ACS Energy Letters

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PERSPECTIVE

Opportunities and Challenges of Lithium Ion Batteries in Automotive Applications

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Cite This: *ACS Energy Lett.* 2021, 6, 621−630

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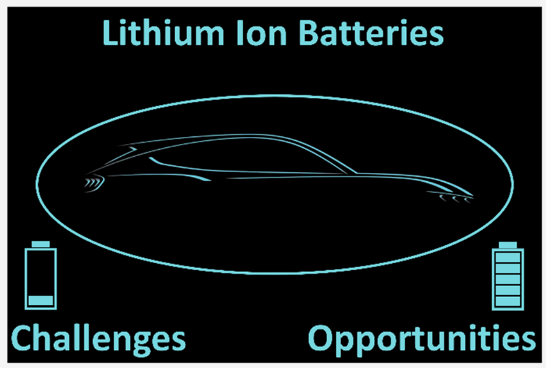
ABSTRACT: Lithium ion batteries (LIBs) have transformed the consumer electronics (CE) sector and are beginning to power the electrification of the automotive sector. The unique requirements of the vehicle application have required design considerations beyond LIBs suitable for CE. The historical progress of LIBs since commercialization is compared against automotive application goals and requirements. Vehicle-driven battery targets are discussed and informed by a set of international research groups and existing production electric vehicles' performance. The opportunities and challenges remaining for the transition of LIBs suitable for CE to the automotive sector are assessed in terms of energy, life, cost, safety, and fast charge capability.



The modern Information Age has been enabled by advances in semiconductor and computing technology, but it has largely been powered by the rise of the lithium ion battery (LIB). Since its introduction in 1991 by Sony Corporation for digital camcorders, the LIB has achieved widespread use in personal electronics and, in the past decade, electrified transportation. The development of this technology has its roots in the 1960s and represents decades of focused R&D.[1,2] Since their initial mass production, the specific energy (Wh/kg) of cylindrical consumer electronics LIBs has improved at about 6%/year (see Figure 1).[3] The ubiquity of LIBs and the understanding of their basic operating mechanisms have matured to the point where chemistry educators have created a building block game to teach their operation to school children.[4]

Although the progress of LIBs has been significant since their introduction (see Figure 1), there are several technical challenges for LIBs to meet the future needs of the automotive application. When compared to consumer electronics, automotive applications have more stringent technical requirements such as calendar life (10 years), cycle life (1000 cycles), temperature range (−30 to 52 °C), and cost ($100/kWh).[5] These higher performance requirements are largely responsible for the 17-year delay between the introductions of LIBs in consumer versus automotive applications.

A variety of challenges and opportunities exist for automotive LIBs in the present day. Further advancements in energy storage efficiency (by both weight and volume) are necessary to improve the competitiveness of electrified vehicles. The ability to quickly and accurately validate and predict long battery life is an important area of development. Likewise, continued cost reduction of LIBs on a $/kWh basis is both challenging and

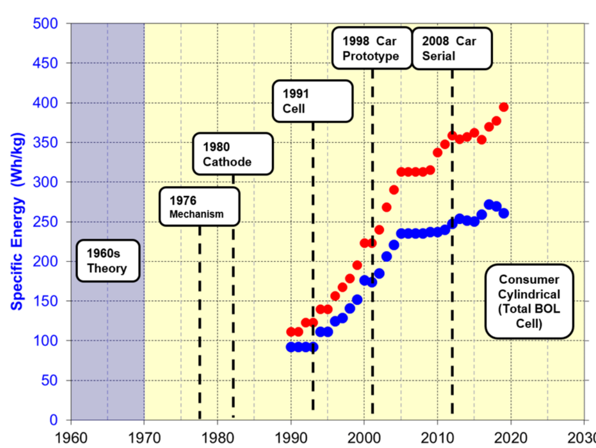


Figure 1. Historical developments in LIB and consumer cylindrical cell-specific energy.[3]

Received: December 14, 2020
Accepted: January 15, 2021
Published: January 29, 2021

https://dx.doi.org/10.1021/acsenergylett.0c02584
ACS Energy Lett. 2021, 6, 621−630

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necessary to increase the opportunity for wide-scale adoption of electric transportation. Automotive LIBs are also expected to meet or exceed the safety performance of existing gasoline-powered internal combustion engine (ICE) vehicles. ICE-powered vehicles have also created a customer expectation of fast refueling, which is a large challenge for LIB technology to match. Although the overall performance of automotive LIBs has improved greatly in recent years, major challenges and opportunities remain.

Vehicle-Based Battery Targets. There are a variety of electrified vehicle types, each with different and unique battery needs that can influence the challenges and opportunity for LIBs. The most commonly known electrified vehicles are those that rely on off-board electrical power for either part (Plug-In Hybrid Electric Vehicles, PHEV) or all (Electric Vehicles, EV) of their traction and on-board power. Electrified vehicles that rely solely on traditional liquid fuels for power are typically sorted by their ability to either electrically power traction drive (Hybrid Electric Vehicle, HEV) or not (Stop/Start Hybrids, S/S).[3]

For each of the electrified vehicle types (EV, PHEV, HEV ,and S/S), the United States Advanced Battery Consortium (USABC), comprising representatives of Stellantis, Ford, and General Motors, has issued a set of battery level technical performance targets.[5−8] Additionally, the USABC has also issued variations on the base EV goals to enable the targeted development of particular use cases, such as the Low-Cost Fast-Charge[9] and Lithium Metal Based EV goals.[10] It should be noted that all USABC goals are written in terms of end-of-life (EOL), battery-pack-level values, whereas researchers and cell developers often highlight beginning-of-life (BOL), cell-level performance.[5] When accounting for energy density lost due to life fade (∼20%), module/pack integration (30−40%),[5] and battery management system and life management uncertainty (5−10%), a factor of 2 reduction in available energy results. Due to the large difference in required energy density depending on the hardware level (cell versus pack), age (BOL versus EOL), and energy level (total versus usable), caution is required when comparing lab-scale or advertised performance versus requirements.

The European Council for Automotive Research and Development (EUCAR, an industry organization in Europe analogous to the USABC) and the New Energy and Industrial Technology Development Organization (NEDO) of the Japanese government have published their own battery system goals.[11,12] When considering the present-day challenges for LIB and future battery chemistries, a comparison against the goals of USABC, EUCAR, and NEDO can be helpful. In Table 1, a side-by-side comparison of EV goals of USABC,[5] EUCAR,[11] and NEDO[12] is shown. As mentioned, the USABC performance goals are for EOL and defined at the battery pack hardware level. The EUCAR goals were published for BOL cells and them converted to EOL pack goals using their published EOL criteria (80% SOH) and pack scale-up cost (+20% of cell cost). The NEDO goals are already in EOL pack format and, except for a currency conversion, are stated as published. A comparison of the USABC, EUCAR, and NEDO specific energy, calendar life, and cycle life goals shows a high degree of alignment. There appears to be global agreement on future energy targets and battery life expectations. For the cost targets, the wide range must be appreciated in terms of the different years of publication and target years: in the case of NEDO, the publication year of their goal is 2017, for EUCAR, the target year is 2030, and for USABC, the target year is 2020. In addition, the relative cost performance of LIBs has rapidly improved in recent years and, as such, cost projections from just a few years ago may no longer be accurate.

The international alignment on energy goals is indicative of a universal need to reduce the weight of vehicles in transportation. Whether a vehicle is powered by fossil fuels or electricity, it is subject to the same basic physics of kinematic weight, aerodynamic drag, tire rolling resistance, and elevation.[3] As a result, the energy consumption rate (Wh/km or L/km) varies linearly with the force required to achieve or maintain a vehicle speed. Although government vehicle drive cycles can vary widely in speed, acceleration, and distance, they are all heavily influenced by these same factors.[3] An illustration of the strong influence of the vehicle weight on consumption is shown in Figure 2. This figure plots the EPA rated consumption of all EVs

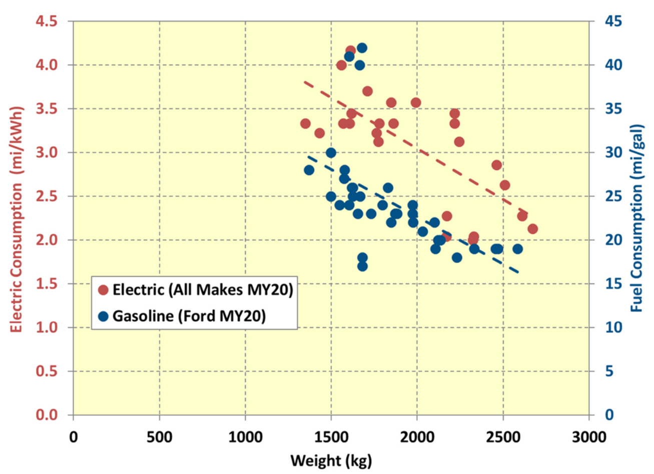

Figure 2. Vehicle energy consumption (electric and fuel) versus weight.[13]

Table 1. International Electric Vehicle Battery Pack Goals at End of Life

group	target year	specific energy (Wh/kg)	specific power (W/kg)	cost ($/kWh)[a]	calendar life (years)	cycles
USABC (ref 5)	2020	235	470	125	10	1000
EUCAR (ref 11)	2030	288	1152	84	vehicle life	24 MWh
NEDO (ref 12)	2020s	250	1500	190	10−15	1000−1500

[a]$1.00 = 1.17€ = 105¥.

on sale in the United States in model year 2020 versus all Ford brand gasoline-powered vehicles. Here we can see that the linear correlation of weight with energy consumption affects both fossil fuel and electric vehicles in a very similar way.

Long-range EVs require heavy battery packs, potentially representing 20−30% of their entire vehicle curb weight.[3] In addition to the beneficial impact on energy consumption, battery weight reduction also has many other related vehicle benefits. For example, reducing the vehicle's weight also would decrease the battery power demands due to the strong correlation to acceleration and deceleration.[3] Vehicle weight reduction could also result in improved battery life by allowing vehicle designers to utilize a smaller state-of-charge (SOC) window. Decreasing vehicle weight also creates the potential for significant cost savings by reducing the quantity of materials needed for manufacture. The challenges and opportunities in battery life and cost are described further in later sections. Modern lithium ion cells have densities ranging from 2 to 3 g/cm^3, and as a result, improvements in battery specific energy (Wh/kg) often translate into greater energy density (Wh/L). When considering the absolute weight of vehicle battery packs, these gains can also translate to significant battery volume reduction.

Challenges and Opportunities in LIB Energy. The higher energy density and specific energy of LIBs have allowed the technology to supplant competing battery chemistries in almost all markets and applications. As a non-aqueous electrolyte system, LIBs are able to maintain cell voltage levels approximately 3 times greater than the incumbent aqueous rechargeable chemistries, such as nickel metal hydride (NiMH) and nickel cadmium (NiCd).[14] However, since their introduction, the basic electrochemical couple of high-energy designs ($Co^{3+/4+}$ and $Ni^{3+/4+}$ versus graphite) has not changed, leading to only marginal improvements in the nominal voltages of LIBs. As a result, almost all of the specific energy improvements shown in Figure 1 are a result of engineering improvements in active material capacity and cell/electrode optimization.[3] However, also as is seen in Figure 1, the rate of performance improvement achieved by these methods has slowed in recent years, posing a significant challenge for future implementations of electrification across all vehicle lines.

To drive future improvements in batteries' specific energy, different materials are needed. The automotive industry has published cathode and anode specific energy goals to help guide the research community with targets (see Figure 3).[12,15] The USABC goals were designed to translate vehicle-level goals[5] to materials-level targets.[15] The EUCAR goals cite previous automotive industry materials requirements studies[16,17] in the context of their vehicle-level targets.[12] For automotive context, the energy storage capability of petrol is also plotted in the figure in green. Gasoline as a liquid fuel has an extremely high energy storage capacity (12.9 kWh/kg), and the value plotted in Figure 3 assumes a best-in-class engine thermal efficiency of 41%, resulting in a practical value of 5.3 kWh/kg.[18] A review of the anode (black font in the figure) goals shows identical gravimetric and similar volumetric goals between the USABC and EUCAR, despite their different publication dates, vehicle-level targets, and goal-setting methodology.[12,15] Existing conventional LIB cell designs were assumed for the cathode (carbon, 0.2 V) and anode (NMC111, 3.8 V) voltage couples. Comparing the cathode (red font in the figure) targets also reveals similar though not identical values for gravimetric and volumetric energy storage. The large degree of materials goal alignment

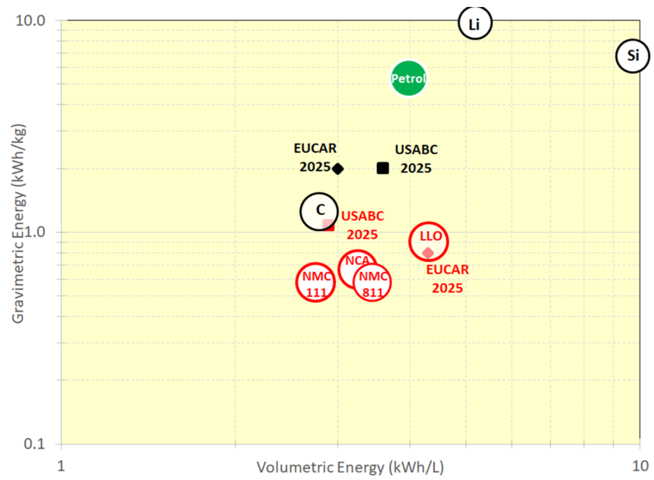

Figure 3. Cathode (red) and anode (black) materials' performance and targets.[15,16]

between two automotive manufacturer-based consortia should inspire confidence in the materials chemistry community of the relevance of these targets.

A review of cathode material performance reveals no practical, mature candidate materials capable of exceeding or meeting future automotive materials targets (see Figure 3). Since initial commercialization, LIB cathodes have been dominated by layered lithium transition metal oxides. The initial cathode, lithium cobalt oxide, has seen its capacity improve steadily over the years as its material stability has improved to tolerate higher and higher charge voltages through coatings, dopants, and processing conditions.[19] In parallel, a variety of different metals such as nickel, manganese, and aluminum have been successfully incorporated in the same crystal structure as cobaltate, enabling reduced cost while balancing available energy.[20] One such derivative, the lithium-rich layered oxide (LLO), is capable of meeting the EUCAR cathode targets; however, since its discovery, this material has suffered from structural stability and voltage fade issues that have limited its utility.[21] The lack of a viable future cathode material to continue LIB energy improvement provides a significant challenge to the continued development of the field.

The LIB anode market has historically been dominated by carbon anodes, which are incapable of meeting future energy targets (Figure 3). The two most widely researched anode materials, lithium and silicon, are both capable of meeting the energy targets, depending on how they are used. For purposes of comparison, Figure 3 displays lithium energy assuming a 50% excess and silicon assuming a 2000 mAh/g capacity. The technical challenges involved in utilizing lithium metal in automotive applications are large and require significant further engineering at the present time.[22] In the near term, silicon is the more viable advanced anode material; carbon/silicon blends have been used commercially for years.[23] As can be seen in Figure 3, silicon has almost an order of magnitude greater gravimetric energy than that of carbon. However, pure silicon has practical limitations due to the large volume changes during cycling and resulting lithium loss due to continuous solid−electrolyte interphase (SEI) formation and particle isolation during cycling. Fortunately, even at a capacity of 2000 mAh/g, less than half that of pure silicon, this material has potential to meet the industrial targets. As a result, silicon as a system can afford to tolerate additional weight and volume from strategies

https://dx.doi.org/10.1021/acsenergylett.0c02584
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that accommodate the lithium storage capacity of silicon while maintaining the electrode structure and stable interfacial chemistry.[24,25]

Challenges and Opportunities in LIB Life. Battery durability is a critical requirement for the continued proliferation of EVs. Many vehicle original equipment manufacturers (OEMs) offer warranties on EV battery packs that guarantee battery replacement if capacity degrades beyond a defined threshold level. This creates engineering challenges during the battery development process, since warranty periods extend for many years and hundreds of thousands of miles, yet batteries must be validated on the order of months for timely implementation of state-of-the-art power and energy density, among other attributes. Additionally, sufficiently precise test equipment is required to avoid propagating uncertainty in derived quantities such as battery power and energy.[26]

Typically, accelerated testing is able to ascertain the long-term battery degradation rate versus operating conditions, if not fully meeting EOL discharge throughput and/or power and energy retention targets, in a matter of several months.[27] There are then two remaining challenges: (1) to select a sufficient set of tests to fully characterize the degradation rate across the operating conditions the battery is expected to encounter during customer use, and (2) to confirm that these degradation rates are not significantly accelerated at some later stage in life, deemed a "rollover" degradation process. The former challenge may be addressed by a combination of intelligent selection of state of charge and temperature windows known to be relevant to the application, and use of advanced diagnostics to equate early-life results to long-term degradation rates. The latter challenge requires either continuing testing until fully reaching EOL conditions, or obtaining a deep understanding of the degradation processes that are occurring and using mathematical models to simulate their contributions to degradation as the battery progresses to EOL. Figure 4 demonstrates the use of Coulombic efficiency testing[28] to rapidly map the effect of current rate on cycle life for a power cell of 5 Ah capacity at 25 °C.

From the trend of Coulombic efficiency versus current, it can be deduced that there are several conditions that would produce results with little differentiation when comparing long-term degradation performance. Using the Coulombic efficiency performance as a guide, the design of experiments could be reduced from several cells to just two bookends, with Coulombic efficiency data used to interpolate the trend between these bookends.

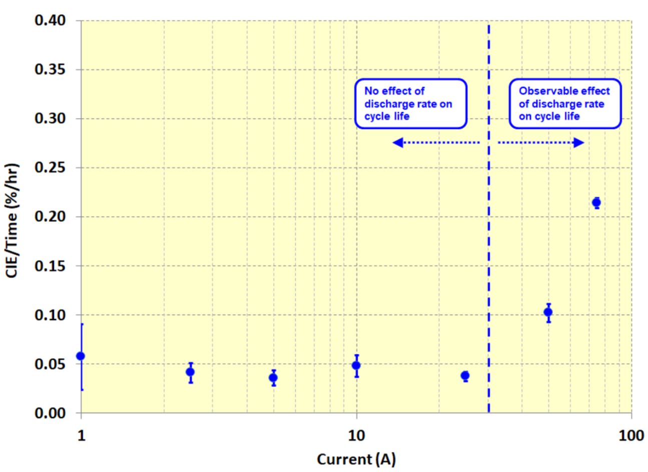

Figure 4. Demonstration of Coulombic inefficiency rate (1 minus Coulombic efficiency, per unit time) versus current. Several conditions will produce similar long-term degradation rates, until a critical threshold of current is exceeded.[28]

In terms of confirming whether degradation rates accelerate as the battery ages, Dahn and others have developed a framework[29−31] for predicting the rollover of graphite-NMC cells that considers two main degradation processes to represent several side reactions: (1) consumption of cyclable lithium and (2) shuttle mechanisms that consume electrolyte solvent. For the graphite-NMC pairing cycled at low charge rates, this set of mechanisms is sufficiently broad to both confirm the long-term degradation rates with high-precision coulometry and verify that kinetic limitations at the cathode can accelerate solvent decomposition. Separately, they have quantified lithium plating using Coulombic efficiency measurements across multiple rates and temperatures.[32]

From an automotive standpoint, this framework is robust for cells with large capacity relative to their power requirements that are comprised of graphite-NMC couples. However, important challenges remain in battery life prediction when cycling cells aggressively and continuously, such as when vehicles tow or haul heavy loads over long distances with multiple fast-charge events followed by extended highway driving or are driven autonomously for durations well beyond those of a typical retail customer. To provide accurate life predictions under these circumstances, it is likely that models based on three-dimensional electrochemistry will be needed to accurately represent the geometry of cell and pack designs and local degradation effects, such as lithium plating.[33−35] These models must quantify thermal and mechanical inhomogeneity inherent to the design of a given pack and apply the above framework to ascertain whether rollover occurs in any cell of an aggressively cycled battery pack. Machine learning is an important supplement to physics-based models in cases where large amounts of data are available or when rapid model execution is required to support optimization modeling.[36,37]

As the automotive industry looks toward next-generation anode and cathode chemistries, important challenges remain in battery life prediction. Although silicon compounds are presently being utilized at low concentrations in graphite anodes, increasing their fraction to improve energy density is likely to cause greater mechanical stability issues. Battery failure, even in the present generation chemistries, may be caused by excessive cell swelling, leading to strains in pack components that compromise the integrity of the pack. Electrically disconnected zones within an electrode are more likely to occur in electrodes with high rates of volume change during cycling, which will lead to greater resistance rise. Though high-precision coulometry studies have been conducted on cells where swelling is present,[38] neither swelling nor resistance rise has been tracked or predicted by high-precision coulometry to date, and large changes in resistance over life complicate interpretation of coulometry results. Therefore, analogous, rapid diagnostic methods for these alternative failure modes are needed. Likewise, high-precision coulometry has been applied to metallic lithium anodes,[39] but these will require novel characterization methods because dendrite formation is a critical degradation mechanism. In this case, the issue is not consumption of cyclable lithium but rather maintaining the morphology and integrity of the lithium metal surface as it is stripped and plated during battery cycling. The formation of

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ACS Energy Lett. 2021, 6, 621−630

high-surface-area dendrites can increase the rate of SEI formation and electrolyte depletion, which would be observable in Coulombic efficiency measurements. However, the possibility that dendrite formation can create soft short-circuit connections between the anode and cathode could lead to premature battery failure via excessive self-discharge, and it is unlikely that efficiency measurements alone are capable of quantifying this outcome.

Challenges and Opportunities in LIB Cost. The high cost of battery electric vehicles (BEVs) when compared to internal combustion engine vehicles (ICEVs) is a key challenge for their acceptance in the marketplace. The battery pack is often the single most expensive component in an EV due to high material costs and engineering complexity. However, over the past decade, tremendous investment in battery manufacturing capacity and the reduction of cobalt in cathodes has driven an 80% decrease in cost (see Figure 5).[40,41] As this trend continues,

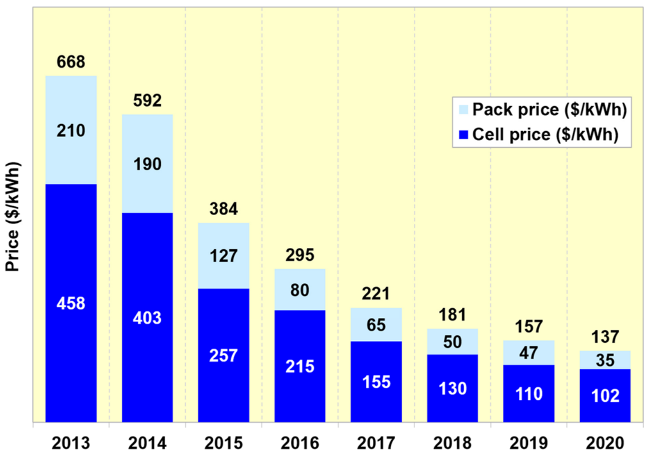

Figure 5. Survey of lithium-ion cell and pack integration prices for automotive years 2013−2020, adjusted to real 2020 \$/kWh. From ref 40 with permission of BNEF.

total cost of ownership parity between BEVs and ICEVs is closer than ever. However, purchase price parity remains elusive, and thus further cost reduction is needed to improve marketplace acceptance.

Opportunities for cost reduction exist across the entire LIB value chain and generally fall into the following categories: manufacturing process improvements, material innovations, performance improvements that reduce integration requirements, and closing the value-chain loop. The merits of a cost reduction opportunity should be evaluated based on the ability to decrease cost on a per-kWh basis, and research efforts should take account for upstream requirements and downstream effects. Use of simulation tools and modeling are critical to validate those effects.

A great deal of the cost reduction already realized is due to improvements in the manufacturing process. Process innovations that further reduce energy, time, volume, and capital all stand to make an impact.[41−43] Material production for anodes and cathodes still relies on batched, high-temperature processes such as calcination for cathodes and graphitization for synthetic graphite. Cell assembly processes involve solvents, binders, and additives that, if eliminated or reduced, could yield further cost reductions. Lastly, the SEI formation process takes days or weeks and requires a large storage volume for staging; streamlining of this process would be a boon to production.[44]

Material selection and innovation is another area where cost reduction opportunities exist. Much of the cost reduction illustrated in Figure 5 is associated with swapping 80% of cobalt with nickel, such as in NMC811 vs LCO. Still, further reduction and ultimately elimination of cobalt and nickel are warranted. These metals are costly and may encounter significant supply chain challenges.[45] Advanced electrodes that increase volumetric and specific energy density can decrease per-kWh costs, even if they represent higher per-kg costs. Furthermore, beyond the opportunities described earlier in this paper, researchers should keep material-level and system-level trade-offs in mind.

Another opportunity for reducing cost is to improve performance attributes that affect pack and system integration. For example, customer-accessible energy and battery-nameplate energy often differ by up to 20%. By improving cycle and calendar lifetime, depth of discharge limits can be widened, which would reduce cost per usable kWh. Additionally, increasing the thermal operation window would reduce system thermal management requirements. Lastly, integrated safety features have the ability to lessen engineering requirements at the pack and structural level, reducing engineering cost.[46]

Closing the value chain loop with recycling also offers an additional cost reduction opportunity. Rapid adoption of EVs will create a wave of EOL batteries in the next decade, and by bringing the EOL battery materials back into the value chain, the raw material economics could shift. At the moment, economical pyro- and hydrometallurgical methods are still being developed. Ultimately, direct recycling of cathodes and anodes offers the most promise, eliminating both the costly geological extraction and initial material processing steps.[47]

Ultimately, direct recycling of cathodes and anodes offers the most promise, eliminating both the costly geological extraction and initial material processing steps.

In addition to manufacturing cost reduction opportunities for LIBs on the horizon, indirect economic benefits to customers may be an important factor to adoption. Fueling and maintenance cost advantages of EVs suggest that total cost of ownership parity will come sooner than expected. For example, new all-electric commercial vehicles are expected to have 40% lower scheduled maintenance costs when compared to gasoline models,[47] and depending on location, electricity as a fuel can cost up to 60% less than gasoline, representing savings of thousands of dollars over the lifetime of the vehicle.[48]

While BEVs have favorable cost traits over ICEVs, the cost of emitting CO_2 and other pollutants is still largely externalized. However, one can still assess the relative environmental benefits of BEVs. Though BEVs do not create point-source emissions, those associated with manufacturing and those associated with fueling should not be ignored. Regarding the former, past work has shown that the manufacturing greenhouse-gas footprint of a BEV is larger than that of a comparable ICEV.[49] Regarding the latter, emissions vary based on the regional electricity supply mix and are usually much better than those of ICEVs.[50,51] It is important to recognize that operational emissions from BEVs are dynamically coupled with the electricity supply mix and are expected to improve as more renewable energy sources come online. In contrast, operational emissions from ICEVs are fixed

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at best and usually deteriorate over the lifetime of the vehicle; this underlines the primary value proposition for BEVs.

Challenges and Opportunities in LIB Safety. Regardless of the energy source for propulsion (gas or electric), there is the same expectation of a high degree of safety in the automotive sector. As a result, LIB-electrified vehicles are expected to meet or exceed the level of safety provided by fossil-fuel-powered vehicles. As a result, vehicle makers provide multiple layers of safety protection, ranging from the cell to the module, pack, and vehicle.[52] In addition, a range of electrical, thermal, and mechanical control and monitoring systems are typically employed surrounding the electrochemical cell.

The widespread adoption of LIB technology has promoted significant safety research in automotive applications on thermal,[54,55] mechanical,[56,57] electrical,[58,59] and systems bases.[53,60] Additionally, a range of battery-safety-related government regulations have been issued from the UN,[61,62] ECE,[63] China,[64] and South Korea[65] to name a few. The result has been a comprehensive study and understanding of the safety considerations in a LIB.

The reaction of a battery during an abuse test is categorized on a scale developed by EUCAR[66] ranging from 0 (no response) to 7 (explosion). For example, Figure 6 describes the boundary conditions around a EUAR 5 (fire) rating under crush, overcharge, and short-circuit abusive testing for a variety of LIB hardware levels.[53] The EUCAR rating system is an effective means to summarize complicated battery abuse responses. Although this rating system is not perfect, its great utility to the research community comes from its ubiquity and simplicity.

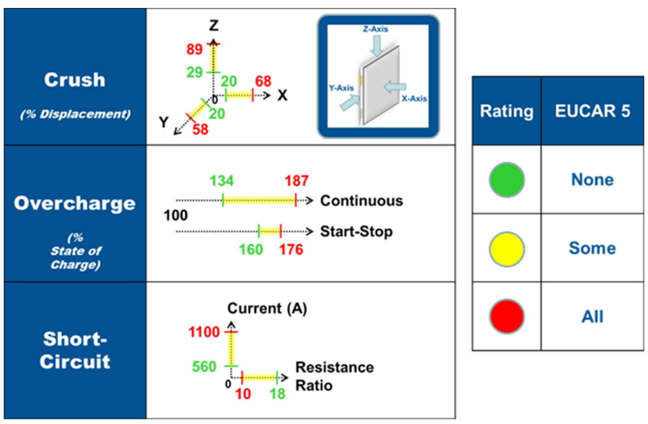

Figure 6. Boundary conditions of EUAR 5 response under crush, overcharge, and short-circuit.[53]

The existing system of layered safety protections at the vehicle, pack, and module levels, all supporting the individual cell, has shown itself to be very effective, with all but a few incidents to-date. The main sensor systems employed to determine the battery's condition are voltage, current, and temperature, measured at either the cell, module, or pack level. A potential future research opportunity would be to develop alternative sensor techniques such as pressure, fiber-optics, and acoustics.[67−69] These additional sensors may be able to provide novel information regarding the LIB's safety conditions and provide a cost, weight, and volume reduction opportunity compared to current solutions. In this context, a future challenge for LIB researchers will be continued energy density improvements while also maintaining the current high level of safety.

Challenges and Opportunities in LIB Fast Charge. Charging time is a key concern for customers who make regular long-distance trips that exceed the total range of their battery. Direct current fast charging (DCFC) allows customers to charge to 80% SOC at rates up to 350 kW, delivering a couple hundred miles of range in about 40 min. However, the ability to charge faster, practically approaching parity with ICEVs, is still desired, particularly for drivers who lack access to overnight or workplace charging. The U.S. DOE's long-term target for extreme fast charging is to deliver 200 miles in 7.5 min of charging.[70] A more near-term fast charge target from the USABC for 2023 calendar year is goal of Δ80% SOC in 15 min.[9]

LIBs offer high energy in part because of the low electrochemical potential for lithium ion insertion in carbon, which maximizes the available cell energy. Unfortunately, this places the negative electrode potential close to that of lithium metal, which raises the risk of unintended lithium plating. This in turn leads to irreversible capacity loss, reduced performance, and increased risk of short-circuiting and thermal runaway.[71] This is the primary factor limiting adoption of faster charging rates.

There are several opportunities at the cell and battery levels for improved fast charging, such as electrode engineering, new electrode chemistry, and thermal management. Electrode engineering can lead to optimized thicknesses and porosities, thereby reducing voltage polarization. This in turn helps stabilize the voltage to allow for lithium intercalation. Electrode engineering is able to modify the power-to-energy ratio (P/E) of cells across a substantial range, even considering the same basic chemistry. For example, Figure 7 shows several production automotive battery cells plotted in terms of their energy density and estimated peak charging power of a hypothetical 100 kWh battery pack, based on the cell P/E.[3]

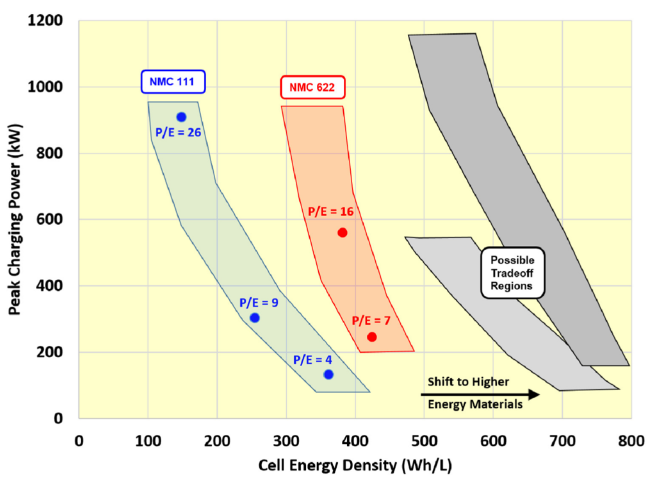

Figure 7. Estimated battery peak charging power versus cell energy density for groupings of similar active material chemistry. All data points feature graphitic carbon anodes paired with the noted cathode chemistry.

Based on the charge rates defined by the highlighted region for each cathode chemistry (Figure 7), it is clear that high power can be achieved through engineering modifications to the electrode structure while using the same active material. As a result, researchers should place greater emphasis on finding high-energy materials while leaving power capability as a secondary goal that can be achieved through cell and electrode engineering. A cell energy density of 700−800 Wh/L can be used to create a vehicle with exceptionally high range, but customers may prefer the moderately high range offered by cells in the 600−700 Wh/L range if they can also deliver fast charge rates that approach gasoline parity. To increase the fast charge rate by 50 kW requires a cell energy density penalty between 1.5% and 4% based on prior cell designs. This trade-off correlation will apply to future cell designs if they have similar versatility to today's designs and are primarily composed of intercalation materials mixed into agglomerate coatings, rather than fundamentally different technology such as solid-state electrolytes with metal electrodes. Furthermore, this does not include any allowance for the enhanced thermal management requirements of charging at faster rates.

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At the system level, thermal management remains paramount. Cooling strategies must be continually improved to keep pace with the demand for greater charging rates. The limiting factor in many present designs is the surface area available for cooling. Due to requirements to maximize pack energy density, OEMs are reluctant to expand their surface area that defines the heat removal pathway. As material energy density increases, some volume within the pack could be repurposed to support greater heat removal capability. Additionally, with greater volume in the pack dedicated to thermal management, more advanced strategies could enable extreme fast charging with existing materials using brief excursions to high temperatures.[72] However, it is critical that high-temperature durability be improved, including capacity loss, resistance rise, and gas generation, if high-temperature charging is expected regularly throughout life.

Future Challenges and Opportunities for LIBs. The lithium ion battery (LIB) has evolved significantly since its first market introduction in 1991, enabling its specific energy content to almost triple (see Figure 1). The life of LIBs has more than tripled, providing the ability to meet most automotive calendar and cycle life requirements, albeit by requiring advanced control systems and maintaining cell energy reserves. While achieving these performance increases, the cost of LIBs has also been reduced by almost 2 orders of magnitude, with sharp declines in recent years (see Figure 5). These improvements in LIB technology have been achieved while maintaining the high degree of safety required of automotive products. In recent years, a new automotive goal of fast charging has evolved as practical pack sizes have increased, resulting in steadily increasing battery system charge rates.

Based on the current progress and materials chemistry research efforts, LIBs will be able to achieve ~350 Wh/kg total cell energy at the beginning of life. Unfortunately, this energy content is not sufficient to meet the automotive energy targets once this performance is converted to usable, pack end-of-life values.[3,5] This looming challenge led to the initiation of various Beyond Lithium Ion (BLI) research initiatives approximately a decade ago. The three main automotive-relevant technologies pursued as a part of initial BLI efforts were lithium air, lithium sulfur, and lithium metal. Lithium air has struggled with life and energy efficiency challenges that have limited its significance to the research lab. Sulfur chemistries have shown greater promise than lithium air, finding use in niche market applications such as unmanned aerial vehicles; however, they too have been limited by life and volume concerns, given their low densities. The BLI technology that has advanced the furthest in the past decade has been the lithium metal system. The progress and potential impact of lithium metal cells are underscored by the recent issuance of automotive lithium metal cell targets from the USABC.[10]

Novel liquid electrolytes compatible with lithium metal have shown significant progress recently by adapting the existing lithium ion cell design.[73,74] The recent discovery of several solid lithium superionic conductors has also renewed the viability of lithium metal solid-state batteries (LMSSBs).[75,76] Most proposed designs for LMSSBs also adopt traditional LIB cathode and cell design parameters. The challenge of developing long-lasting lithium metal cells will require an improved understanding of lithium mechanics[77,78] and the lithium/electrolyte interface, be it liquid or solid.[79] Regardless of whether solid or liquid electrolyte systems are used, the basic design of those lithium metal cells will inherit the majority of the engineering and materials chemistry improvements of the LIB system.

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Notes

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The authors declare no competing financial interest.

https://dx.doi.org/10.1021/acsenergylett.0c02584
ACS Energy Lett. 2021, 6, 621−630

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■ REFERENCES

(1) Blomgren, G. E. The Development and Future of Lithium Ion Batteries. *J. Electrochem. Soc.* **2017**, *164* (1), A5019−A5025.

(2) Tarascon, J. M.; Armand, M. Issues and challenges facing rechargeable lithium batteries. *Nature* **2001**, *414* (6861), 359−367.

(3) Masias, A. Lithium Ion Battery Design for Transportation. In *Behavior of Lithium-Ion Batteries in Electric Vehicles: Battery Health, Performance, Safety, and Cost*; Pistoia, G., Liaw, B., Eds.; Springer International Publishing, 2018; pp 1−34.

(4) Driscoll, E. H.; Hayward, E. C.; Patchett, R.; Anderson, P. A.; Slater, P. R. The Building Blocks of Battery Technology: Using Modified Tower Block Game Sets to Explain and Aid the Understanding of Rechargeable Li-Ion Batteries. *J. Chem. Educ.* **2020**, *97*, 2231−2237.

(5) United States Advanced Battery Consortium. USABC Goals for Advanced High-Performance Batteries for Electric Vehicle (EV) Applications, 2019. Retrieved from http://www.uscar.org/commands/files_download.php?files_id=364 (accessed Sept 9, 2020).

(6) United States Advanced Battery Consortium USABC Goals for Advanced Batteries for PHEVs for FY 2018 to 2020 Commercialization, 2018. Retrieved from http://www.uscar.org/commands/files_download.php?files_id=436 (accessed Nov 12, 2020).

(7) FreedomCAR Battery Test Manual for Power-Assist Hybrid Electric Vehicles, DOE/ID-11069, October 2003. Retrieved from http://www.uscar.org/commands/files_download.php?files_id=58 (accessed Nov 12, 2020).

(8) United States Advanced Battery Consortium (USABC). Development of Advanced High-Performance Batteries for 12V Start Stop Vehicle Applications, 2020. Retrieved from http://www.uscar.org/commands/files_download.php?files_id=347 (accessed Nov 12, 2020).

(9) United States Advanced Battery Consortium (USABC). Development of Advanced Low-Cost/Fast-Charge (LC/FC) Batteries for Electric Vehicle (EV) Applications, 2020. Retrieved from https://www.uscar.org/commands/files_download.php?files_id=477 (accessed Jan 7, 2021).

(10) United States Advanced Battery Consortium (USABC). Development of Lithium Electrode Based Cell and Manufacturing for Automotive Traction Applications: Appendix A - Lithium Electrode Based Cell Goals, April 2020. Retrieved from https://www.uscar.org/commands/files_download.php?files_id=509 (accessed Nov 13, 2020).

(11) Nagai, T.. The Japanese policy and NEDO activity for future mobility, New Energy and Industrial Development Organization (NEDO), Dec 13, 2017. Retrieved from https://www.nedo.go.jp/content/100873093.pdf (accessed Sept 29, 2020).

(12) European Council for Automotive R&D (EUCAR). Battery requirements for future automotive applications, July 2019. Retrieved from https://eucar.be/wp-content/uploads/2019/08/20190710-EG-BEV-FCEV-Battery-requirements-FINAL.pdf (accessed Sept 29, 2020).

(13) The Official U.S. Government Source for Fuel Economy Information, U.S. Environmental Protection Agency. Retrieved from www.fueleconomy.gov (accessed Jan 6, 2021).

(14) Winter, M.; Barnett, B.; Xu, K. Before Li Ion Batteries. *Chem. Rev.* **2018**, *118*, 11433−11456.

(15) United States Advanced Battery Consortium (USABC). Development of Active Materials for Cells to be Used in Automotive Traction Applications, Sept 2020. Retrieved from https://www.uscar.org/commands/files_download.php?files_id=455 (accessed Oct 23, 2020).

(16) Andre, D.; Kim, S.; Lamp, P.; Lux, S. F.; Maglia, F.; Paschos, O.; Stiaszny, B. Future generations of cathode materials: an automotive industry perspective. *J. Mater. Chem. A* **2015**, *3*, 6709−6732.

(17) Andre, D.; Hain, H.; Lamp, P.; Maglia, F.; Stiaszny, B. Future high-energy density anode materials from an automotive application perspective. *J. Mater. Chem. A* **2017**, *5*, 17174−17198.

(18) Yamaji, K.; Tomimatsu, M.; Takagi, I.; Higuchi, A.; Yoshida, T.; Murase, E. New 2.0L I4 Gasoline Direct Injection Engine with Toyota New Global Architecture Concept, SAE Technical Paper 2018-01-0370, 2018. DOI: 10.4271/2018-01-0370.

(19) Zhang, J.; Li, Q.; Ouyang, C.; Yu, X.; Ge, M.; Huang, X.; Hu, E.; Ma, C.; Li, S.; Xiao, R.; Yang, W.; Chu, Y.; Liu, Y.; Yu, H.; Yang, X.; Huang, X.; Chen, L.; Li, H. Trace doping of multiple elements enables stable battery cycling of $LiCoO_2$ at 4.6V. *Nature Energy* **2019**, *4*, 594−603.

(20) Myung, S.; Maglia, F.; Park, K.; Yoon, C. S.; Lamp, P.; Kim, S.; Sun, Y. Nickel-Rich Layered Cathode Materials for Automotive Batteries: Achievements and Perspectives. *ACS Energy Letters* **2017**, *2*, 196−223.

(21) Thackeray, M. M.; Kang, S.; Johnson, C. S.; Vaughey, J. T.; Benedek, R.; Hackney, S. A. Li_2MnO_3-stabilized $LiMO_2$ (M = Mn, Ni, Co) electrodes for lithium-ion batteries. *J. Mater. Chem.* **2007**, *17*, 3112−3125.

(22) Chen, S.; Dai, F.; Cai, M. Opportunities and Challenges of High-Energy Lithium Metal Batteries for Electric Vehicle Applications. *ACS Energy Letters* **2020**, *5*, 3140−3151.

(23) Choi, J. W.; Aurbach, D. Promise and reality of post-lithium-ion batteries with high energy densities. *Nature Reviews Materials* **2016**, *1*, 16013.

(24) Shen, X.; Tian, Z.; Fan, R.; Shao, L.; Zhang, D.; Cao, G.; Kou, L.; Bai, Z. Research progress on silicon/carbon composite anode materials for lithium-ion battery. *J. Energy Chem.* **2018**, *27*, 1067−1090.

(25) Salah, M.; Murphy, P.; Hall, C.; Francis, C.; Kerr, R.; Fabretto, M. Pure silicon thin-film anodes for lithium-ion batteries: A review. *J. Power Sources* **2019**, *414*, 48−67.

(26) Wold, J.; Marcicki, J.; Masias, A. Derived Quantities Uncertainty Propagation in High Precision Battery Testing. *J. Electrochem. Soc.* **2017**, *164* (9), A2131−A2137.

(27) Belt, J.; Utgikar, V.; Bloom, L. Calendar and PHEV cycle life aging of high-energy, lithium-ion cells containing blended spinel and layered-oxide cathodes. *J. Power Sources* **2011**, *196*, 10213−10221.

(28) Marcicki, J.; Masias, A.; Coupar, T.; Elsayed, O. Quantitative Life Prediction Using High Precision Coulometry with Automotive Current Rates, 229th Electrochemical Society Meeting, May 29−June 2, 2016.

(29) Smith, A. J.; Burns, J. C.; Xiong, D.; Dahn, J. R. Interpreting High Precision Coulometry Results on Li-Ion Cells. *J. Electrochem. Soc.* **2011**, *158* (10), A1136−A1142.

(30) Burns, J. C.; Kassam, A.; Sinha, N. N.; Downie, L. E.; Solnickova, L.; Way, B. M.; Dahn, J. R. Predicting and Extending the Lifetime of Li-Ion Batteries. *J. Electrochem. Soc.* **2013**, *160* (9), A1451−A1456.

(31) Ma, X.; Harlow, J.; Li, J.; Ma, L.; Hall, D.; Buteau, S.; Genovese, M.; Cormier, M.; Dahn, J. R. Hindering Rollover Failure of $Li[Ni_{0.5}Mn_{0.3}Co_{0.2}]O_2$/Graphite Pouch Cells during Long-Term Cycling. *J. Electrochem. Soc.* **2019**, *166* (4), A711−A724.

(32) Burns, J. C.; Stevens, D. A.; Dahn, J. R. In-Situ Detection of Lithium Plating Using High Precision Coulometry. *J. Electrochem. Soc.* **2015**, *162* (6), A959−A964.

(33) Bach, T.; Schuster, S.; Fleder, E.; Muller, J.; Brand, M.; Lorrmann, H.; Jossen, A.; Sextl, G. Nonlinear aging of cylindrical lithium-ion cells linked to heterogeneous compression. *J. Energy Storage* **2016**, *5*, 212−223.

(34) Sauerteig, D.; Hanselmann, N.; Arzberger, A.; Reinshagen, H.; Ivanov, S.; Bund, A. Electrochemical-mechanical coupled modeling and parameterization of swelling and ionic transport in lithium-ion batteries. *J. Power Sources* **2018**, *378*, 235−247.

(35) Spingler, F.; Wittmann, W.; Sturm, J.; Rieger, B.; Jossen, A. Optimum fast charging of lithium-ion pouch cells based on local volume expansion criteria. *J. Power Sources* **2018**, *393*, 152−160.

(36) Severson, K. A.; et al. Data-driven prediction of battery cycle life before capacity degradation. *Nature Energy* **2019**, *4*, 383−391.

(37) Attia, P.; et al. Closed-loop optimization of extreme fast charging for batteries using machine learning. *Nature* **2020**, *578*, 397−402.

(38) Burns, J. C.; Sinha, N. N.; Jain, G.; Ye, H.; VanElzen, C. M.; Scott, E.; Xiao, A.; Lamanna, W. M.; Dahn, J. R. The Impact of Intentionally Added Water to the Electrolyte of Li-Ion Cells. *J. Electrochem. Soc.* **2013**, *160* (11), A2281−A2287.

(39) Genovese, M.; Louli, A. J.; Weber, R.; Hames, S.; Dahn, J. R. Measuring the Coulombic Efficiency of Lithium Metal Cycling in Anode-Free Lithium Metal Batteries. *J. Electrochem. Soc.* **2018**, *165* (14), A3321−A3325.

(40) Frith, J. *2020 Lithium Ion Battery Price Survey*; Bloomberg New Energy Finance, New York, 2020.

(41) Duffner, F.; Wentker, M.; Greenwood, M.; Leker, J. Battery cost modelling: A review and directions for future research. *Renewable Sustainable Energy Rev.* **2020**, *127*, 109872.

(42) Ahmed, S.; Nelson, P.; Gallagher, K.; Susarla, N.; Dees, D. Cost and energy demand of producing nickel manganese cobalt cathode material for lithium ion batteries. *J. Power Sources* **2017**, *342*, 733−740.

(43) Schmuch, R.; Wagner, R.; Horpel, G.; Placke, T.; Winter, M. Performance and cost of materials for lithium-based rechargeable automotive batteries. *Nature Energy* **2018**, *3*, 267−278.

(44) Wood, D.; Wood, M.; Li, J.; Du, Z.; Ruther, R.; Hays, K.; Muralidharan, N.; Geng, L.; Mao, C.; Belharouak, I. Perspectives on the relationship between materials chemistry and roll-to-roll manufacturing for high-energy lithium-ion batteries. *Energy Storage Materials* **2020**, *29*, 254−265.

(45) Turcheniuk, K.; Bondarev, D.; Amatucci, G. G.; Yushin, G. Battery materials for low-cost electric transportation. *Mater. Today* **2020**, DOI: 10.1016/j.mattod.2020.09.027.

(46) Ye, Y.; et al. Ultralight and fire-extinguishing current collectors for high-energy and high-safety lithium-ion batteries. *Nat. Energy* **2020**, *5*, 786−793.

(47) Chen, M.; Ma, X.; Chen, B.; Arsenault, R.; Karlson, P.; Simon, N.; Wang, Y. Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries. *Joule* **2019**, *3*, 2622−2646.

(48) Borlaug, B.; Salisbury, S.; Gerdes, M.; Muratori, M. Levelized Cost of Charging Electric Vehicles in the United States. *Joule* **2020**, *4*, 1470−1485.

(49) Kim, H. C.; Wallington, T. J.; Arsenault, R.; Bae, C.; Ahn, S.; Lee, J.; et al. Cradle-to-Gate Emissions from a Commercial Electric Vehicle Li-Ion Battery: A Comparative Analysis. *Environ. Sci. Technol.* **2016**, *50*, 7715−7722.

(50) Wu, D.; et al. Regional Heterogeneity in the Emissions Benefits of Electrified and Lightweighted Light-Duty Vehicles. *Environ. Sci. Technol.* **2019**, *53* (18), 10560−10570.

(51) Tong, F.; Azevedo, I. What are the best combinations of fuel-vehicle technologies to mitigate climate change and air pollution effects across the United States? *Environ. Res. Lett.* **2020**, *15*, 074046.

(52) Ressler, G. Application of System Safety Engineering Processes to Advanced Battery Safety. *SAE International Journal of Engines* **2011**, *4*, 1921−1927.

(53) Masias, A. Ford Safety Performance of Rechargeable Energy Storage Systems, Report No. DOT HS 812 756; National Highway Traffic Safety Administration: Washington, DC, July 2019; https://rosap.ntl.bts.gov/view/dot/41840.

(54) Liu, X.; Stoliarov, S.; Denlinger, M.; Masias, A.; Snyder, K. Comprehensive calorimetry of the thermally-induced failure of a lithium ion battery. *J. Power Sources* **2015**, *280*, 516−525.

(55) Liu, X.; Wu, Z.; Stoliarov, S.; Denlinger, M.; Masias, A.; Snyder, K. A thermos-kinetic model of thermally-induced failure of a lithium ion battery: development, validation and application. *J. Electrochem. Soc.* **2018**, *165* (11), A2909.

(56) Sahraei, E.; Meier, J.; Wierzbicki, T. Characterizing and modeling mechanical properties and onset of short circuit for three types of lithium-ion pouch cells. *J. Power Sources* **2014**, *247*, 503−516.

(57) Sahraei, E.; Hill, R.; Wierzbicki, T. Calibration and finite element simulation of pouch lithium-ion batteries for mechanical integrity. *J. Power Sources* **2012**, *201*, 307−321.

(58) Xia, B.; Chen, Z.; Mi, C.; Robert, B. External Short Circuit Fault Diagonsis for Lithium-Ion Batteries, 2014 IEEE Transportation Electrification Conference, Dearborn, MI, June 15−18, 2014.

(59) Xia, B.; Mi, C.; Chen, Z.; Robert, B. Multiple Cell Lithium-Ion Battery System Electric Fault Online Diagnostics, 2015 IEEE Transportation Electrification Conference and Expo, Dearborn, MI, June 14−17, 2015.

(60) Deng, J.; Bae, C.; Marcicki, J.; Masias, A.; Miller, T. Safety modeling and testing of lithium-ion batteries in electrified vehicles. *Nature Energy* **2018**, 3 (4), 261−266.

(61) Recommendations on the Transport of Dangerous Goods. Manual of Tests and Criteria, 7th rev. ed.; Document No. ST/SG/AC.10/11/Rev.7; United Nations: New York and Geneva, 2019; Section 38.3.

(62) Global Technical Regulation on Electric Vehicle Safety (EVS), Addendum 20: Global Technical Regulation No. 20, ECE/TRANS/180/Add.20, May 3, 2018;United Nations: New York and Geneva, 2018. Retrieved from https://www.unece.org/fileadmin/DAM/trans/main/wp29/wp29wgs/wp29gen/wp29registry/ECE-TRANS-180a20e.pdf (accessed Oct 17, 2020).

(63) Uniform provisions concerning the approval of vehicles with regard to specific requirements for the electric power train, Addendum 99: Regulation No. 100, rev. 2, Aug 12, 2013; United Nations: New York and Geneva, 2013. Retrieved from https://www.unece.org/fileadmin/DAM/trans/main/wp29/wp29regs/2013/R100r2e.pdf (accessed July 24, 2020).

(64) Electric vehicles traction battery safety requirements, GB 38031-2020; National Standard of the People's Republic of China, May 12, 2020.

(65) Test Procedures for Traction Battery Safety, KMVSS Annex 1 - Part 48; Korean Motor Vehicle Safety Standards, 2009.

(66) Doughty, D. H.; Crafts, C. C. FreedomCAR: Electrical Energy Storage System Abuse Test Manual for Electrical and Hybrid Electric Vehicle Applications, SAND2005-3123; Sandia National Laboratories: Albuquerque, NM, August 2006. Retrieved from https://prod-ng.sandia.gov/techlib-noauth/access-control.cgi/2005/053123.pdf (accessed Nov 13, 2020).

(67) Oh, K.; Epureanu, B. I. A phenomenological force model of Li-ion battery packs for enchanced performance and health management. *J. Power Sources* **2017**, *365*, 220−229.

(68) Ganguli, A.; Saha, B.; Raghavan, A.; Kiesel, P.; Arakaki, K.; Schuh, A.; Schwartz, J.; Hegyi, A.; Sommer, L. W.; Lochbaum, A.; Sahu, S.; Alamgir, M. Embedded fiber-optic sensing for accurate internal monitoring of cell state in advanced battery management systems part 2: Internal cell signals and utility for state estimation. *J. Power Sources* **2017**, *341*, 474−482.

(69) Hsieh, A. G.; Bhadra, S.; Hertzberg, B. J.; Gjeltema, P. J.; Goy, A.; Fleischer, J. W.; Steingart, D. A. Electrochemical-acoustic time fo flight: in operando correlation of physical dynamics with battery charge and health. *Energy Environ. Sci.* **2015**, *8*, 1569−1577.

(70) Howell, D.; et al. Enabling Fast Charging: A Technology Gap Assessment; U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, October 2017; https://www.energy.gov/sites/prod/files/2017/10/f38/

XFC%20Technology%20Gap%20Assessment%20Report_FINAL_10202017.pdf.

(71) Bommier, C.; et al. *In Operando* Acoustic Detection of Lithium Metal Plating in Commercial $LiCoO_2$/Graphite Pouch Cells. *Cell Reports Physcial Science* **2020**, *1* (4), 100035.

(72) Yang, X.-G.; Liu, T.; Gao, Y.; Ge, S.; Leng, Y.; Wang, D.; Wang, C.-Y.; et al. Asymmetric Temperature Modulation for Extreme Fast Charging of Lithium-Ion Batteries. *Joule* **2019**, *3* (12), 3002−3019.

(73) Weber, R.; Genovese, M.; Louli, A. J.; Hames, S.; Martin, C.; Hill, I. G.; Dahn, J. R. Long cycle life and dendrite-free lithium morphology in anode-free lithium pouch cells enabled by a dual-salt liquid electrolyte. *Nature Energy* **2019**, *4*, 683−689.

(74) Liu, J.; Bao, Z.; Cui, Y.; Dufek, E. J.; Goodenough, J. B.; Khalifah, P.; Li, Q.; Liaw, B. Y.; Liu, P.; Manthiram, A.; Meng, Y. S.; Subramanian, V. R.; Toney, M. F.; Viswanathan, V. V.; Whittingham, M. S.; Xiao, J.; Xu, W.; Yang, J.; Yang, X.; Zhang, J. Pathways for practical high-energy long-cycling lithium metal batteries. *Nature Energy* **2019**, *4*, 180−186.

(75) Murugan, R.; Thangadurai, V.; Weppner, W. Fast Lithium Ion Conduction in Garnet-Type $Li_7La_3Zr_2O_{12}$. *Angew. Chem., Int. Ed.* **2007**, *46* (41), 7778−7781.

(76) Kamaya, N.; Homma, K.; Yamakawa, Y.; Hirayama, M.; Kanno, R.; Yonemura, M.; Kamiyama, T.; Kato, Y.; Hama, S.; Kawamoto, K.; Mitsui, A. A lithium superionic conductor. *Nat. Mater.* **2011**, *10*, 682−686.

(77) Masias, A.; Felten, N.; Garcia-Mendez, R.; Wolfenstine, J.; Sakamoto, J. Elastic, plastic and creep mechanical properties of lithium metal. *J. Mater. Sci.* **2019**, *54* (3), 2585−2600.

(78) Masias, A.; Felten, N.; Sakamoto, J. Characterizing the mechanical behavior of lithium in compression. *J. Mater. Res.* **2021**, 1−12.

(79) Hatzell, K. B.; Chen, X. C.; Cobb, C. L.; Dasgupta, N. P.; Dixit, M. B.; Marbella, L. E.; McDowell, M. T.; Mukherjee, P. P.; Verma, A.; Viswanathan, V.; Westover, A. S.; Zeier, W. G. Challenges in Lithium Metal Anodes for Solid-State Batteries. *ACS Energy Letters* **2020**, *5*, 922−934.