery IP space was relatively inactive, reflecting its niche and academic nature at the time. However, this picture has changed dramatically during the past five years, as since 2013, patent publications have been steadily rocketing up in solid-state batteries. While the United States and Japan have been long-time leaders in pushing innovation in this area, China has been a big story in the past few years, as it now accounts for more than 30% of patent filings in the space. Toyota, Bosch, Panasonic, LG Energy Solution, and NGK Insulators lead patent activity, with more than 1000 patents each. Key developments include three promising solid electrolyte materials, organic polymer electrolytes, inorganic sulfide-based electrolytes, and inorganic oxide-based electrolytes, as illustrated in Figure 8 [56].

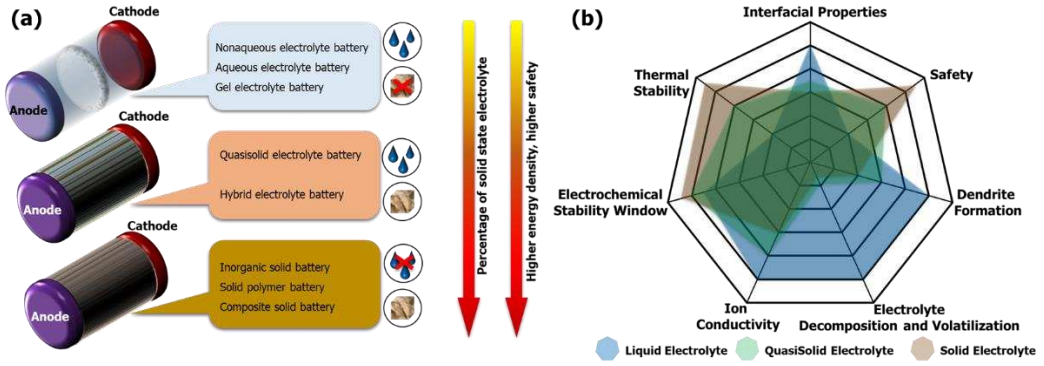


Figure 7. (**a**) Schematic illustration and (**b**) radar chart of three types of batteries based on the liquid content and SSE percentage in the assembled batteries.

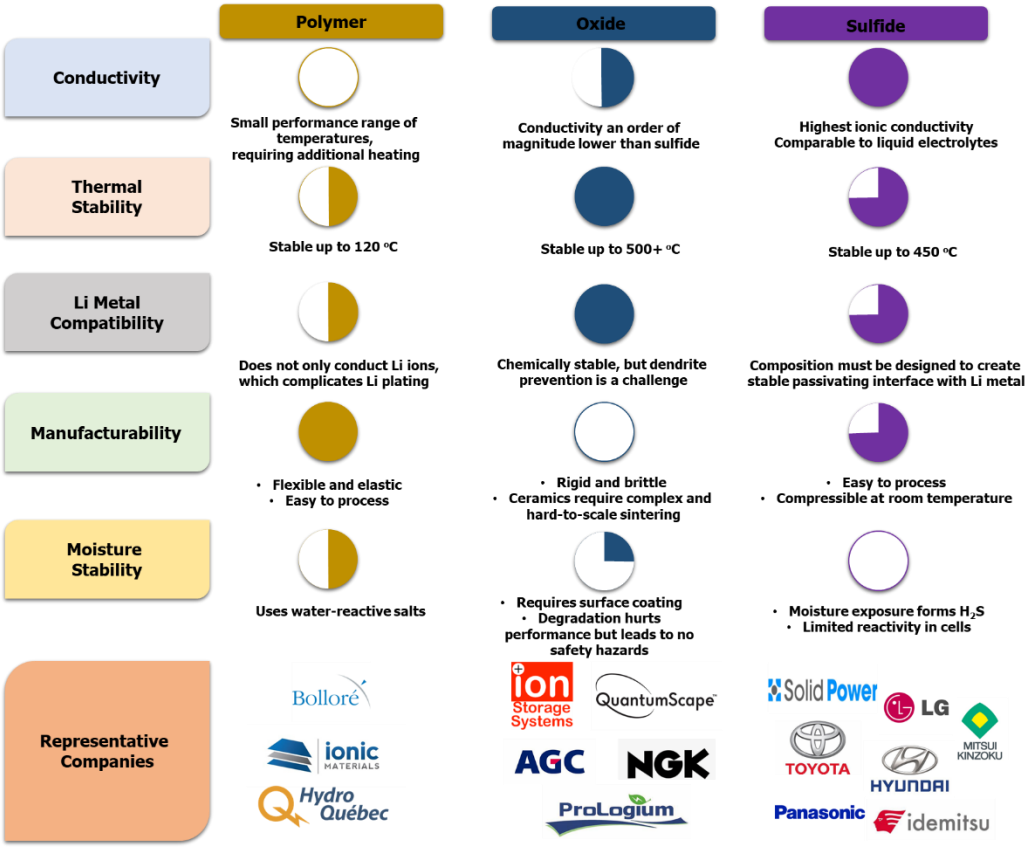


Figure 8. An overview of key material properties for the design of SSBs.

(i) Organic polymer solid electrolytes: Polymers are the most mature solid electrolytes that have been adapted to large-format applications such as EVs and stationary storage. Polyethylene oxide (PEO) is the most common material that has seen commercial viability, and others being researched include polyacrylonitrile (PAN), polyvinylidene fluoride (PVDF), and polyphenylene sulfide (PPS) [57]. Most polymers can be adapted to conventional manufacturing techniques, and polymer extrusion is a well-understood industrial process. PEO polymers have low room-temperature ionic conductivity, which results in poor power output and slow charging. Low voltage stability also limits cathode choice and cutoff voltage.

(ii) Inorganic sulfide-based solid electrolytes: Sulfide-based inorganic electrolytes are the closest to commercialization in large-format cells. LGPS systems ($Li_{10}GeP_2S_{12}$) and LPS systems ($Li_7P_3S_{11}$) are the most pursued materials [57]. Sulfide-based ma-

terials exhibit mechanical properties that allow for roll to-roll processing without unreasonably high pressures and are electrochemically stable at both high and low voltages. Exposed to air or water, sulfide materials generate toxic hydrogen sulfide gas. This is both a risk, potentially exposing customers to these gases, and an added cost, requiring additional dry-room requirements during manufacturing.

(iii) Inorganic oxide-based solid electrolytes: Oxide-based electrolytes attract less attention than organic polymers or sulfide-based inorganics but show promising performance. Lithium lanthanum zirconium oxide (LLZO) is the most investigated material, while lithium phosphorous oxynitride (LiPON) has been investigated for thin-film batteries [57]. Very high ionic conductivity and wide electrochemical stability offer promising performance, while LiPON can be deposited in thin layers for thin-film and flexible batteries. These materials are very brittle and cannot typically be adopted for use in conventional roll-to-roll manufacturing, requiring time- and energy-intensive deposition processes or tape-casting. High interfacial resistance with lithium leads to lower power output and low efficiency. Perovskite has the general formula ABO_3 (A = Ca, Sr or La; B = Al, Ti) [58]. The most studied perovskite electrolyte is $Li_{3x}La_{2/3-x}TiO_3$ (LLTO), with bulk Li+ ion conductivity of 10^{-3} S. cm^{-1} at 25 °C [59]. Ti-based perovskite electrolyte is thermally and chemically stable in air/humid conditions. However, LLTO can be deteriorated by the reduction of Ti^{4+} to Ti^{3+} when it touches the Li metal anode as LATP [60]. In addition, Na superionic conductor, or NASICON ($Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$, $0 < x < 3$), has been found to exhibit high ionic conductivity [61]. The substitution in NASICON generates structural modification and allows improving the conductivity of compounds [62]. The maximum ionic conductivity is $\sim 1 \times 10^{-3}$ S. cm^{-1} at 25 °C for $Li_{1+x}Ti_{2-x}Al_x(PO_4)_3$ (LATP), where $x \approx 0.3$ [63]. However, at low potential ($\leq$1.5 V vs. Li/Li+), the reduction reaction (Ti^{4+} to Ti^{3+}) can occur when NASICON is in direct contact with lithium metal, which induces undesirable electronic conductivity and short circuit of the cell [64].

Based on numerous chemistry approaches, polymer electrolytes represent the most mature technology, while ceramic and composite electrolytes will require more time to commercialize. Table 3 below illustrates three different production approaches (wet [50,65], powder-based [66], and solvent-free processes [50]) that are currently utilized in academia and industry for solid electrolytes. Early adoption of solid-state electrolytes is motivated by a greater need for safety and potentially lower production costs. Developers such as Ionic Materials have achieved comparable results to current technologies, but their success hinges on integration with current production capabilities. Solid electrolytes that require additional environmental considerations, such as sulfides, or are too rigid for roll-to-roll manufacturing, such as oxides, will take longer to be implemented. Early manufacturability will help decide which technology gets to market first, but in the long run, we expect a diversity of solid-state electrolyte materials.

A Lux industrial report expected to see commercial use of solid-state electrolytes in BEVs no earlier than 2030 [56]. What will push solid electrolytes into commercial production over liquid systems will be cost and performance metrics. Enhanced performance will depend on other components of the battery system, such as anodes and cathodes. The solid-state battery landscape is led by a mix of automotive players, materials developer, and battery makers. Start-ups such as Solid Power and QuantumScape have emerged as key players, with Solid Power focusing on manufacturability and QuantumScape on low-temperature operation and fast-charging solid-state batteries. Toyota Motor Corporation has led solid-state battery development efforts, including a joint venture with Panasonic to commercialize solid-state technology. Research centers such as Hydro-Quebec CETEES have historically focused on lithium metal anodes and have set their sights on polymer–ceramic composite electrolytes. Solid-state battery investments were sluggish until 2018, when QuantumScape received $100 million in a corporate round with Volkswagen to commercialize its technology. The company has cumulatively raised $1 billion in total. Since 2018, numerous strategic investments have boosted the field's investment activity

to more than $200 million. In the first half of 2021, Solid Power took a major step toward commercialization in addition to a round for $150 million led by BMW Group, Ford Motor Company, and Volta Energy Technologies [43,56]. QuantumScape and Solid Power are favored to be the first start-ups to commercialize the technology, having joined the SPAC craze. Automakers and energy companies are betting on solid-state technology to be the winning innovation in Li-ion batteries; however, start-ups will require far more support to successfully displace liquid electrolyte manufacturing. Automakers such as Toyota have made large investments in solid-state batteries, including a joint venture (JV) with Panasonic, but have yet to commercialize the technology. The difficulty of manufacturing at scale remains a key issue, as typically, industries remain too small today, and the processes they use remain too exotic. Moreover, power output remains a concern, and performance advantages require using the elusive metallic lithium anode.

Table 3. Processing routes for solid electrolytes including their advantages and challenges.

Methodologies	Pros	Cons and Challenges
Aerosol deposition	■ Solvent-free ■ Thin, dense, and high quality layers ■ Oxide electrolytes: sintering can possibly be omitted	■ Low deposition rate ■ Vacuum needed ■ Immature and expensive process
Solvent-free (or minimized) processes (e.g., dry calendaring, extrusion)	■ Solvent-free or -minimized ■ Polymer electrolytes: extrusion is an established process ■ Elimination of the solvent and drying process to lead cost and energy savings	■ Inorganic electrolytes: challenging to generate uniform and thin layers ■ Oxide electrolytes: sintering (excess lithium needed, low throughput, expensive, and energy intensive)
Wet processing	■ High deposition rate ■ Similar process and equipment as for AAM/CAM processing ■ Sulfide electrolytes: subsequent calendaring/ cold-pressing possible	■ Film drying and solvent recovery ■ Challenging to generate thin layers with low porosity ■ Oxide electrolytes: sintering (excess lithium needed, low throughput, expensive, and energy intensive) ■ Sulfide electrolytes: limited options of binders and solvents

AAM: Anode Active Material & CAM: Cathode Active Material.

4. Conclusions and Outlook

Li-ion technologies (generations 3 and 4) will globally govern mobility applications by 2030+. Battery materials represent 50–70% of the cost, including the anode, cathode, electrolyte, and separator. These advanced chemistries are vital for better-performing, safer, more cost-efficient, and more sustainable battery cells, enabling market uptake of battery technology. Most of the battery technologies beyond Li-ion chemistries are still in either in the prototype or research phase and may not appear on the commercial market until 2025. Figure 9 summarizes key recommendations for Li-ion batteries for mobility (generations 3 and 4), along with short- and long-term research needs.

The most straightforward strategy among major battery manufacturers and automotive OEMs has been to reduce the use of expensive cobalt by switching to high-nickel chemistries. However, this swift shift entails demand for higher-quality electrolytes and higher-purity lithium and may lead to battery stability and safety issues. Thus, complete removal of cobalt from batteries seems undesirable. In the short term (<2025 period), EVs will depend on the mature battery technologies, including NCA, NMC, and blended LMO/NMC chemistries, for developing lower-cost and higher-specific-energy batteries such as NMC-811 and Si. However, heavy-duty applications that require long-cycle-life batteries will not rely on nickel-rich cathodes and lithium metal anodes. Instead, they

will use low-nickel NMC or LFP and graphite. Considering the cell potential is limited to 4.2 V vs. Li/Li^+, as the electrolyte will start oxidizing above that, the adoption of a new electrolyte with a higher oxidation potential will enable the usage of high-voltage cathodes in the battery cell, leading to higher specific energy and lower weight and costs, therefore improving the driving range of electric vehicles. In the long term (>2025 period), solid-state batteries seem to be the most promising technology for use in EVs and the cornerstone for the future of gigafactories for their production. Particularly, Li metal solid-state batteries could lead EV applications to new opportunities, because metallic Li is one of the highest-performing anodes for next-generation LIBs. SSEs also assure battery safety via their superior physical and chemical and physical stabilities and by suppressing Li dendrite problems and side chemical reactions that originated in conventional LIB systems. In the meantime, developing LIB recycling is a crucial approach to ease a supply and demand matching problem in the future.

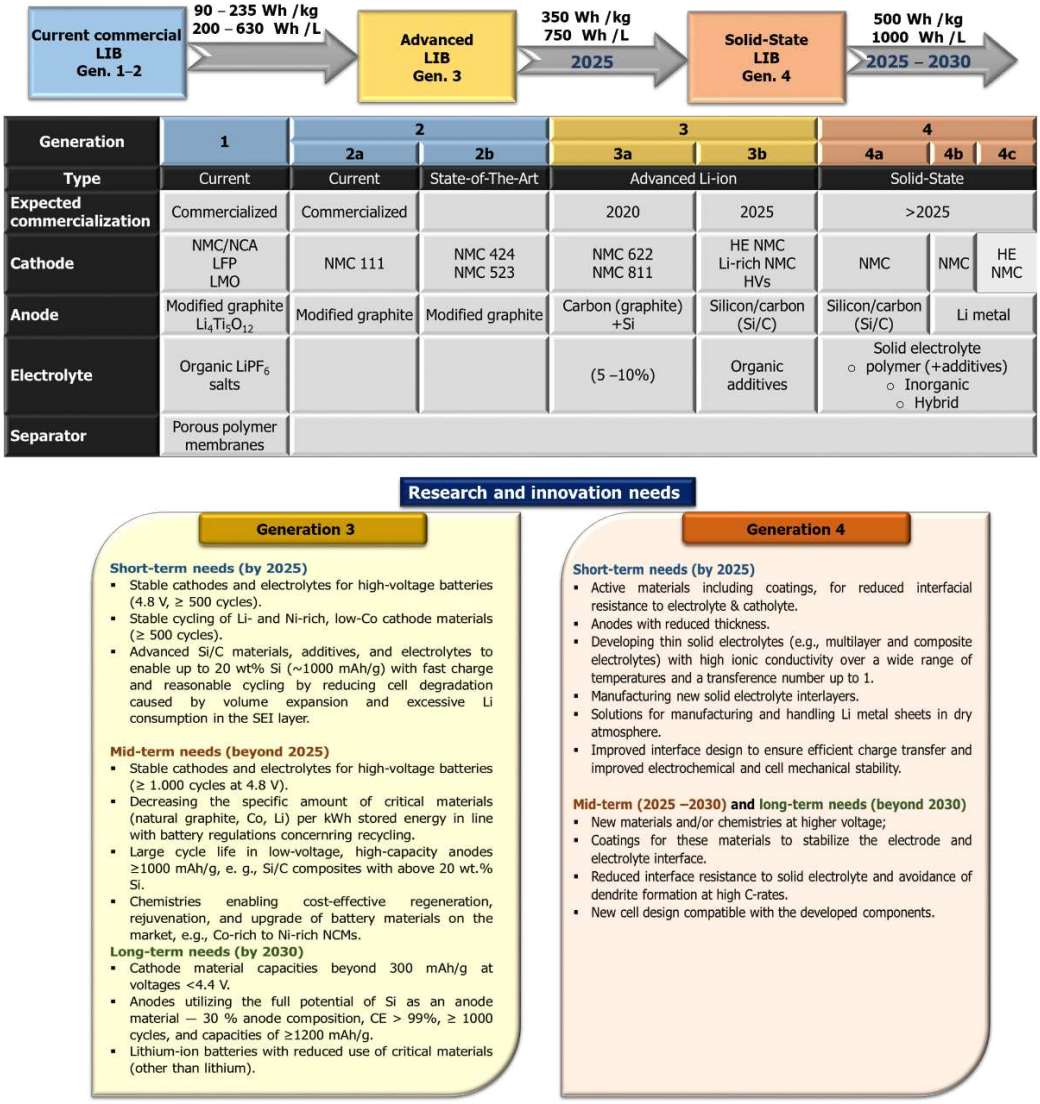


Figure 9. An overview of key recommendation of different Li-ion batteries generations for mobility along with short- and long-term research needs.

One major aspect that should be considered is if mineral prices spike. If lithium, cobalt, and nickel all experience price spikes in the future, battery costs will skyrocket. Thus, it is imperative that suppliers work closely with downstream customers and develop innovative processes to mitigate the worst effects of tight commodity supply [43].

Besides the concerns for the material development for LIBs, the cell structure of battery cells, especially of prismatic cells, has a significant impact on EV performance. However, prismatic cells require high-end passive BTMSs (battery thermal management systems) because of their thermal instability. Cylindrical cells are used for longer range, recharge cycles, and thermal stability, as they are spaced out in the module with passive BTMSs. In the future, new cell structures that permit a cell to pack design (eliminating EV modules) could significantly reduce the complexities and material in battery pack designs [67].

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