

# Key Challenges for Grid-Scale Lithium-Ion Battery Energy Storage

Yimeng Huang and Ju Li\*

**A rapid transition in the energy infrastructure is crucial when irreversible damages are happening quickly in the next decade due to global climate change. It is believed that a practical strategy for decarbonization would be 8 h of lithium-ion battery (LIB) electrical energy storage paired with wind/solar energy generation, and using existing fossil fuels facilities as backup. To reach the hundred terawatt-hour scale LIB storage, it is argued that the key challenges are fire safety and recycling, instead of capital cost, battery cycle life, or mining/manufacturing challenges. A short overview of the ongoing innovations in these two directions is provided.**

## 1. Eight Hours of Energy

Greta Thunberg commented on Twitter about the 2021 UN Climate Change Conference: “COP26 is over ... But the real work continues outside these halls. And we will never give up, ever.”<sup>[1]</sup> Energy storage is the real work. To halve the global CO<sub>2</sub> emission by Jan. 3, 2040, Greta’s 37<sup>th</sup> birthday, there are only 18 years left. Based on historical engineering experiences, there is no time left for a newborn, “baby” heavy industry (the so-called “B: Beyond-2040” technologies in the MIT A+B conference language<sup>[2]</sup>) to emerge from a university lab, mature, up-scale, and save the world in time from the irreversible damages of ocean acidification, loss of habitat, and societal upheaval. The Earth today is like a house on fire, and only the so-called “A: Action” type technologies that already exist today, with demonstrable terawatt scale capabilities, can dampen the raging fire by 2040. This means nuclear fission (specifically, light-water reactors), wind/solar generations, plus some forms of energy storage (heat, mechanical, battery, chemicals). Nuclear is type-A, as

in the 1970s it has already been demonstrated to lead the largest decarbonization actions to date, but is presently beset by very high construction cost.<sup>[3]</sup> “Desperate Times Call for Desperate Measures”, and energy storage seems more and more a human survival skill.

Here, we focus on the lithium-ion battery (LIB), a “type-A” technology that accounts for >80% of the grid-scale battery storage market,<sup>[4]</sup> and specifically, the market-prevalent battery chemistries using LiFePO<sub>4</sub> or LiNi<sub>x</sub>Co<sub>y</sub>Mn<sub>1-x-y</sub>O<sub>2</sub> on Al foil as the cathode, graphite on Cu foil as

the anode, and organic liquid electrolyte, which currently cost as low as US\$90/kWh(cell). LIBs can be deeply charged and discharged on the order of 10<sup>3</sup> cycles,<sup>[5]</sup> although this cycle life can vary greatly depending on cycling conditions and temperature. Going from LIB cells to battery packs to energy systems, one faces another 2× to 4× increase in cost, after thermal management, power electronics, safety measures, and controls<sup>[6]</sup> are added. In the past decade, there has been a 10-fold increase in cycle life and 6-fold decrease in pack-level cost,<sup>[7]</sup> assisted by the exponential growth in the electric vehicle (EV) supply chains. China broke the 1 million EV annual sales threshold in 2018. Realistically, one is probably looking at US\$200 to US\$300/kWh(system) capital expenditure (CAPEX) for LIB storage by 2025.

Among the existing electricity storage technologies today, such as pumped hydro, compressed air, flywheels, and vanadium redox flow batteries, LIB has the advantages of fast response rate, high energy density, good energy efficiency, and reasonable cycle life, as shown in a quantitative study by Schmidt et al.<sup>[8]</sup> In 10 of the 12 grid-scale application scenarios (ranging from black start, power quality, to primary, secondary, and tertiary responses), except for seasonal energy storage and primary response, LIB is expected to beat all other technologies by 10% or more in 2040, the time that matters.

The first question is: how much LIB energy storage do we need? Simple economics shows that LIBs cannot be used for seasonal energy storage. The US keeps about 6 weeks of energy storage in the form of chemical fuels, with more during the winter for heating.<sup>[9]</sup> Suppose we have reached US\$200/kWh battery cost, then US\$200 trillion worth of batteries (10× US GDP in 2020) can only provide 1000 TWh energy storage, or 3.4 quads. As the US used 92.9 quads of primary energy in 2020, this is only 2 weeks’ worth of storage, and not quite sufficient to heat our homes in the winter. Thus, very large-scale heat storage<sup>[9]</sup> and nuclear generations are likely needed for a 100% clean-energy infrastructure that can survive the winter. A real

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game-changer would come if we can synthesize liquid fuels efficiently, but day by day, this is looking more like a type-B, not type-A, projection.

The above does not mean LIBs cannot greatly help the low-carbon energy transition. It is clear from quantitative modeling<sup>[10]</sup> that just 8 h of battery energy storage, with a price tag of \$5 trillion (3 months of US GDP), would unlock significant wind/solar generations to be of some real utility in the direction of deeply reducing global CO<sub>2</sub> emission. A study by Ziegler et al.<sup>[10]</sup> showed that in warm states such as Arizona and Texas, the equivalent availability factor (EAF) of wind/solar + LIB can reach 95% and achieving cost parity with fossil fuel generations, if the LIB cost drops below US\$150/kWh(system). In other words, one can use wind/solar + LIB in 19 of 20 days, to reduce CO<sub>2</sub> emission by 80% or more. In the one unlucky day out of the 20 days, the week-long wind/solar drought would require us to fire up our natural-gas power plants, and rescue the wind/solar + LIB based grid in places such as Arizona and Texas. In colder states such as Massachusetts, this decarbonization solution would not be as thorough: we would need to fire up natural gas more often and use heavy oil to heat our homes, especially during the winter, but something like 50% reduction is still entirely possible. This plan says that we do not demolish our fossil-fuel powerplants and delivery infrastructure (indeed why should we, as they are sunk costs), but use them as backup systems at least before 2040–2050, while still deeply blunting the rate of ocean acidification and climate change, enabled by an economically feasible amount of LIB storage. We must still maintain and operate the fossil-fuel industry, albeit using them much less often by a factor of 5× or 10×, until the day type-B technologies mature. The corollary of this proposal is that we must expand the total workforce and acreage of the energy industries, by keeping two parallel, “legacy” and “A” systems. Thus, the levelized cost of electricity will be more expensive than what we have now, perhaps by as much as 50%. But “Desperate Times Call for Desperate Measures”. In sum, the

actionable solution appears to be ≈8 h of LIB storage stabilizing wind/solar + nuclear with heat storage, with the legacy fossil fuel systems as backup power (Figure 1).

LiFePO<sub>4</sub>//graphite (LFP) cells have an energy density of 160 Wh/kg(cell). Eight hours of battery energy storage, or 25 TWh of stored electricity for the United States, would thus require 156 250 000 tons of LFP cells. This is about 500 kg LFP cells (80 kWh of electricity storage) per person, in which there is about 6.5 kg of Li atoms (need to multiply by 5.32× for the corresponding lithium carbonate equivalent, LCE), and 29 kg of phosphorous atoms. To put this in perspective, oil tankers move about 2 billion tons of oil globally every year across ocean surfaces. The world’s per capita consumption of oil is 750 kg, and a US person’s consumption is 3.5 ton, per year, which are mostly oxidized and freely released into the atmosphere now.

While an endowment of 500 kg LFP cells (80 kWh of electricity storage) per person sounds reasonable, does Earth actually have enough lithium and other minerals to support it? The short answer is yes, if we are careful about recycling, which is one of the two key challenges ahead. The U.S. Geological Survey has identified “total about 80 million tons” of lithium atoms globally,<sup>[11]</sup> or 10.3 kg of Li atoms per person, on this planet, so there is enough lithium for everyone on Earth. This is not yet accounting for lithium from seawater, which costs 3× to 30× to extract than from hard rocks and brines.<sup>[12]</sup>

Granted, to scale up the global LIB industry by another factor of 10<sup>2</sup>× would be a herculean task,<sup>[13]</sup> causing tremendous industrial, ecological, and societal stresses. It is a civilization-scale endeavor and should not be taken lightly. The amount of investment (\$5 trillion) for the United States will be comparable to that of the War on Terror, the longest war in the US history. Nonetheless, it is doable by 2040 if one keeps 30% growth rate year-over-year, starting from now. Also note that “8 h of energy” is a colloquial term to show the scale in contrast to primary energy use, but if normalized by just electrical

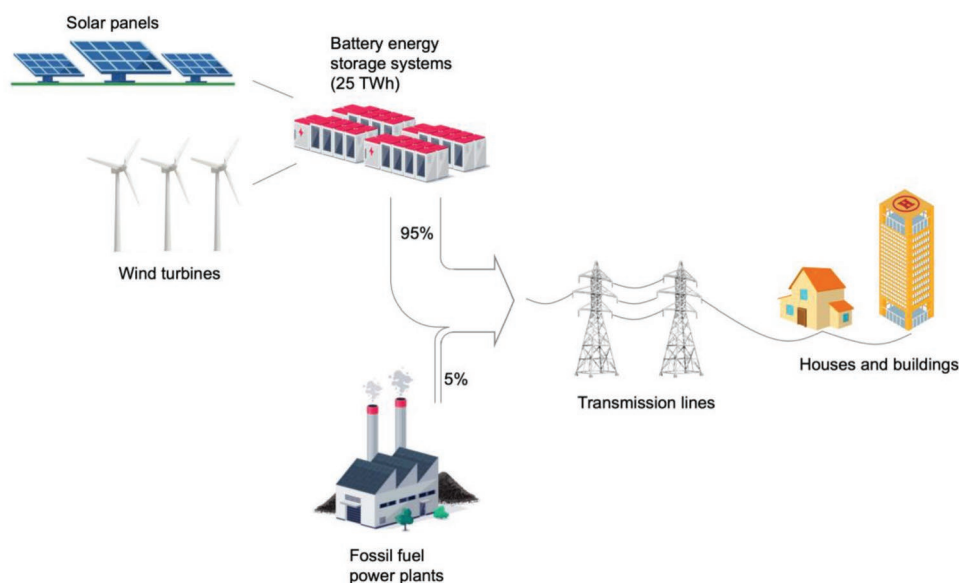


Figure 1. Schematic of sustainable energy production with 8 h of lithium-ion battery (LIB) storage.

energy use, it is more like 60 h, or 2.5 days, of electrical energy storage.

Aside from CAPEX, what about the operating expense (OPEX) that is closely related to the LIB cycle life? It turns out that the LIB cycle life has a very nonlinear relationship with the depth of charge–discharge, temperature, as well as the charging rate. Thus, better software and battery management systems<sup>[14]</sup> are tremendously important to operate safely and maximize long-term economic value. Hsu et al.<sup>[15]</sup> and Lu et al.<sup>[16]</sup> used deep neural networks to predict battery state of health (SOH), remaining useful life (RUL) and capacity–voltage curves, which are key for selecting newly manufactured or used cells for at-home or grid-scale battery packs and dynamic load-balancing.

The long-term LIB cycle life sensitively depends on the so-called Coulombic inefficiency, which is the percentage of Li atom inventory that becomes deactivated each time an inventory is deposited into and extracted from the electrodes. By tuning the electrolyte chemistry<sup>[17,16]</sup> and electrode coatings,<sup>[18]</sup> it is possible to reduce the Coulombic inefficiency and increase the cycle life exponentially.<sup>[19,20]</sup> Recently, our group used robotic arms and automatic testing apparatus paired with active learning algorithms to conduct high throughput electrolyte testing (ongoing).<sup>[21]</sup> Prof. Jeff Dahn has shown that 10 000–20 000 cycles are achievable with electrolyte tuning,<sup>[16]</sup> thus reducing the environmental impact of the EV industry and facilitating vehicle-to-grid storage.

While there is a tremendous amount of work remaining to be done, cycle life, mining/manufacturing, or capital cost per se will not be the showstopper to LIB energy storage systems (ESS) becoming a type-A solution in tackling climate challenges in the next decade. But fire safety and recycling challenges could well be, as we outline below.

## 2. Fire Safety

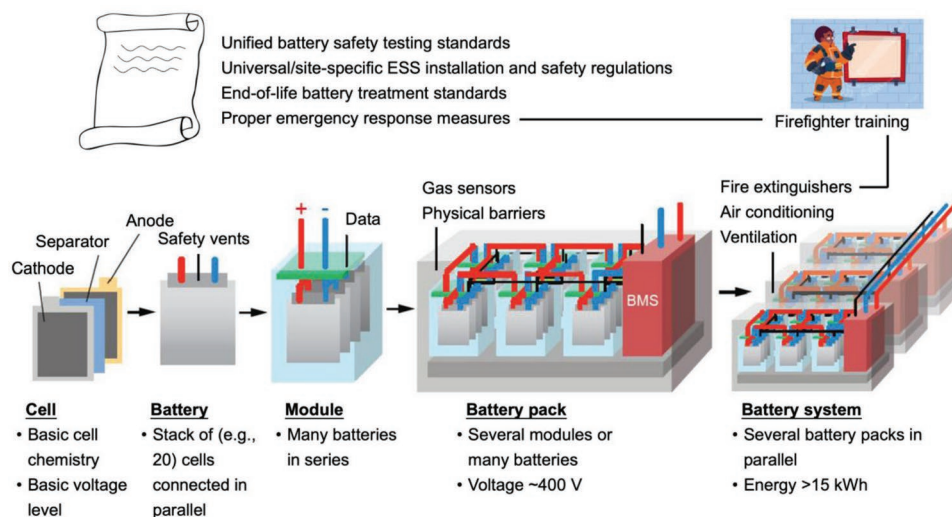
On April 19, 2019, the fire and explosion at a 2 MWh  $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$  (NCM)//graphite ESS facility in Arizona caused eight firefighter injuries. On April 16, 2021, the explosion at a 25 MWh LFP ESS station in Beijing, China caused the death of two firefighters. In South Korea alone, between 2017 and 2019 there had been 28 fire accidents, leading to the shutdown of 522 ESS units after regulatory review, or  $\approx 35\%$  of all ESS installations.<sup>[22]</sup> While the chance of an individual battery cell failure under normal use is on the order of  $10^{-7}$  in its life, due to the cascading nature of fire accidents, the probability of an ESS facility catching fire with millions of cells, leading to severe accidents, is apparently not low with the present generations of ESS. These accidents result in electricity supply disruptions, severe pollution, and huge economic losses. The liabilities need to be added to the OPEX and are often underestimated.

For safer battery cells in ESS,  $\text{LiFePO}_4$ -based chemistries are chosen over the layered oxides (preferred for EV applications for their higher energy density and power) for their much-improved cycling and thermal stability, as well as low cost.<sup>[23]</sup> Nevertheless, the initial cause of an ESS fire may not be a battery cell at all but faulty wiring, electric shock protection, control-system bugs, etc.<sup>[24]</sup> For comparison, consider the technology of AC transformers on electrical grid: even after

100+ years of development and heavy usage, the “probability of a serious transformer fire is on the order of 0.06% to 0.1% per service year.”<sup>[25]</sup> Grid-scale ESS is much newer and more complex than transformers, and the thermal design is more challenging (the up-to-30% round-strip energy loss needs to be dissipated as heat<sup>[6]</sup>), so fault diagnostics and risk mitigation are more demanding. Natural disasters such as tornados, flooding, and man-made causes such as cyberattacks and even arson must be considered. A defense-in-depth design and rapid response strategy are needed to minimize life loss and collateral damage.

Regardless of where the thermal runaway first initiates, there is a tremendous driving force for this runaway to amplify. The cathode material in LIBs is highly oxidative at high states of charge (SOC) and can, especially at high temperatures, release oxygen (contributed by surface oxygen ions as well as mobile lattice oxygen ion<sup>[26,27]</sup>), whereas the anode material is highly reducing, separated by just 10  $\mu\text{m}$ -thick nanoporous battery separator made of polypropylene or polyethylene. Both the anode and the liquid electrolyte can act as fuels. No external  $\text{O}_2$  supply is required for a battery cell to heat itself up to several hundred degrees Celsius, by internal short-circuiting (ISC) or external short-circuiting (ESC).<sup>[28]</sup> The battery separator tends to shrink in total area above  $\approx 110^\circ\text{C}$ , exposing more naked contact between the cathode and the anode for ISC. Furthermore, the solvent of the liquid electrolyte is volatile, generating larger vapor pressure (boiling) at high temperatures. After the cell packaging is breached, convective mixing of external  $\text{O}_2$  with battery materials will exacerbate violent, rocket-like bursts and explosions. The total heat of combustion of NCM batteries is on the order of 5–10 MJ(heat)/kg(cell), which is nearly 10 $\times$  of its reversible electrical energy storage ( $\approx 200\text{ Wh kg}^{-1}$ ), and higher than the embedded energy of TNT ( $4.6\text{ MJ kg}^{-1}$ ). Thus, container-scale ESS systems are somewhat similar to an ammunition dump, which also actively gives off heat! The raw-energy comparison is of course a bit misleading since paper and plastic both have heat of combustion significantly higher than TNT. The rate, or kinetics, of heat release at different time- and length-scales in different scenarios is critical for quantifying the flammability of battery cells, and innovations in materials and electrolytes, cell architecture, sensors and safety systems, battery management systems, national/local safety regulations, and firefighting preparations are all essential in retarding the exothermicity (summarized in **Figure 2**). Again, it is important to emphasize the active nature of the heat release (up to 0.2 MJ(heat)/kg(cell) per deep charge–discharge cycle) in ESS, compared to the passive ammunition dump.

Most research efforts on fire safety thus far have been focused on improving battery materials at the cell level. These include but are not limited to: optimizing thermally stable  $\text{LiFePO}_4$  and stabilizing high-voltage  $\text{LiCoO}_2$ /NCM cathodes via doping<sup>[29–34]</sup> or coating<sup>[18,35–39]</sup> methods, improving graphite anodes via mild oxidation,<sup>[40]</sup> coating,<sup>[41]</sup> and morphological modification<sup>[42]</sup> methods to obtain a stable SEI layer and finding alternative anodes (such as silicon nanowires,<sup>[43,44]</sup>  $\text{Fe}_3\text{O}_4$ ,<sup>[45]</sup> and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ <sup>[46]</sup>), reducing the flammability of electrolytes via strategies such as replacing salts and solvents,<sup>[47,48]</sup> using functional additives,<sup>[49,50]</sup> and finding nonflammable alternatives (such as ionic liquids,<sup>[51]</sup> gelled polymer-based electrolytes,<sup>[52]</sup>



**Figure 2.** Schematic of different levels of lithium-ion battery (LIB) storage and critical components considered for fire safety. Reproduced with permission under the terms of the Creative Commons CC BY license.<sup>[61]</sup> Copyright 2020, the Authors. Published by MDPI.

and inorganic solid electrolytes<sup>[53]</sup>). Although cell-level materials development has seen significant progress, thermal runaway risks within the cell cannot be fully eliminated. This calls for additional layers of protection from proper cell architecture and extrinsic safety devices, which is beyond the cell level. Research in these fields has risen rapidly in recent years, but technologies are still immature. Topics of interest include safety vents,<sup>[54]</sup> current interrupt device,<sup>[55]</sup> positive temperature coefficient device,<sup>[56]</sup> shutdown separator,<sup>[57]</sup> gas sensors,<sup>[58]</sup> battery management systems<sup>[14]</sup> and their integration with energy/power management systems,<sup>[24]</sup> and cell compartmentation (cell spacing and physical barriers<sup>[59]</sup>). Fire suppression is another emerging field of research, which is nontrivial since LIB fires differ drastically from traditional fires, due to factors such as lithium's high reactivity with water, explosion risk of the battery, un-necessity of external oxygen supply to maintain a LIB fire, and emission of toxic fluoride gases.<sup>[59,60]</sup> Investigations on different suppressants

(e.g., water mists, halon-based products<sup>[61]</sup>), additives (e.g., C<sub>6</sub>F<sub>4</sub>-ketone<sup>[59]</sup>), and strategies are underway, but progress is still at its infancy and requires further work.

Aside from technological advances, the control of fire hazards is also decided by developments in regulations and management. LIBs must undergo a series of safety tests to be used in applications such as EVs and stationary storage. Although several standards and regulations have been published internationally and domestically, (see **Table 1**) there is still a lack of consistency in testing conditions, testing parameters, and pass/fail criteria for safety tests.<sup>[59]</sup> For instance, there is great variability in nail material, size, and penetration depth used for penetration tests, and in testing conditions such as the SOC, temperature, and charging rates. The inconsistencies of safety tests result in large fluctuations in LIB quality across the industry, which may bring difficulties to troubleshooting and safety policy making. Another concern is that most tests are performed on the easily accessible cell level, which might not

**Table 1.** Selected standards and regulations for grid-storage battery safety.

Organization	Code	Content	Reference
International Electrotechnical Commission	IEC 62619	Requirements and tests for safety operation of lithium-ion batteries (LIBs) in industrial applications (including energy storage systems [ESS])	[64]
National Fire Protection Association	NFPA 855	Standard for installation of ESS	[65]
	NFPA 70	National electrical code (for safe electrical design, installation, and inspection)	
	NFPA 13	Standard for sprinkler systems installation	
	NFPA 1	Fire code regulation and hazard management	
American National Standards Institute	NFPA 1620	Standard for pre-incident planning	
	UL 9540	Test standards for the interaction between battery and power electronics in ESS	[66]
	UL 9540A	Evaluation of thermal runaway fire propagation in ESS	
International Code Council	UL 1973	Batteries safety standards for stationary and motive auxiliary power applications	
	RB154	Restrictions for ESS size and siting location for homeowners	[66]
Standardization Administration of the P.R.C.	GB/T 34131	Technical specifications for LIB management systems for ESS	[67]
	GB/T 36276	Safety standards of LIBs for power energy storage	

be indicative of pack/system level safety performances. A lack of consensus on system design and evaluation of system-level safety still exists in the industry.

Emergency responses to LIB fire accidents are another big part of safety, since minor thermal runaway in cells can lead to chain reactions that release huge amounts of energy in ESS with millions of cells, greatly endangering human lives and assets. It is thus critical to implement proper emergency response measures to contain ESS fire hazards. In the case of the Arizona ESS fire, the firefighters opened the ESS door, which brought the flammable gases inside into contact with a spark or heat source that led to the explosion, causing severe injuries.<sup>[62]</sup> Despite the existence of standards and regulations on international/domestic levels, they are absent on the local level specific to each individual ESS site. Site-specific regulations are critically needed because each site has its unique conditions, such as the cell material, quantities of cells, module component types, system design, temperature and humidity of the site, and availability of water resources and first responders. For instance, most of the ESS in the South Korean fire incidents are located in mountainous and coastal areas with large temperature swings and high humidity, which led to condensation that resulted in residue after drying, and eventually degrading the electrical insulating components. All these factors play a role in deciding the operation mode, type of fire extinguishing agents used, and the specific steps first responders should take. These should be transparent and taken seriously. The site-specific guidelines, as well as the general (inter)national ones, should be inspected and updated frequently to keep up with the rapid changes in the battery energy storage industry. Stakeholders should also make sure that firefighters are well-educated and have up-to-date trainings, as methods for extinguishing LIB fires are largely different from typical fires.

End-of-life treatment of LIBs also creates serious fire hazards and should not be taken lightly. For both recycling and disposal, LIBs can be damaged during various steps, such as jostling during collection and transportation, and crushing during mechanical disassembly.<sup>[63]</sup> These physically intense processes greatly increase fire hazards and should be restricted with tight regulations, but they are lacking at the moment. A summary of the critical aspects considered for LIB fire safety is schematically shown in Figure 2.

### 3. Truly 'Green' Renewable Energy

Besides safety concerns, whether renewable energy is heavily "green" remains questionable.<sup>[68]</sup> The mineral-intense

production of wind turbines, solar panels, and LIBs creates a tough problem for supply chains and Earth's limited mineral deposits. For example, LIB production today already consumes 40% and 25% of all lithium and cobalt mining capacities, respectively, and with batteries becoming more dominant in the future, global mining capacities would have to expand by 200% or more for resources such as copper, lithium, cobalt, graphite, and rare-earth elements.<sup>[13]</sup> In addition, mineral mining and LIB production both produce substantial amounts of CO<sub>2</sub>, and these green devices mostly end up in landfills or oceans upon retirement, generating large amounts of waste plastic and heavy metals that pose serious threats to our environment. It is thus imperative to recycle these devices to ensure a sustainable mineral supply chain and reduce pollution.

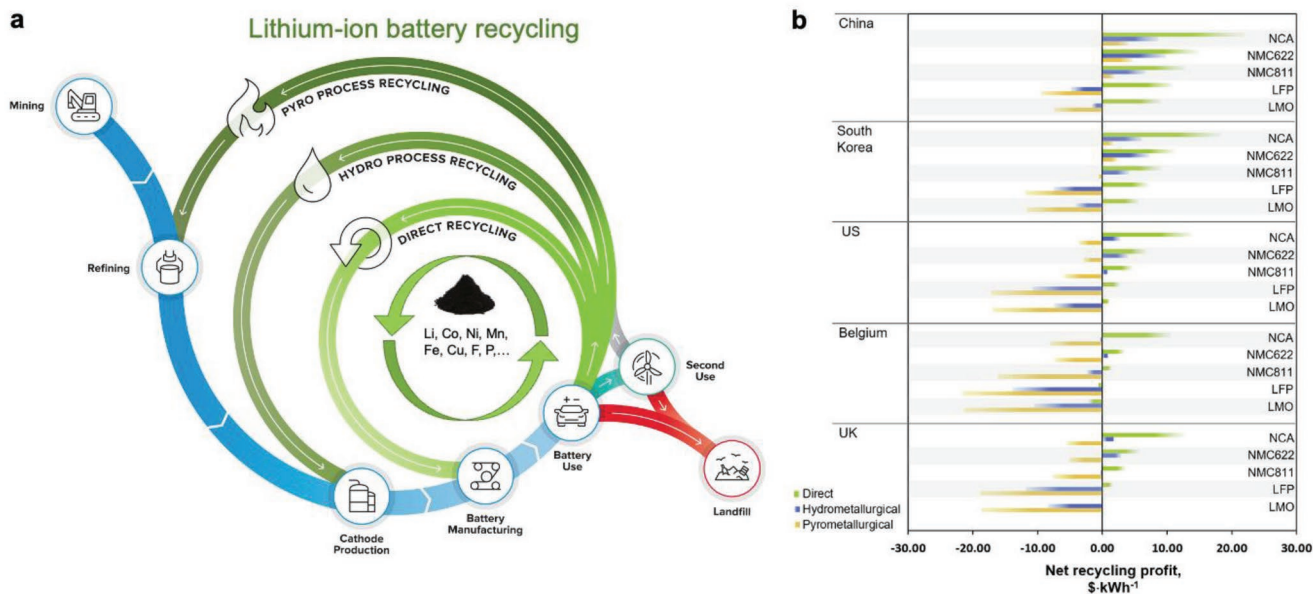
As of 2019, a mere 5% of LIBs are recycled worldwide,<sup>[69]</sup> resulting from factors such as the absence of regulations, complex and expensive recycling processes, and the lack of recycling technologies and facilities.<sup>[70]</sup> Pyrometallurgy and hydrometallurgy are the main methods of recycling today that both aim for the extraction of valuable metals such as Co and Ni in their metallic form, but they are energy-consuming and not environmentally/economically friendly.<sup>[71]</sup> They have even been shown to bring negative CO<sub>2</sub> emission reduction compared to not recycling at all.<sup>[70]</sup>

Direct recycling has been developed in the past few years for being more environmentally/economically viable, which repairs the active materials that have undergone lithium loss or structural transformation, instead of extracting constituent elements.<sup>[72]</sup> Compared with pyro/hydrometallurgy, direct recycling methods consumes only ≈15% of the energy, produces ≈25% of the CO<sub>2</sub> emission, and cost ≈50% less<sup>[73]</sup> (see **Table 2** for detailed numbers). This is especially important for ESS applications that heavily depend on chemistries with less valuable elements, such as LiFePO<sub>4</sub> or LiMn<sub>2</sub>O<sub>4</sub>, of which the direct recycling can potentially be profitable (**Figure 3**). A common direct recycling strategy is to mix the spent active material with new active material or extra lithium sources and then perform heat treatment, with the aim of recovering lost lithium or repairing the damaged crystal structure. The reconditioned active materials can be directly made into new electrodes for battery remanufacture, which greatly reduces the cost and emissions from extracting metal constituents and resynthesis of active materials. Other facile and inexpensive direct recycling methods include hydrothermal regeneration,<sup>[74]</sup> selective healing,<sup>[75]</sup> mechanochemical activation,<sup>[76]</sup> microwave,<sup>[77]</sup> repair using deep eutectic solvent,<sup>[78]</sup> rapid thermal radiation,<sup>[79]</sup> and rapid Joule heating.<sup>[80,81]</sup> To ensure both practicality and quality, novel direct recycling (shallow recycling) methods can be used in combination with conventional

**Table 2.** Comparison of recycling methods.

Method	Energy consumption [MJ kg <sup>-1</sup> cell]			CO <sub>2</sub> emission [kg kg <sup>-1</sup> cell]			Cost [\$ kg <sup>-1</sup> cell]			Net profit [\$ kg <sup>-1</sup> cell]		
	LFP <sup>[75]</sup>	LMO <sup>[85]</sup>	LCO <sup>[78]</sup>	LFP <sup>[75]</sup>	LMO <sup>[85]</sup>	LCO <sup>[78]</sup>	LFP <sup>[75]</sup>	LMO <sup>[85]</sup>	LCO <sup>[78]</sup>	LFP <sup>[75]</sup>	LMO <sup>[85]</sup>	LCO <sup>[78]</sup>
Pyrometallurgy	18.4	18.6	152.5	2.5	2.3	11.3	3.4	2.4	4.1	-2.6	-0.5	0.3
Hydrometallurgy	30.6	30.7	160.7	2.4	2.3	10.8	2.4	1.3	3.8	-1.4	0.4	0.2
Direct Recycling	3.6	4.1	112.1	0.7	0.5	8.3	2.1	0.8	3.7	1.1	2.0	1.7

<sup>a</sup>LFP: LiFePO<sub>4</sub>, LMO: LiMn<sub>2</sub>O<sub>4</sub>, LCO: LiCoO<sub>2</sub>



**Figure 3.** a) Schematic of closed-loop recycling of lithium-ion batteries. Image adapted from Argonne National Laboratory ReCell Center. Adapted with permission under the terms of the Creative Commons CC BY license.<sup>[86]</sup> Copyright 2021, the Authors. Published by MDPI. b) Net recycling profit of batteries by country and chemistry. Reproduced with permission under the terms of the Creative Commons CC BY license.<sup>[87]</sup> Copyright 2017, the Authors. Published by Elsevier Inc.

metallurgical processes (deep recycling). For instance, one could perform a deep recycling step for LIBs that have undergone 10 shallow recycling steps. A summary of the recycling processes for LIBs is shown schematically in Figure 3.

Parallel with the advancements in recycling technologies should be the development in waste management and policy making, for ensuring a true circular economy. Since game-changing recycling technologies cannot mature within a short period of time, it is likely in the near future that the majority of LIBs still remain un-recycled, ending up stockpiled, landfilled, or incinerated.<sup>[82]</sup> Toxic organic solvents, plastics, and heavy metals from waste LIBs can leach into the soil and pollute oceans if not properly managed and disposed. There are several existing standards and regulations for LIB disposal, both on federal and state levels, such as the Resource Conservation and Recovery Act in the U.S. and hazardous waste handling regulations in China,<sup>[83]</sup> but problems still exist such as ambiguities on the classification of LIBs, failure to keep up to date with technological advances, lack of standards for data collection, reporting, and tracking,<sup>[84]</sup> which should all be addressed promptly.

#### 4. Outlook

With growing energy demands, resources being drained and the climate worsening at exponential rates, the decarbonization of energy production is inevitable. At the current technological stage with economic and environmental considerations, 8 h of LIB storage paired with wind/solar (type-A technologies) generating energy fulfilling 95% of demand, and using conventional fossil fuels as backup should be the realistic strategy for energy decarbonization in the near future, until Type-B technologies (e.g., fusion power engineering<sup>[88]</sup> and superconducting transmission) mature. With continuous efforts in LIB energy

density, cost efficiency, and cycle life, the numbers (8 h, 95%, etc.) will improve, but the two real challenges that lie ahead are fire safety and recycling, which have been relatively overlooked in the past compared to the pursuits of low cost, long cycle life, and high energy density, but are critical for ensuring battery reliability and true environmentally friendliness. Luckily, they have attracted growing attention recently and have seen significant innovations and progress. Technologically, for fire safety, people have gained a deeper understanding of the origins of heat generation and thermal runaway,<sup>[89]</sup> developed better designs on both materials and engineering levels, including reliable high-voltage cathodes, anodes with stable SEI, flame-resistant liquid electrolytes, solid electrolytes with good cyclability, and better extrinsic safety devices and battery management systems. For recycling, people are pursuing more economically and environmentally viable metallurgical processes (deep recycling), as well as innovative direct recycling (shallow recycling) methods. An optimized combination of shallow and deep recycling could further boost the economic and environmental performance of recycling, forming a closed loop of valuable elements such as Co, Ni, and Li within the renewable energy industry, and thus greatly reducing resource and mining burdens under the rapid growth in energy storage demands. In addition, regulations and management must be improved synchronously with advancements in technology to further enhance safety and sustainability. Up-to-date site-specific installation/safety guidelines and emergency response measures need to be developed, end-of-life fire risks need to be taken seriously, and waste management policies need to be developed and enforced. Achieving a circular economy for renewable energy paired with LIB storage will require widescale collaboration between academia, industry, and governments. With both technological and managerial improvements, we will be closer to having reliable <US\$90/kWh battery packs that could cycle stably up to 20 000 times and beyond for safe and sustainable grid storage.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

battery recycling, fire safety, grid-scale storage

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- [1] G. Thunberg, <https://twitter.com/GretaThunberg/status/1459612735294029834> (accessed: November 2021).
- [2] ICAE, *Applied Energy Symposium: MIT A+B*, MIT, Cambridge, USA, August 2021, <https://applied-energy.org/mitab2021/index>.
- [3] P. Eash-Gates, M. M. Klemun, G. Kavlak, J. Mcnerney, J. Buongiorno, J. E. Trancik, *Joule* **2020**, *4*, 2348.
- [4] *Market Analysis Report*, Grand View Research, San Francisco, CA, USA **2022**, pp. 2020–2027, <https://www.grandviewresearch.com/industry-analysis/grid-scale-battery-storage-market>.
- [5] Y. Preger, H. M. Barkholtz, A. Fresquez, D. L. Campbell, B. W. Juba, J. Romā N-Kustas, S. R. Ferreira, B. Chalamala, *J. Electrochem. Soc.* **2020**, *167*, 120532.
- [6] M. Schimpe, M. Naumann, N. Truong, H. C. Hesse, S. Santhanagopalan, A. Saxon, A. Jossen, *Appl. Energy* **2018**, *210*, 211.
- [7] L. Trahey, F. R. Brushett, N. P. Balsara, G. Ceder, L. Cheng, Y.-M. Chiang, N. T. Hahn, B. J. Ingram, S. D. Minter, J. S. Moore, K. T. Mueller, L. F. Nazar, K. A. Persson, D. J. Siegel, K. Xu, K. R. Zavadil, V. Srinivasan, G. W. Crabtree, *Proc. Natl. Acad. Sci. USA* **2020**, *117*, 12550.
- [8] O. Schmidt, S. Melchior, A. Hawkes, I. Staffell, *Joule* **2019**, *3*, 81.
- [9] C. Forsberg, *Electr. J.* **2021**, *34*, 107042.
- [10] M. S. Ziegler, J. M. Mueller, G. A. D. Pereira, J. Song, M. Ferrara, Y.-M. Chiang, J. E. Trancik, *Joule* **2019**, *3*, 2867.
- [11] *Mineral Commodity Summaries 2020*, U.S. Geological Survey, VA, USA **2020**, <https://doi.org/10.3133/mcs2020>.
- [12] P. Greim, A. A. Solomon, C. Breyer, *Nat. Commun.* **2020**, *11*, 4570.
- [13] M. P. Mills, 2019. *The “New Energy Economy”: An Exercise in Magical Thinking*, The Manhattan Institute, New York, NY, USA **2019**, <https://www.manhattan-institute.org/green-energy-revolution-near-impossible>
- [14] H. Rahimi-Eichi, U. Ojha, F. Baronti, M.-Y. Chow, *IEEE Ind. Electron. Mag.* **2013**, *7*, 4.
- [15] C.-W. Hsu, R. Xiong, N.-Y. Chen, Ju Li, N. -T. Tsou, *Appl. Energy* **2022**, *306*, 118134.
- [16] E. R. Logan, H. Hebecker, A. Eldesoky, A. Luscombe, M. B. Johnson, J. R. Dahn, *J. Electrochem. Soc.* **2020**, *167*, 130543.
- [17] W. Song, J. Harlow, E. Logan, H. Hebecker, M. Coon, L. Molino, M. Johnson, J. Dahn, M. Metzger, *J. Electrochem. Soc.* **2021**, *168*, 090503.
- [18] M. Yoon, Y. Dong, J. Hwang, J. Sung, H. Cha, K. Ahn, Y. Huang, S. J. Kang, J. Li, J. Cho, *Nat. Energy* **2021**, *6*, 846.
- [19] L. Suo, W. Xue, M. Gobet, S. G. Greenbaum, C. Wang, Y. Chen, W. Yang, Y. Li, J. Li, *Proc. Natl. Acad. Sci. USA* **2018**, *115*, 1156.
- [20] W. Xue, M. Huang, Y. Li, Y. G. Zhu, R. Gao, X. Xiao, W. Zhang, S. Li, G. Xu, Y. Yu, P. Li, J. Lopez, D. Yu, Y. Dong, W. Fan, Z. Shi, R. Xiong, C.-J. Sun, I. Hwang, W.-K. Lee, Y. Shao-Horn, J. A. Johnson, J. Li, *Nat. Energy* **2021**, *6*, 495.
- [21] Liquid Handling Robot with 3D Printed Pipette Handler, <https://www.youtube.com/watch?v=ScDYSUFRFZO> (accessed: (September 2020).
- [22] A. Elder, D. Sandahl, *Energy Storage News*, <https://www.energy-storage.news/preventing-thermal-runaway-in-lithium-ion-energy-storage-systems/> (accessed: May 2021).
- [23] B. Dunn, H. Kamath, J.-M. Tarascon, *Science* **2011**, *334*, 928.
- [24] Li-ion Tamer, <https://liiontamer.com/south-korea-identifies-top-4-causes-that-led-to-ess-fires/> (accessed: June 2019).
- [25] A. Petersen, *T&D World*, <https://www.tdworld.com/substations/article/20964130/risk-equals-probability-times-consequences> (accessed: April 2014).
- [26] P. Yan, J. Zheng, Z.-K. Tang, A. Devaraj, G. Chen, K. Amine, Ji-G Zhang, Li-M Liu, C. Wang, *Nat. Nanotechnol.* **2019**, *14*, 602.
- [27] Y. Dong, L. Qi, A. Alvarez, J. Li, I.-W. Chen, *Acta Mater.* **2022**, *232*, 117941.
- [28] R. Xiong, S. Ma, H. Li, F. Sun, J. Li, *iScience* **2020**, *23*, 101010.
- [29] P. Yan, A. Nie, J. Zheng, Y. Zhou, D. Lu, X. Zhang, R. Xu, I. Belharouak, X. Zu, J. Xiao, K. Amine, J. Liu, F. Gao, R. Shahbazian-Yassar, Ji-G Zhang, C.-M. Wang, *Nano Lett.* **2015**, *15*, 514.
- [30] Y. Huang, Y. Zhu, H. Fu, M. Ou, C. Hu, S. Yu, Z. Hu, C. T. Chen, G. Jiang, H. Gu, H. Lin, W. Luo, Y. Huang, *Angew. Chem., Int. Ed.* **2021**, *60*, 4682.
- [31] R. Alcántara, P. Lavela, J. L. Tirado, R. Stoyanova, E. Zhecheva, *J. Solid State Chem.* **1997**, *134*, 265.
- [32] Z. Feng, R. Rajagopalan, S. Zhang, D. Sun, Y. Tang, Y. Ren, H. Wang, *Adv. Sci.* **2021**, *8*, 2001809.
- [33] Q. Liu, X. Su, D. Lei, Y. Qin, J. Wen, F. Guo, Y. A. Wu, Y. Rong, R. Kou, X. Xiao, F. Aguesse, J. Bareño, Y. Ren, W. Lu, Y. Li, *Nat. Energy* **2018**, *3*, 936.
- [34] S. J. Do, P. Santhoshkumar, S. H. Kang, K. Prasanna, Y. N. Jo, C. W. Lee, *Ceram. Int.* **2019**, *45*, 6972.
- [35] S.-H. Lee, C. S. Yoon, K. Amine, Y.-K. Sun, *J. Power Sources* **2013**, *234*, 201.
- [36] M. K. Azar, M. A. R. Kholari, M. Esmaeili, E. Heidari, S. M. Hosseini-Hosseiniabad, R. S. Moakhar, A. Dolati, S. Ramakrishna, *ACS Appl. Energy Mater.* **2021**, *4*, 934.
- [37] H.-J. Kweon, J. Park, J. Seo, G. Kim, B. Jung, H. S. Lim, *J. Power Sources* **2004**, *126*, 156.
- [38] S. Oh, J. K. Lee, D. Byun, W. I. Cho, B. W. Cho, *J. Power Sources* **2004**, *132*, 249.
- [39] A. T. Appapillai, A. N. Mansour, J. Cho, Y. Shao-Horn, *Chem. Mater.* **2007**, *19*, 5748.
- [40] H.-J. Guo, X.-H. Li, Z.-X. Wang, W.-J. Peng, Y.-X. Guo, *J. Cent. South Univ.* **2005**, *12*, 50.
- [41] A. Friesen, S. Hildebrand, F. Horsthemke, M. Börner, R. Klöpsch, P. Niehoff, F. M. Schappacher, M. Winter, *J. Power Sources* **2017**, *363*, 70.
- [42] L. G. Bulusheva, A. V. Okotrub, A. G. Kurennya, H. Zhang, H. Zhang, X. Chen, H. Song, *Carbon* **2011**, *49*, 4013.
- [43] V. Chakrapani, F. Rusli, M. A. Filler, P. A. Kohl, *J. Power Sources* **2012**, *205*, 433.
- [44] Y. Yao, N. Liu, M. T. Mcdowell, M. Pasta, Y. Cui, *Energy Environ. Sci.* **2012**, *5*, 7927.
- [45] G. Zhou, D.-W. Wang, F. Li, L. Zhang, N. Li, Z.-S. Wu, L. Wen, G. Q. (.M.). Lu, H.-M. Cheng, *Chem. Mater.* **2010**, *22*, 5306.
- [46] T.-F. Yi, Y. Xie, Y.-R. Zhu, R.-S. Zhu, H. Shen, *J. Power Sources* **2013**, *222*, 448.
- [47] K. Xu, S. Zhang, T. R. Jow, W. Xu, C. A. Angell, *Electrochem. Solid-State Lett.* **2001**, *5*, A26.
- [48] J. Wang, Y. Yamada, K. Sodeyama, C. H. Chiang, Y. Tateyama, A. Yamada, *Nat. Commun.* **2016**, *7*, 12032.

- [49] S. S. Zhang, *J. Power Sources* **2006**, 162, 1379.
- [50] Q. Wang, L. Jiang, Y. Yu, J. Sun, *Nano Energy* **2019**, 55, 93.
- [51] M. Ishikawa, T. Sugimoto, M. Kikuta, E. Ishiko, M. Kono, *J. Power Sources* **2006**, 162, 658.
- [52] P. Yao, H. Yu, Z. Ding, Y. Liu, J. Lu, M. Lavorgna, J. Wu, X. Liu, *Front. Chem.* **2019**, 7, 522.
- [53] F. Zheng, M. Kotobuki, S. Song, M. O. Lai, L. Lu, *J. Power Sources* **2018**, 389, 198.
- [54] J.-H. Kim, K.-H. Lee, D.-C. Ko, S.-B. Lee, B.-M. Kim, *J. Mech. Sci. Technol.* **2017**, 31, 2505.
- [55] F. Lin, J. Li, X. Hu, M. Sun, *Int. J. Energy Res.* **2020**, 44, 11185.
- [56] X. M. Feng, X. P. Ai, H. X. Yang, *Electrochem. Commun.* **2004**, 6, 1021.
- [57] M. Baginska, B. J. Blaiszik, R. J. Merriman, N. R. Sottos, J. S. Moore, S. R. White, *Adv. Energy Mater.* **2012**, 2, 583.
- [58] Y. Jin, Z. Zheng, D. Wei, X. Jiang, H. Lu, L. Sun, F. Tao, D. Guo, Y. Liu, J. Gao, Yi Cui, *Joule* **2020**, 4, 1714.
- [59] L. B. Diaz, X. He, Z. Hu, F. Restuccia, M. Marinescu, J. V. Barreras, Y. Patel, G. Offer, G. Rein, *J. Electrochem. Soc.* **2020**, 167, 090559.
- [60] J. Thibeau, *CellBlock Fire Containment Systems*, <https://cellblockfcs.com/why-is-a-lithium-fire-worse-than-a-regular-fire/> (accessed: February 2020).
- [61] M. Ghiji, V. Novozhilov, K. Moinuddin, P. Joseph, I. Burch, B. Suendermann, G. Gamble, *Energies* **2020**, 13, 5117.
- [62] A. Colthorpe, *DNV GL says. Energy Storage News*, <https://www.energy-storage.news/arizona-battery-fires-lessons-can-be-learned-by-industry-to-prevent-further-incidents-dnv-gl-says/> (accessed: July 2020).
- [63] M. R. Timpane, [https://rethinkwaste.org/wp-content/uploads/legacy\\_media/7-a-attachment-d-lithium-based-battery-assessment-2017.original.pdf](https://rethinkwaste.org/wp-content/uploads/legacy_media/7-a-attachment-d-lithium-based-battery-assessment-2017.original.pdf) (accessed: 2017).
- [64] *BatteryStandards.Info*, <https://batterystandards.info/standard/iec-62619-nwp>
- [65] National Fire Protection Association, <https://www.nfpa.org/media/Files/Code%20or%20topic%20fact%20sheets/ESSFactSheet.pdf> (accessed: June 2020).
- [66] P. Dailey, <https://www.pv-magazine.com/2020/08/21/how-standards-can-ensure-battery-safety/> (accessed: August 2020).
- [67] DGBell, <https://belltestchamber.com/lithium-ion-battery-energy-storage-safety-standards-part-2.html> (accessed: March 2022).
- [68] Youtube, The Contradictions of Battery Operated Vehicles, <https://www.youtube.com/watch?v=S1E8SQde5rk> (accessed: January 2020).
- [69] M. Jacoby, Chemical and Engineering News, <https://cen.acs.org/materials/energy-storage/time-serious-recycling-lithium/97/i28> (accessed: July 2019).
- [70] R. E. Ciez, J. F. Whitacre, *Nat. Sustainability* **2019**, 2, 148.
- [71] O. Velázquez-Martínez, J. Valio, A. Santasalo-Aarnio, M. Reuter, R. Serna-Guerrero, *Batteries* **2019**, 5, 68.
- [72] X. Wu, J. Ma, J. Wang, X. Zhang, G. Zhou, Z. Liang, *Met. Sustainability* **2022**, 2200067.
- [73] D. Steward, A. Mayyas, M. Mann, *Procedia Manuf.* **2019**, 33, 272.
- [74] Q. Jing, J. Zhang, Y. Liu, W. Zhang, Y. Chen, C. Wang, *ACS Sustainable Chem. Eng.* **2020**, 8, 17622.
- [75] P. Xu, Q. Dai, H. Gao, H. Liu, M. Zhang, M. Li, Y. Chen, K. An, Y. S. Meng, P. Liu, Y. Li, J. S. Spangenberg, L. Gaines, J. Lu, Z. Chen, *Joule* **2020**, 4, 2609.
- [76] X. Meng, J. Hao, H. Cao, X. Lin, P. Ning, X. Zheng, J. Chang, X. Zhang, B. Wang, Z. Sun, *Waste Manage.* **2019**, 84, 54.
- [77] J. Sun, Z. J. , G. Yu, P. Jia, W. Wang, *Huanjing Gongcheng Xuebao* **2021**, 15, 2191.
- [78] J. Wang, Q. Zhang, J. Sheng, Z. Liang, J. Ma, Y. Chen, G. Zhou, H.-M. Cheng, *Natl. Sci. Rev.* **2022**, 9, nwa097.
- [79] M. Jiao, Qi Zhang, C. Ye, Z. Liu, X. Zhong, J. Wang, C. Li, L. Dai, G. Zhou, H.-M. Cheng, *Proc. Natl. Acad. Sci. USA* **2022**, 119, e2202202119.
- [80] W. Fan, Z. Ren, Z. Sun, X. Yao, B. Yildiz, Ju Li, *ACS Energy Lett.* **2022**, 7, 1223.
- [81] Y. Yao, Z. Huang, P. Xie, S D. Lacey, R. J. Jacob, H. Xie, F. Chen, A. Nie, T. Pu, M. Rehwoldt, D. Yu, M R. Zachariah, C. Wang, R. Shahbazian-Yassar, Ju Li, L. Hu, *Science* **2018**, 359, 1489.
- [82] R. A. Meidl, *Waste Management and the Energy Transition: The Path to Sustainability and a Circular Economy*, Baker Institute, Houston, TX, USA **2021**, <https://doi.org/10.25613/DJ5H-D776>.
- [83] AZO Materials, <https://www.azom.com/news.aspx?newsID=57992> (accessed: January 2022).
- [84] US Environmental Protection Agency, [https://www.epa.gov/system/files/documents/2021-08/lithium-ion-battery-report-update-7.01\\_508.pdf](https://www.epa.gov/system/files/documents/2021-08/lithium-ion-battery-report-update-7.01_508.pdf) (accessed: July 2021).
- [85] H. Gao, Q. Yan, P. Xu, H. Liu, M. Li, P. Liu, J. Luo, Z. Chen, *ACS Appl. Mater. Interfaces* **2020**, 12, 51546.
- [86] L. Gaines, Q. Dai, J. T. Vaughey, S. Gillard, *Recycling* **2021**, 6, 31.
- [87] L. Lander, T. Cleaver, M. A. Rajaeifar, V. Nguyen-Tien, R. J. R. Elliott, O. Heidrich, E. Kendrick, J. S. Edge, G. Offer, *iScience* **2021**, 24, 102787.
- [88] B. N. Sorbom, J. Ball, T. R. Palmer, F. J. Mangiarotti, J. M. Sierchio, P. Bonoli, C. Kasten, D. A. Sutherland, H. S. Barnard, C. B. Haakonsen, J. Goh, C. Sung, D. G. Whyte, *Fusion Eng. Des.* **2015**, 100, 378.
- [89] X. Feng, S. Zheng, D. Ren, X. He, L. Wang, H. Cui, X. Liu, C. Jin, F. Zhang, C. Xu, H. Hsu, S. Gao, T. Chen, Y. Li, T. Wang, H. Wang, M. Li, M. Ouyang, *Appl. Energy* **2019**, 246, 53.