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REVIEW

The Promise of 3D Printed Solid Polymer Electrolytes for Developing Sustainable Batteries: A Techno-Commercial Perspective

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Abstract

The year 1975 can be claimed to be the year of inception for the research and development of solid polymer electrolytes (SPEs) for Lithium-Ion Batteries (LIB), when the ionic conductivity of polyethylene oxide–alkaline metal ion complex was found by Peter Wright from the University of Sheffield. However, SPE research has undergone a leapfrog development, with conductivity values improving from 1×10^{-7} S·cm^{-1} to 1×10^{-1} S·cm^{-1}. The seed of development of SPEs spurs from the need for introducing design freedom to battery structures as well as the need for leak-proof electrolytes, greater operational safety, higher energy density, and other considerations. While the benefits of SPEs are evident, poor interfacial contact is a major factor limiting their application. This review presents the history of SPEs and shows how the additive manufacturing (AM) could prove beneficial for the improvement of performance and the functional implementation of SPEs. While the article articulates a technical review of additively manufactured SPEs, it also provides a lab-to-market perspective that could aid in shaping the future of green technology in energy storage. It also aims to provide an overall picture about the evolution and diversity of research advances in the development of greener SPEs through AM technology.

Keywords Solid-state battery · Additive Manufacturing · 3D Printing · Li-ion battery · Solid Polymer Electrolyte · Sustainability

1 Introduction

The industrial revolutions of various eras have largely been fueled by the accessibility of reliable and cost-effective sources of energy. With the electrification of a greater number of devices, the demand for electrical energy storage has continued to increase. Electrical energy storage offers a more dependable and eco-friendlier source of energy, especially for the storage of energy derived from renewable sources over long periods of time. Lithium-ion (Li-ion) batteries, which emerge to the current industry standard, are used in various applications such as wearables, textiles, electric vehicles, and Internet of Things devices [1–3]. Such advanced alkali-ion or alkali-metal batteries require high-performance electrolytes, but the standard liquid electrolytes used in these batteries pose safety concerns. Solid electrolytes have the potential to improve the energy density and safety of batteries. The rise of additive manufacturing (AM) or 3D printing has enabled the creation of functional electronics, including batteries [4–7]. Several companies are working to create 3D-printed batteries for large-scale adoption in order to offer a greener and more sustainable product. Despite the development of new generations of high-energy–density rechargeable batteries, Lithium-ion batteries remain the most popular. However, these batteries are reaching their performance limits. As society moves towards zero-carbon emissions, batteries with higher energy and power density, improved ecological footprint, and exceptional lifetime, reliability, safety, and environmental sustainability will be required. Achieving these goals may require the development of batteries that approach their theoretical limits.

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1.1 Why Battery Sustainability is Important?

Batteries have played a crucial role in the transition to green energy by eliminating emissions from fossil fuel technologies. However, the manufacturing process still depends on natural resource mining, which negatively impacts the environment. To promote sustainability, action is needed at every stage of a battery's life cycle, from the extraction of raw materials used in its manufacturing to the recycling of the battery at the end of its service life. Improving battery performance through the use of novel methods, by itself, is not enough to achieve sustainability; resource consumption must also be minimized. Some battery materials, such as copper and lithium, are expected to become scarce due to the annual consumption rates in the devices that use these batteries [8]. Therefore, adopting a circular economy approach could reduce environmental impacts and costs by recovering energy and converting material waste into by-products or energy to avoid the consumption of virgin materials [9–12]. The circular economy has already shown positive applications in the battery manufacturing industry with automobile companies such as Tesla, Toyota, etc. already partnering with Redwood Materials, a battery materials recycling company to reduce dependencies on mineral.

1.2 Challenges and Opportunities for Lithium-Ion Batteries as Green Technology

The energy storage capabilities of lithium-ion batteries (LIBs) have been challenged on various grounds, as some dispute the claims that LIBs are a green technology. The following paragraphs discuss the energy storage capacity, durability, costs, safety, and charging time of LIBs as well as the mileage range for vehicles that use LIBs.

Energy density and retention: The introduction of LIBs has motivated technological advances in electrochemical energy storage [13]. Due to their non-aqueous electrolyte systems, LIBs are able to maintain three times greater cell voltage levels than other energy storage technologies that use aqueous rechargeable chemistries [14]. However, only marginal improvements in the LIBs have been made. To date, nearly all of the specific energy improvements can be attributed to engineering improvements such as active material capacity and cell/electrode optimization [15]. The rate of performance improvement has slowed over the years, posing hurdles for the electrification of vehicles. The automotive industry has various cathode- and anode-specific targets to guide research on batteries [16, 17], and the United States Advanced Battery Consortium has material-level goals [17] that were translated from vehicle-level goals [18]. Since initial commercialization, cathodes for LIBs have been made from layered lithium metal oxides, which have seen improved material stability that enables them to tolerate higher voltages through coatings, dopants, and improves their operating conditions [19]. In parallel, the use of various metals (i.e., nickel, manganese, and aluminum) instead of cobalt has enabled a reduction in cost [20, 21]. A lithium-rich layered oxide is capable of meeting European Council for Automotive R&D standards for cathodes [16]. The LIB anode market has been predominantly captured by carbon compositions, which are barely able to meet the energy targets. Today, lithium and silicon are the most explored anode materials due to their potential to meet these targets. There remain numerous technical hurdles surrounding the utilization of lithium that will require significant engineering to be overcome. While carbon blends have been commercially used for years, silicon is a viable material to use for anodes [22, 23]. However, pure silicon hinders performance due to the large volume changes during cycling and the resulting lithium loss due to continuous solid–electrolyte interphase formation and particle isolation. Silicon as a system has the intercalation ability to tolerate additional mass that results from the lithium storage capacity of the battery [24, 25].

Battery durability: Durability is the biggest success metric for batteries in electric vehicles (EVs). Original equipment manufacturers who offer warranties generally require strict quality control to avoid any inaccuracies of derived parameters of interest such as Coulombic efficiency and capacity [26]. Accelerated testing is able to confirm long-term battery degradation in a shorter frame of time, as compared to tests under actual operating conditions over the course of several months [27]. The two remaining challenges in battery testing are (1) to select a set of tests that can 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 the life of the battery (which is considered to be a "rollover" degradation process). To test the hypothesis that the degradation rate accelerates as a battery ages, some research groups [28–30] have developed a framework to predict the rollover of graphite–lithium nickel–manganese–cobalt oxide cells that exhibit two degradation mechanisms: (1) the consumption of cyclable lithium and (2) shuttle mechanisms that consume electrolyte solvent. Lithium plating is quantified separately using Coulombic efficiency measurements across multiple rates and temperatures [31]. In order to provide accurate service life predictions under real-world conditions, 3D electrochemistry models are needed to accurately represent the cell and pack designs and the corresponding degradation process [32–34]. The machine learning approach

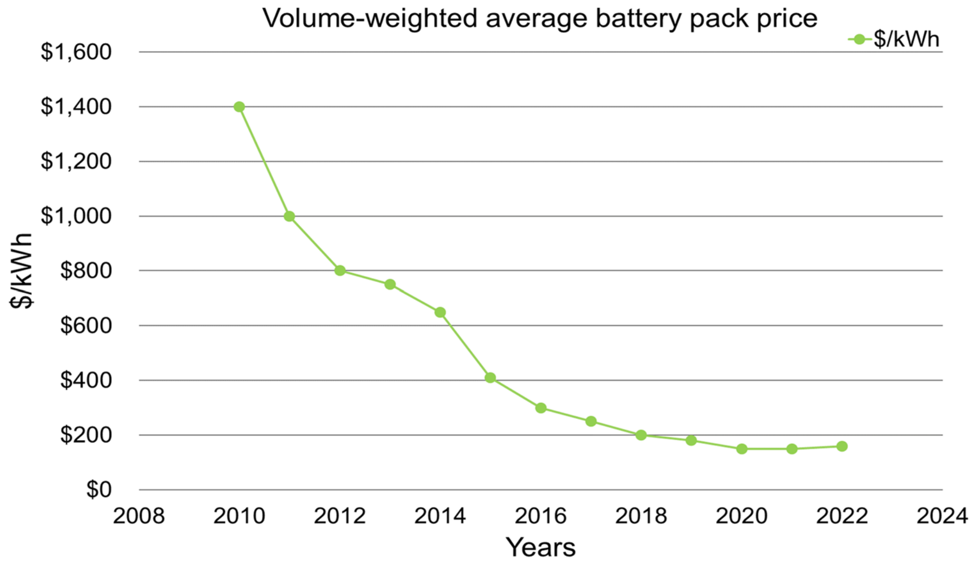


Fig. 1 Cost trend for LIBs from 2010 to 2022. Reproduced with permission [37] Copyright 2023, Volta Foundation

is a vital complement to physics-based data analytics, as it allows available data to be mined to support optimization modelling [35, 36].

Cost reduction: Cost reduction practices are often detrimental to the environment. However, the high cost of EVs as compared to the cost of internal combustion engine vehicles (ICEVs) is a major hurdle for the large-scale adoption of EVs. Battery manufacturing has undergone a major overhaul, with an 80% cost reduction over the past decade dropping from around $1300/kWh to $100/kWh as shown in the Fig. 1 [38,39]. Much of the cost reduction in recent years was realized due to optimization of current manufacturing processes, as process innovations that reduce energy consumption, manufacturing time, and capital expenditure all stand to make an impact [38 ,40, 41]. Most of the cost reduction for producing LIBs can be attributed to the swapping of 80% cobalt with nickel, as metals such as cobalt are costly and have significant supply chain challenges [42]. Complimentary cost reduction opportunities arise out of the indirect monetary benefits that arise from battery recycling, as electric commercial vehicles tend to have 40% lower maintenance costs [43], and lifetime fuel costs can be reduced by 60% with the use of electricity as fuel (depending on location) [44]. Although the greenhouse footprint for manufacturing EVs is higher than that for ICEVs [45], the emissions associated with the electricity supply mix are lower for EVs than ICEVs[46, 47].

Safety: The large-scale adoption of LIB technology has motivated significant research efforts on safety in automotive applications in terms of thermal [48, 49] mechanical [50,51] electrical[52, 53] and safety modelling and testing systems [54, 55]. Various regulations on LIB technology have been adopted by regulatory agencies in Europe [56] the Republic of Korea[57] and the United Nations [58, 59]. Electrolytes have been the focus of a global dialogue surrounding battery safety, as electrolytes are one of the main reasons why LIBs have experienced thermal runaway, causing vehicles to catch fire. Electrolytes, in spite of being electrochemically inert, are made from volatile and flammable organic solvents, salts, and additives, and their safe operating temperature is below 80 °C[55, 60]. The separators that serve as a barrier between the electrodes are very thin (perhaps 10–30 µm in thickness). Cathodes, on the other hand, actively undergo phase transformation and oxygen release in the overcharging state, which has a significant impact on the battery's thermal stability [61]. Lithium dendrite formation on the anodes is problematic, as lithium dendrite is able to break the contact between the active materials, and it can pierce the separator (causing a short circuit and thermal runaway). Although these issues have been mitigated, traces of dendrites can still be found in the edges of graphite anodes [62]. One way to deal with this issue is to accelerate the discovery of novel materials for electrodes and electrolytes using techniques like Material Acceleration Platforms (MAPs) which is discussed under Sect. 1.3.

Charging time and mileage range: Two of the key parameters determining the success of any EV are charging time and mileage range. Direct current fast charging allows up to 80% state of charge at 350 kW in about 40 min that can deliver a range of a few hundred miles. The United States Department of Energy is aiming to deliver fast charging that can provide a range of 200 miles in 7.5 min of charging [63]. LIBs have high energy because of the low electrochemical potential for lithium-ion intercalation in carbon. However, this leads to unintended lithium plating (in which metallic lithium forms around the anode of the battery during charging) due to negative electrode potential that is close to that of lithium which can lead to performance loss, capacity fade, and an increased risk of short circuiting [64].

Performance optimization is one of the main strategies towards achieving the green revolution promised by the adoption of LIBs. The success of this revolution depends on the development of batteries that incorporate high-performance electrolytes with a high degree of compatibility between the anode and cathode. As liquid electrolytes are highly volatile, they may experience thermal runaway—especially at elevated temperatures. Hence, there exist strong motivations for replacing the liquid electrolytes that are currently used in batteries for Internet of Things devices, electric vehicles, and other devices.

1.3 The Future of Sustainable Batteries

The European Commission has set a goal to reduce greenhouse gas emissions by 55% and transition to a carbon–neutral society by 2030, which requires advanced batteries with improved energy and power density as well as excellent longevity, reliability, safety, and environmental sustainability. In addition, the production of batteries must be scalable, sustainable, and cost-effective. BATTERY 2030 +, a large-scale and long-term European research initiative, aims to address these challenges and guide the development of batteries of the future through a chemistry-neutral approach whose aim is not to only develop specific battery chemistry but to provide a generic toolbox for future to transform the way we develop and design batteries.

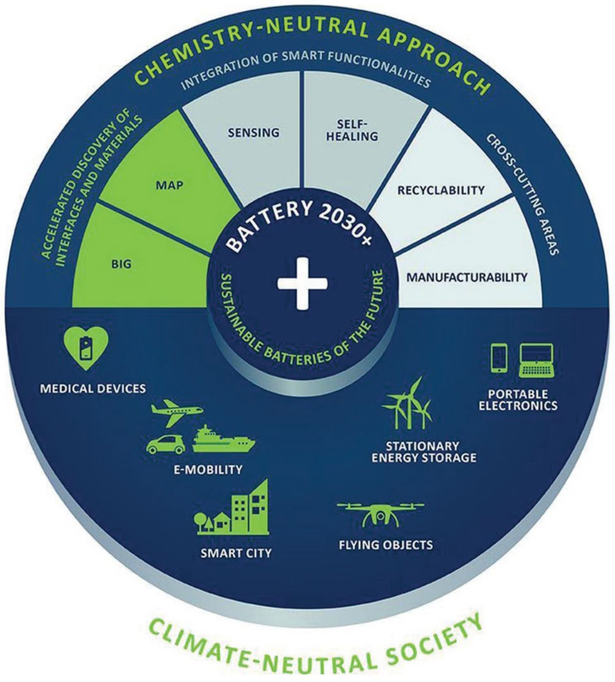


Fig. 2 Developing sustainable batteries of the future through synergy between a climate-neutral approach and a chemical-neutral approach [65]. Copyright 2022, Wiley

An accelerated path to a climate-neutral society can be achieved through three chemistry-neutral approaches: (1) accelerated discovery of battery interfaces and materials, (2) integration of smart functionalities, and (3) cross-cutting areas (Fig. 2). These goals align with the roadmaps published by several organizations and nations, including the European Association for Storage of Energy, the Energy Materials Industrial Research Initiative, the European Council for Automotive R&D, the Joint Research Centre of the European Commission as well as national roadmaps developed in China, Finland, India, Japan, and the United States [65].

1.3.1 Accelerated Discovery of Battery Interfaces and Materials

The development of high-performance materials and components is essential for creating batteries with higher energy and power. The BATTERY 2030 + initiative proposes the creation of a materials acceleration platform (MAP) for batteries that will utilize high-throughput automated synthesis and characterization, materials and interface simulations, autonomous data analysis and data mining, and artificial intelligence (AI) and machine learning to accelerate the discovery and optimization of battery materials, interfaces (which play a critical role in battery reactions but are not well understood), and cell designs. BATTERY 2030 + proposes to develop a battery interface genome (BIG) to understand and optimize these processes. A shared European data infrastructure can be developed to acquire, handle, and analyze data from all aspects of the battery development cycle. AI-based tools and physical models can be used to analyze large amounts of data generated by the various approaches. The integration of BIG and MAP (each of which is discussed in greater detail below) is expected to result in a significant increase in the speed of new discoveries for developing safe, long-lasting, and sustainable ultra-high-performance batteries [65].

Materials Acceleration Platform (MAP): The development of advanced materials is crucial for clean energy technologies, particularly for emerging battery technologies. Traditional trial-and-error–based development processes for high-performance battery materials and cell designs are expensive and time-consuming; and more than a decade can pass from the initial discovery to commercialization. BATTERY 2030 + proposes the creation of a battery MAP to accelerate the discovery and optimization of battery materials, interfaces, and cells. The proposed BIG-MAP infrastructure is modular and versatile, accommodating all emerging battery chemistries, material compositions, structures, and interfaces [66]. MAP utilizes AI to integrate and orchestrate data acquisition and utilization from various approaches and technologies such as computational materials design, modular and autonomous synthesis, robotics, and advanced characterization. The integration of all MAP elements will enable AI-orchestrated and fully autonomous discovery of battery materials and cells with unprecedented breakthroughs in development speed and performance [67]. The creation of autonomous, self-driving laboratories capable of designing and synthesizing novel battery materials and of conducting and interpreting experiments on the fly will create an efficient, closed-loop materials discovery process [68]. Its implementation will require the integration of all relevant European expertise.

Battery Interface Genome (BIG): In addition to the interface between the electrode and the electrolyte, batteries also have interfaces between other important components (such as the current collector and the active material). To master and design these interfaces, there is a need to combine their characterization in both time and space using physical and data-driven models—including models to study ion transport mechanisms and the role of electrons in interfacial reactions. Control of interfacial reactivity can help extend the stability of the battery; otherwise, it can limit the battery's cycle life. Interphases are complex, as they involve multiple reactions and processes that span a range of time and length scales, and their properties depend on various factors such as the composition of the electrolyte and the external conditions. Therefore, understanding, controlling, and designing the function of these interfaces and interphases [69] is essential for developing high-performing, sustainable batteries. The concept of BIG is based on the idea of catalyst design [70] where the binding energy of the reaction intermediates is related to a descriptor, and the identification and measurement of the descriptor value can help inform the design of the materials used for the catalyst. To achieve this for battery interfaces and interphases, which are complex phenomena with multiple interrelated factors, will require the use of multi-scale modeling, AI, and high-throughput characterization techniques to generate comprehensive and accurate data sets. The goal of BIG is to establish a fundamental understanding of battery interfaces and interphases across various chemistries, from established lithium-ion technology to other emerging technologies.

1.3.2 Integration of Smart Functionalities

The development of smart sensing and self-healing functionalities is essential to improve the safety, reliability, and cycle life of batteries. Despite the inevitability of battery degradation, efforts can be made to alleviate external factors such as extreme temperatures, mechanical stress, or aging in order to enhance sustainability, economic efficiency, and reliability. Achieving this requires that smart sensing and self-healing capabilities be incorporated into batteries that would enable non-invasive sensing technologies with spatial and temporal resolution to be used to monitor key parameters, identify defective components for self-repair, and observe reactions during real-world operation as well as to detect early signs of battery failure, thermal runaway, and aging. This particular approach focuses on developing self-healing functionalities in batteries to realize predictable lifetimes and documented states of health and safety, thereby improving acceptance rates in primary and secondary applications.

Sensing: The development of on-board [71] electrochemical impedance spectroscopy devices and battery management systems has had limited success for improving battery quality, reliability, and service life [35]. To enhance battery cycle life and longevity, direct temperature monitoring at the cell level is needed as well as an understanding of physical parameters and parasitic chemical processes to improve knowledge of battery science. Battery state of charge estimation has been a longstanding problem, and although various monitoring methods have been developed over the years, there is still no accurate solution. Most of the sensing activity relies on sensors located outside the battery cells that provide only limited information regarding the macroscopic properties of the battery. Recent research has focused on implantable sensors, with fiber Bragg grating sensors and other sensors showing promise for accurately monitoring the temperature, pressure, and strain upon cycling to support imaging of the cell temperature and estimation of the battery state of charge without interfering with cell performance. While implementation of sensing technologies for battery modules and systems has been attempted, challenges remain for the various types of sensing technology used. These obstacles must be overcome in order to make non-invasive battery sensing a reality.

Self-healing: The quality, reliability, lifetime, and safety (QRLS) of rechargeable batteries depend on the electrochemical and chemical ageing processes in the battery cell. To improve the QRLS of the cell, the first step is to detect irreversible changes [72]. The self-healing functionalities built into the cell, which can be autonomous or non-autonomous [73] can significantly contribute to the QRLS of the cell. The self-healing functionalities are in addition to preventive functionalities such as designed interfaces and optimized materials. Different self-healing functionalities have different kinetics, depending on the material or energy that is transported to the site of damage [35]. These functionalities should be developed alongside sensors and integrated into the cell/packs along with advanced battery management systems. To date, only a limited number of self-healing approaches in the battery field have benefited from the general strategies and formalisms that are well established in nature. Battery scientists have developed polymers having intrinsic self-healing properties based on dynamic supramolecular assembly, such as hydrogen bonding, electrostatic crosslinking, and host–guest or van der Waals interactions [74, 75].

1.3.3 Areas of Convergence

The battery of the future will prioritize sustainability and circular economy principles, including life cycle assessment [70] to ensure efficient manufacturability and recyclability at an early stage. Collaborative efforts in materials discovery and development, manufacturing, and recycling will drive the development of new battery technologies. The Fourth Industrial Revolution (Industry 4.0) [76] and digitalization will be leveraged to optimize cell designs and manufacturing methodologies through modeling and AI [77], while new recycling concepts (such as reconditioning and reusing active materials and electrodes) will be explored to reduce waste. The feasibility of scaling up new materials and battery cells, in addition to recycling and reusing components at low cost and using climate-neutral approaches, will be a key consideration.

Manufacturability: The manufacturing of future battery technologies needs to be viewed through the lens of Industry 4.0 and digitalization, as it involves integrating the outcomes of efforts to achieve BIG-MAP, self-healing, and sensors into a holistic process for manufacturing battery cells [78]. The use of modelling and AI can enable the creation of digital twins for innovative cell designs and manufacturing processes, which would minimize the need for traditional trial-and-error experiments. By employing fully digital manufacturing techniques, it will be possible to optimize process parameters and determine their impacts on the intermediate and final product. These virtual models can then be used to improve control over battery manufacturing facilities and production lines.

Recyclability: To accommodate new materials and cell architectures, new recycling concepts [79] such as reconditioning or reusing electrodes will be required, and this requires input from and coordination between material suppliers, cell and battery manufacturers, main application actors, and recyclers. Sustainability, dismantling, and recycling will be considered in battery designs and manufacturing processes, enabling a circular economy with reduced waste, a small CO_2 footprint, and the intelligent use of strategic resources. Design for recycling will be integrated into the algorithms for automated materials discovery, with input parameters such as the criticality of raw materials as well as raw material toxicity and socioeconomic aspects. Since the European Union implemented Batteries Directive 2006/66/EC [79], the battery recycling industry has significantly evolved. This directive established extended producer responsibility, forcing battery producers or third parties to finance the costs of recycling battery waste. The European Union also issued supporting/guidance documents and recycling efficiency regulations. Recycling methods are classified based on the battery chemistry and process used. The most widely applied recycling method for battery waste is pyrometallurgy, where battery components that have been fragmented by physical means and sorted before they undergo heating to extract/purify metals.

2 Solid Polymer Electrolytes for sustainable Batteries

Different methods and strategies have been explored to enhance battery performance. The modification of electrolytes is broadly accepted and has been explored as one way to improve the current performance levels of LIBs. Modifications by incorporating additives help induce the formation of solid electrolyte interface layer at the active material/electrolyte interface [80–87]. In a solid polymer electrolyte (SPE), a specific class of polymers is considered as an alternative to the currently used organic liquid electrolytes, as the polymers in this class have superior mechanical properties and electrochemical stability [88]. However, SPEs exhibit low ionic conductivity at room temperature and have high contact resistance.

An SPE can be defined as a solid-state membrane with a moderate to high ionic conductivity ($\geq 10^{-4}$ S·cm^{-1}) at room temperature. The ionic conductivity of this class of polymers was first demonstrated in 1973 by Peter Wright's group at University of Sheffield using polyethylene oxide (PEO) blended with an alkali metal salt [89]. The first practical demonstration of an all-solid-state battery was published by Armand et al. [90]. In 1991, Armand reported a Li-ion conducting electrolyte made using a novel salt, lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), on a single ion conductive polymer [91]. Since then, electrolytes that incorporate various transport ions (H +, Li +, Na +, K +, Ag +, Mg2 +, etc.) have been reported. Various theoretical approaches have been applied to understand the ion transfer process as well the physical/chemical processes occurring at the polymer electrolyte/electrode interface [92, 93].

Generally, the SPE functions as an ion-conducting medium between electrodes while serving as an electronically insulating layer. Some of the common selection criteria to be considered for a sustainable SPE are as follows: high ionic conductivity and lithium-ion transference number; a wide electrochemical window to ensure stability at the electrode/electrolyte interface; high mechanical stability to ensure good processibility as well as to curtail dendrite growth; minimal electrode/electrolyte resistance that aids in reducing polarization during battery operation; good electrical insulation to prevent short circuits and self-discharge;

and other characteristics such as low cost, ease of manufacturing, and environmentally friendly production. These selection criteria act as the chief motivations for advancing solid polymer electrolytes toward commercialization. In the following sections, we will discuss broadly studied polymer electrolytes and how some of these electrolytes can be used to leverage additive manufacturing processes (i.e., 3D printing) to produce sustainable batteries in the future [93–96].

2.1 Types of Polymer Electrolytes

Since the discovery of ion-conductive polymer electrolytes in 1973 [89] , the prime objective of researchers has been focused on enhancing the ionic conductivity of the polymer electrolytes—along with the other properties mentioned in the previous section—so that their conductivity would be close to those for liquid/aqueous electrolytes. To accomplish this, it is necessary to understand and deconstruct the effects of the different parameters on the performance of the polymer electrolyte. Currently, various articles have appeared in the literature that describe studies on different classes of SPEs. The most fundamental and comprehensive classification of SPEs is based on their physical properties and the methods used for their preparation [97, 98]. Based on the studies in the literature, SPEs can be classified into the following five broad groups.

Conventional polymer-salt complex/dry SPEs: Conventional SPEs are prepared by dissolving ionic salts in a polar polymer host such as polyethylene oxide (PEO) or polypropylene oxide (PPO). PEO and PPO are chosen as polymer hosts mainly because they are able to form a stable dry complex that exhibits a relatively high ionic conductivity [99]. The stability of the SPE is improved when the lattice energies of the polymer and the inorganic salt are low. A higher ionic conductivity is achieved when the ratio of salt to ethylene oxide is lower. However, in general, as the concentration of the lithium-ion salt increases, both the conductivity and the lithium-ion transference number decrease [100].

Plasticized polymer salt complex: A common method used to improve the performance of the SPE is to mix a large amount of a liquid plasticizer, such as polyethylene glycol (PEG) and/or aprotic organic solvents (such as dimethylsulfoxide) of low molecular weight, with the dry SPE matrix. This not only reduces the degree of crystallinity but also enhances the movement of the polymer chains. Additionally, the addition of plasticizers may also promote ion dissociation and, thus, increase the number of ions available for charge transport [101]. Plastic crystals can also be considered as a type of solid plasticizer: one example is succinonitrile (SCN), which has a plastic crystal phase between its crystal melting temperature of − 388 °C and its plastic crystal–isotropic melt transition at 58.8 °C [102]. Long and coworkers were the first to show fast ion conduction in molecular plastic crystals containing SCN plastic crystals and lithium salts [103]. However, these SCN/lithium salt mixtures have a paste-like appearance and lack mechanical strength [104, 105]. Later, plastic crystalline SCN was used as a solid plasticizer in the polymer electrolyte membrane (PEM) containing PEO/lithium salt composites, where PEO serves as the matrix [106–108]. Recently, Echeverrie et al. successfully fabricated completely amorphous plasticized solid polymer electrolytes under the guidance of ternary mixtures of PEO/SCN/LiTFSI [102]. They demonstrated that at room temperature, the ionic conductivity of some completely amorphous compositions can reach the level of superionic conductors, with a conductivity as high as 2.9×10^{-3} S·cm^{-1} for a 35/35/30 PEO/SCN/LiTFSI composition. Amorphous SCN not only plasticizes the polymer matrix, but it can also ionize the LiTFSI salt. Since PEO is a linear polymer, these completely amorphous PEMs can form self-standing films, but the films are fragile and have low mechanical strength and modulus. To address these issues, photo-crosslinking strategies have been adopted to fabricate various polymeric electrolyte membranes to provide strong mechanical support and flexibility to the PEM composites.

Gel polymer electrolytes: The addition of liquid plasticizers can enhance the ionic conductivity of PEM by effectively preventing crystallization and/or reducing the glass transition temperature of the host polymers in gel-type electrolytes. Aprotic organic solvents such as ethylene carbonate (EC) [108–110] and propylene carbonate (PC) [111, 112] are commonly used liquid plasticizers, but oligomeric polyethylene glycols and their derivatives[113–115] and phthalate derivatives [116] can be also used for this purpose. In general, the ionic conductivity of the PEMs is reported to reach the level of 10^{-5} to 10^{-4} S·cm^{-1} by using liquid plasticizers [111]. However, the reintroduction of liquid plasticizers in PEMs can negatively impact battery safety, and the thermal stability of electrolytes containing liquid plasticizers has been a concern, as evaporation of the liquid plasticizer during annealing in vacuum can lead to a rapid decline in ionic conductivity, as noted by Nicotera et al. [117]. Although this may not reflect the actual conditions in battery operation, the thermal stability of such gel-type electrolytes should be closely monitored, especially when operated under vacuum.

Rubbery electrolytes: Angell and co-workers introduced the concept of a “rubbery electrolyte,” which is a new type of polymer electrolyte system created by mixing different lithium salts into polymer hosts such as PEO and PPO. This polymer electrolyte is known as the “polymer-in-salt” system and has a high concentration of salt and a small amount of polymer host [118], unlike conventional “salt-in-polymer” systems where the concentration of salt is lower. When a small amount of polymer is added to the salt mixture, it forms a rubbery material with a low glass transition temperature; however, the salt tends to crystallize at lower temperatures, which affects the electrochemical stability of these electrolytes and limits their application. There remains a lack of understanding of the mechanism for ion transport in rubbery electrolyte systems, and it is believed that the high degree of ion clusters and their transport through the bulk material are responsible for the high ionic transport in polymer-in-salt systems [119–121].

Composite polymer electrolytes: The major challenges in the operation of lithium polymer batteries involve the reactivity at the Li/Li$^+$ interface and the growth of a passivation layer, leading to an increase in internal resistance. Safety concerns are another major issue. Recent studies have suggested that these issues can be mitigated by using composite solid polymer electrolytes (CSPEs), which are solid polymer electrolytes dispersed with nano or micro-sized filler particles of inert ceramic materials. The use of these particles in SPE host materials improves the morphological, electrochemical, and mechanical properties of the SPE membranes [122–125] and provides improved electrode/electrolyte compatibility and safety. The effects of various types of ceramic filler particles on the properties of CSPEs have been studied by several researchers [126–133].

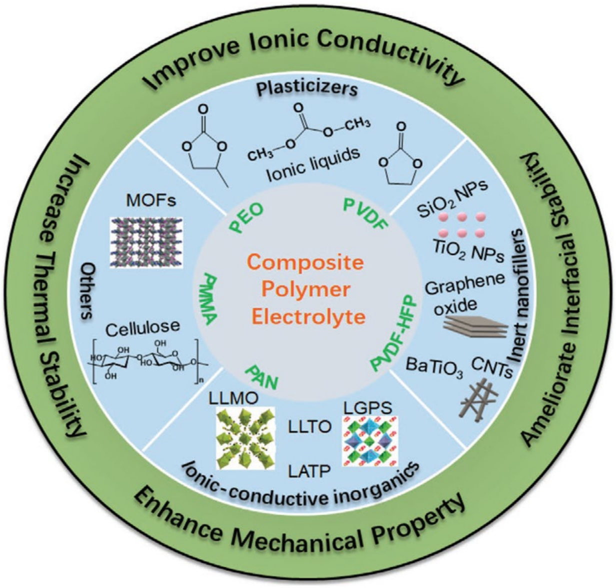


Fig. 3 Diagram showing the composite polymer electrolytes that contribute to various battery properties [148] Copyright 2020, Wiley

2.2 Composite Polymer Electrolytes as a Sustainable Choice

Polymer electrolytes consisting of a high-molecular-weight polymer matrix and dissolved lithium salt are advantageous in that they are flexible and adhere well to electrodes. However, they have a low ionic conductivity (from 10^{-4} to 10^{-6} S·cm^{-1}) and low lithium transference numbers (ranging from 0 to 0.3) at room temperature. Thus, PEO-based polymer electrolytes are considered a preferred option for lithium batteries. Generally, ion transport in polymer electrolytes is thought to be influenced by the relaxation of the polymer [134–139]. To achieve a high ionic conductivity, a high rate of polymer relaxation is required, which has not yet been achieved in dry polymer electrolytes at room temperature. The types of polymer electrolytes can be alternatively grouped into two subtypes based on the type of additive used for the improvement of ionic conductivity: composite polymer electrolytes and quasi-solid-state electrolytes. Composite polymer electrolytes (Fig. 3) are electrolytes in which the inorganic ceramics are dispersed as fillers in an ion-conductive polymer host. Various research efforts have been devoted to the study of polymer-rich electrolytes (known as ceramics in polymer), which are typically based on PEO. These electrolytes exhibit easy film processibility and, in some cases, ionic conductivity values are observed to increase incrementally at room temperature when inactive ceramics are added in small amounts. This enhancement is attributed to small amounts of ceramic particles that can curtail the polymer crystallization [135–140]. An alternative method that is gaining attention is the use of polymer-in-ceramic electrolytes, which have a significant ceramic component. This approach is considered a promising way to improve the development of all-solid-state batteries. Such electrolytes allow for efficient ion conduction through the ceramic material and its interface [141–147].

Quasi-solid-polymer electrolytes (QSPEs) include a wide variety of composites consisting of liquid electrolyte and polymer matrix. Plasticized polymer electrolyte and gel polymer electrolytes (GPEs) are similar, and the main difference between them is the concentration of plasticizer: a plasticized polymer electrolyte has a low concentration, while a GPE has a higher concentration. Since they exhibit similar properties, the distinction is not always clear. QSPEs comprise a spectrum of composite compounds classified as either GPEs or ionogel polymer electrolytes. QSPEs have widely varying mechanical properties that range from an absolute solid to a paste-like form. Polymer hosts include PEO, polyacrylonitrile, poly(methyl-methacrylate), poly (vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) and polyethylene glycol diacrylate (PEGDA), among others. Succinonitrile (SCN), ethylene carbonate (EC), polyethylene glycol (PEG) and diethyl carbonate (DEC) are commonly used plasticizers/solvents used as GPEs [149–152]. Ionogels are materials made of ionic liquids and a polymer or inorganic nanomaterials, which can conduct ions through a solvent or a softened polymer. Ionogels have a solvation structure and conduction mechanism that are like those of high-concentration liquid electrolytes, and the addition of inorganic nanomaterials are known to improve the mechanical properties and interfacial stability. Ionogel Solid Electrolytes are ionic liquids confined within ceramic scaffolds that can be inert or a superionic conductor [153–155].

2.3 Recyclable Polymer Electrolytes

Various polymers can be used for making polymer electrolyte membranes. For example, some studies report the use of PEO [156] or polyvinyl chloride (PVC) for producing PEM, while others report the use of polyvinylidene fluoride (PVDF) [157, 158] , polymethyl methacrylate (PMMA) [159, 160], and polyvinyl alcohol (PVA) [161]. If PVC is buried in the ground, biodegradation is difficult, and environmental problems may arise, as it remains in the ground for a long time. In the case of PVC, it is usually used up when it is burned and treated. However, even if PVC is burned completely, some amount of microplastic remains; these tiny particles of plastic are persistent organic pollutants that become concentrated in the tissues of living organisms [162].

Research on the use of PVDF as a PEM is being actively conducted. Since this material is also used as a binder for the cathode and anode in a battery, there will be no sense of alienation between the materials when positioned between them. In addition, the melting point of PVDF, which is a fluorine-based polymer, is about 177 °C (which is about 150 °C lower than that for polytetrafluoroethylene); since PVDF is dissolved in a polar solvent, it has excellent processability. It is strong in acids and bases as compared to other polymers due to its excellent chemical resistance, and it has excellent flame-retardant performance because it contains fluorine elements. It is also used as an insulator due to its large dielectric constant and low electrical conductivity. When a PEM made of PVDF is positioned between a cathode and an anode, it can effectively function as a separator. Therefore, various studies on the application of filters and membranes made of PVDF have been conducted [163, 164]. However, fluoropolymer has high resistance to various environments, and research on how to recycle this material is very limited. J. Wu and co-workers studied ways to restore membrane function when the PVDF membrane has been damaged by using a small amount of solvent and a catalyst [165]. In another study, by Zhao et al., the separation of all components in the lithium iron phosphate cathode was demonstrated through a three-step separation process [166]. In this and other studies, the PVDF used in the cathode was retrieved by dissolving in a N-methyl pyrrolidone (NMP) solvent [163–167].

Polymethyl methacrylate (PMMA) is a polymer that is non-toxic when it meets human skin, and it is a low-cost material. PMMA is very compatible with aprotic liquid electrolytes, has high ion conductivity, and has better anodic stability than a membrane made with PEO [168]. Unlike other polymers and condensation polymers, PMMA is difficult to recycle into its component monomers. Therefore, the monomer (methyl methacrylate) is obtained from PMMA through pyrolysis [169].

PVA, a water-soluble polymer with low polymerization stability that is typically made from polyvinyl acetate through saponification, has low toxicity and can be used to easily form a solid. Because PVA is a biodegradable material, it can be readily decomposed by Fusarium lini, a phytopathogenic fungus that penetrates the soil and lives in plants [170]. Alipoori et al. prepared a GPE using PVA [171] and they used a battery test to verify the ion conductivity and electrochemical stability of the GPE. However, a membrane made from this material is not easy to recycle, as it is difficult to separate the polymer used in the PEM, as combustion is required to separate the cathode and anode materials [172].

2.4 Biodegradable Materials for Polymer Electrolytes

To achieve a biodegradable polyethylene, numerous studies have been conducted on chitosan, polycaprolactone, and cellulose. Cellulose polymers, which are made by polymerizing several glucose molecules to form a glycosidic bond, are used as the raw material for natural fibers [173]. Many hydroxide groups in the monomer can be hydrogen-bonded with oxygen atoms in neighboring chains, thereby achieving sufficient tensile strength for the polymer [174]. The Du's research team studied the GPE of lithium-ion batteries using cellulose (which is biodegradable and is made from renewable sources) that is low in cost [175]. The results showed that the GPEs in these batteries achieved an ion conductivity of 6.34×10^{-3} S·cm^{-1}, and a membrane with good interfacial compatibility was manufactured. A cellulose nanofiber separator for lithium-ion batteries was studied by Chun et al. [176], who demonstrated that the cellulose-containing separator had better ion conductivity and electrolyte wettability than an existing polypropylene-based separator.

Jaishankar and coworkers produced solid polymer electrodes by blending chitosan and polyethylene glycol in various ratios to obtain ion conductivity of 0.46×10^{-3}·cm^{-1}, and they demonstrated that linear sweep voltammetry was stable up to 3.3 V [177]. The chitosan was polymerized with poly(ethylene glycol) diglycidyl ether (PEGGE) to synthesize a reusable and low-cost GPE with a 3D cross-linked structure [178]. The resulting material showed excellent lithium transference characteristics, good electrochemical stability to the lithium anode, and acceptable battery cycle stability.

In another study, a membrane for lithium-ion batteries was manufactured using an electron spun biodegradable polylactide with poly 3-hydroxybutyrate [179]. It was shown that each polymer has relatively low electrochemical stability but has high stability at a specific ratio and an ion conductivity of 14.7×10^{-3} S·cm^{-1}. More recently, a GPE was manufactured with a cellulose acetate/poly-L-lactic acid composite [180]. The ion conductivity and electrochemical stability of the composite were found to be excellent and, in particular, the GPE had a high initial discharge capability as compared to that of a Celgard separator.

2.5 Fabrication of Reusable Polymer Electrolytes

To recycle the polymer electrolyte membrane, the membrane must be well dissolved in a specific solvent or chemically replaced to another functional group. In addition, to facilitate its separation from the components of the battery, it should not have any side reactions with other substances. Piana and coworkers reported that about 5 tons per year of polyglycidol (which has a similar chemical structure to polyethylene oxide), can be recycled from the epichlorohydrin industry waste [181]. In addition, GPE was polymerized using this polymer, and sodium-ion batteries were fabricated to demonstrate renewable polymer materials. The use of water-soluble cellulose for GPE in lithium-ion energy storage devices was also studied [182]. It was shown that unused GPEs could be recycled using protic solvents.

2.6 Methods Used for Processing Electrolytes

Commonly reported and commercially used methods for thin film fabrication include radio-frequency sputtering, direct current magnetron sputtering, magnetron sputtering, thermal evaporation, and vapor deposition techniques such as pulsar laser deposition, electron cyclotron resonance sputtering, and aerosol spray coating [183]. Thick ceramic electrolytes are also synthesized from powdered materials by a hot-pressing method, in the form of pellets thicker than 0.5 mm. A dry-milling process is used to prepare fine powders, but these thick ceramic electrolytes are rigid and brittle, and they create poor interfacial contacts that lead to high battery impedance. Additionally, void space in the electrolyte and electrode/electrolyte interface can promote lithium dendrite growth, causing short circuits and mechanical degradation. Therefore, high pressure (5–70 MPa) is required for cell operation [184–197].

Solid polymer electrolytes can be manufactured through powder-based processing or wet-chemical processing. The powder-based processing involves dry ball milling and dry-pressing or isostatic hot-press processing like that used in the manufacture of inorganic solid electrolytes. This is a simple process, but it is time-consuming and is difficult to scale up. Wet-chemical processing involves dispersing powdered materials in a solvent, forming polymer electrolytes through solution casting, coating, or electrospinning. Wet-chemical processing is a high-throughput method but requires removal of the solvent. Gel electrolytes are produced using similar methods as polymer electrolytes, but the solvents are incorporated in the polymer matrix [189–193].

Carbon paste electrodes, which consist of polymer and inorganic components, are designed to achieve exceptional synergistic effects such as enhanced conductivity and improved interfacial properties [193–195]. Their manufacturing strategies include electrospinning, which produces interlaced and highly porous nanofibers with a large surface-to-volume ratio and improved mechanical strength [183, 195]. Limited mass loading of inorganic materials is typically used in composite solid electrolytes to prevent agglomeration or the potential deterioration of the ionic conductivity and mechanical strength of the electrolytes. A recent development in composite solid electrolytes is the use of electrophoretic deposition to create novel, thin-film solid polymer-in-ceramic electrolytes using a simple and inexpensive method [142,143, 147].

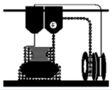
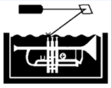
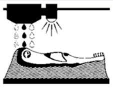
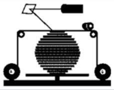
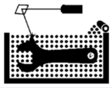
2.7 Societal Need for 3D Printing of Solid-State Batteries

The recent technological disruptions around the Internet of Things and artificial intelligence are the most obvious reasons for the increased implementation of wireless remote sensing operation and automation-based systems. These systems impose specific and stringent requirements on the design and properties of energy sources such as batteries. Conventional battery geometries are a major restriction: as products are designed around batteries, designers are forced to compromise a product's functionality in terms of the footprint, weight, operating range, capacity, and other aspects, hence making the end user application more sustainable.

Additive manufacturing is a promising method for creating battery components that can overcome limitations of traditional fabrication methods. This technique is particularly useful for creating solid-state electrolytes, which are now being used in various types of batteries. Printed solid-state electrolytes are easier to create using 3D printing methods than with liquid-infused separators. They are also less sensitive to environmental conditions and, as they have non-porous shapes, they can be used as a substrate for creating complete battery structures with arbitrary shapes [198–207].

Additive manufacturing (AM), involves the layer-by-layer deposition of materials to produce 3D structures based on a computer-aided design model [208–211]. Since its inception in the 1980s, various 3D printing techniques have been developed, and many unique frameworks have emerged in recent years. The American Society for Testing and Materials categorizes 3D printing techniques into seven groups (shown in Table 1): material extrusion, vat

Table 1 Classification of additive manufacturing processes

Material Extrusion		Technical Variants: Fused Filament Fabrication (FFF), Fused Deposition Modelling (FDM) ™ ,Direct Ink Write Material: Pellets and filaments of Polymer, Polymer Composites Bonding: Thermal curing, UV curing
Vat Photopolymerization		Technical Variants: Stereolithography Apparatus™ (SLA), Digital Light Processing™ (DLP) Material: Photopolymer liquid Resins Bonding: Laser and UV curing
Material Jetting		Technical Variants: Inkjet Printing , Aerosol Jet Printing, MultiJet Modeling™, Polyjet™ Material: Wax, polymer, photopolymers Bonding: UV curing, thermal curing
Binder Jetting		Technical Variants: 3D Printing Material: Powdered metal, ceramic, glass, sand, plastic Bonding: Thermal curing, adhesive bonding
Sheet Lamination		Technical Variants: Laminated Object Manufacturing, Selective Deposition Lamination, Material: Sheets of papers, metals and polymers Bonding: Thermal bonding using adhesives under heat and pressure.
Powder Bed Fusion		Technical Variants: Selective Laser Sintering ™, Selective Laser Melting ™, Multi jet Fusion ™ Material: Powdered metals, plastics, ceramic and sand Bonding: Thermally fused with laser and electron beam
Directed Energy Deposition		Technical Variants: Laser Metal Deposition, Laser Engineered Net Shaping ™, Direct Metal Deposition ™ Material: Metallic wire and powder Bonding: Thermally fused with laser

photopolymerization, material jetting, binder jetting, sheet lamination, powder bed fusion, and directed energy deposition [212]. Lanceros-Méndez and Costa, in their 2018 book Printed Batteries: Materials, Technologies and Applications [198], discussed methods for producing printed electrodes and current collectors. The relevant AM techniques for polymer electrolytes are direct ink writing, filament fabrication, stereolithography and material jetting. Despite the significant amount of research conducted, the available literature on the subject is limited and does not provide sufficient information to fully understand the relationship between the composition, properties, ion transport, and interface effects of materials printed using advanced 3D printing techniques. The scale of the part size and the resulting properties of the part depend on the 3D printing method and the material used for printing. In general, 3D printing provides the freedom to design, enables rapid prototyping, mass customization, and opens up new possibilities for application development [198, 213, 214].

3 Relevant Methods of Additive Manufacturing for Solid Polymer Electrolytes

Of the general techniques for 3D printing, four are appropriate for producing solid polymer electrolytes. These four techniques are described in greater detail below:

Material Extrusion: Direct ink writing (DIW) a material extrusion–based 3D printing method where a relatively viscous or gel ink is fed from a pressurized reservoir to a needle that ejects ink onto the printing substrate. It is typically used for continuous filament writing, which requires the ink to have specific properties such as shear thinning property of the ink, where the viscosity of the ink reduces under shear within the nozzle and holding shape quickly setting on the substrate holding after being extruded out of the nozzle. While inks for many types of active materials have been designed to meet these criteria, adapting inks from other fabrication methods to be used in DIW is not always a straightforward process [215]. Another method relevant here would be fused filament fabrication. This method involves using a heated dispenser nozzle to extrude semi-molten thermoplastic materials onto a substrate. The materials are then layered and cooled, forming a solid final product [216].

Vat photopolymerization (VPP): In this 3D printing method, solid objects are created by layers from a liquid resin. Light energy (e.g., laser or lamp) is used to solidify the resin by photopolymerization, and a stage moves a platform with the solid object to allow a new area to be covered with the resin. The resins used for VPP are photopolymers that react with photons. The resins used for printing battery components should incorporate both electrochemically active materials and printable resin, and they must be adjusted to meet the specific requirements for VPP printing [217].

Material Jetting (MJP): Generically MJP is a category of AM methods in which droplets of feedstock material. Aerosol jet printing (AJP) is sub type MJP relevant for printing PE’s. This technique involves using a carrier gas to break down a fluid into tiny droplets and depositing the droplets onto a printing substrate. The method produces even and thin layers of material. However, ink formulation can be difficult, as the ink must be able to be formed into small droplets, must have a specific viscosity and surface tension, and must be able to maintain a consistent particle size distribution without aggregation or settling [218].

3.1 3D Printable Polymer and Composite electrolytes

Although the materials used as polymer electrolytes are like those used in 3D printing inks, they may not be ideal for printing purposes. Various research groups have been working to modify the properties of these inks to make them both printable and ion conductive. Table 2 draws a contrast between 3D printed batteries and conventional type batteries.

The use of VPP to print a PEO-based polymer electrolyte was demonstrated by He et al. (Fig. 4a), in which UV-curable additives such as PEG-diacrylate, succinonitrile precursors, and lithium bis(trifluoromethanesulfonyl)imide salt were added to the ink. The ink was used to produce a functional 3D-printed PEO-based polymer electrolyte, which was tested in lithium iron phosphate/PE/lithium metal half-cells. The electrolyte exhibited a relatively good ionic conductivity of 3.7×10^{-4} S·cm^{-1} at room temperature and over 1.0×10^{-3} S·cm^{-1} at 50 °C. The ionic conductivity showed a typical Vogel–Tammann–Fulcher behavior for amorphous polyethylenes produced by casting [219]. In a different study, Chen et al. used a micro-stereolithography technique (Fig. 4b) and incorporated a liquid electrolyte into a photopolymer to produce a gel electrolyte [220]. This gel electrolyte showed better ionic conductivity at room temperature (4.3×10^{-3} ·cm^{-1} at room temperature, 80% liquid-electrolyte content),

Table 2 State of the art comparison of value proposition and competitive edge

Battery type	Design freedom	Volumetric, energy and power density	Manufacturing scalability	Hype cycle	Competitive edge
3D printed solid state batteries	High	High	Low	Peak of inflated expectation	Customizability and high performance
Conventional batteries	Low	Medium	High	Plateau of productivity	Scalability and low cost

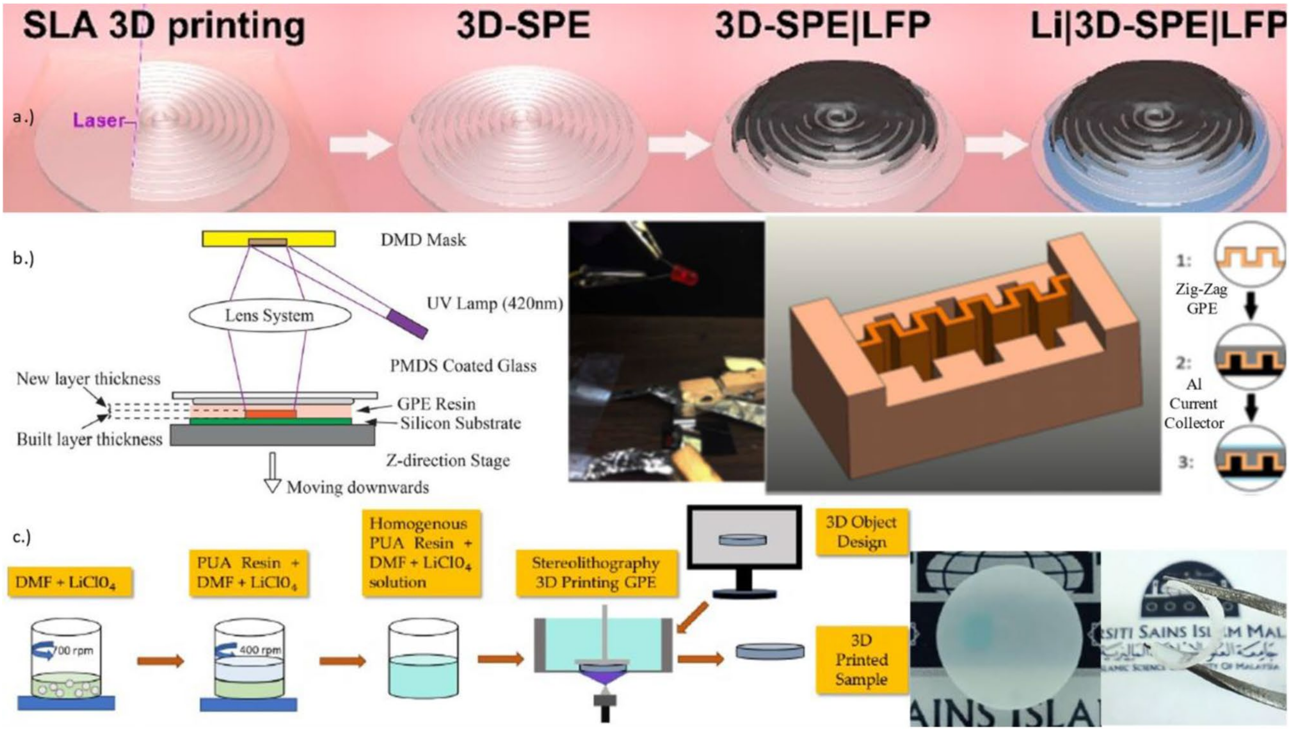


Fig. 4 VPP 3D printing of polymer electrolytes: **a** spiral polymer electrolyte Reproduced with permission [219] Copyright 2020, ACS Publications, **b** micro-scale interdigitated GPE, Reproduced with permission [220] Copyright 2023, IOP publishing **c**) coin-shaped GPE Reproduced with permission [221] Copyright 2022 MDPI, Basel, Switzerland

whereas the all-solid polymer electrolyte studied by He et al. was less durable and was effective for only a few cycles before the cell short-circuited [219, 220]. Recently, Norjeli et al. used a VPP method (Fig. 4c) to 3D-print a micro-scale GPE that exhibited an ionic conductivity of 1.2 mS·cm^{-1} at a low lithium salt content (10 wt%) in ambient conditions [221].

Deiner et al. used AJP to produce a PEO solid polymer electrolyte without a UV-curable system by using a suspension-based ink that was dried in a post-processing step. They tested two types of lithium salts, lithium difluoro(oxalato) borate and lithium trifluoromethanesulphonate, with the addition of alumina as a filler, and they achieved a conductivity of about 5×10^{-4} S·cm^{-1} at 80 °C. The most conductive printed electrolyte was able to achieve a conductivity greater than 10^{-5} S·cm^{-1} at 45 °C. The printed electrolyte showed either Vogel–Tammann–Fulcher or Arrhenius behavior based on the polymer-to-salt ratio. The printed electrolyte was used in lithium/lithium iron phosphate batteries, which had a 15-h discharge rate (C/15) with a capacity of 85 mAh/g at 45 °C and a capacity of 160 mAh/g at 75 °C [222].

Cheng et al. used a DIW method with a PVDF-based ink and a TiO_2 ceramic additive [199]. The solvent in the PVDF was replaced with an ionic liquid electrolyte before printing, which required a mild elevated temperature but no post-processing. The printed gel electrolyte showed better room-temperature ionic conductivity than most solid electrolytes but was not as high as $LiPF_6$-based gel electrolytes [223–225]. Li/MnO_2 cells with a DIW-printed electrolyte had an average Coulombic efficiency of 98.6% for over 100 cycles and a higher capacity than a cell with a cast electrolyte [199]. Wei et al. [226] used a DIW ink that incorporated all electrolyte components, which avoided the need for post-processing, but they used PC as the solvent and used UV-curing for the binder matrix after printing that required the entire process to be completed inside an argon-filled glovebox (see Fig. 5).

Three recent studies show that it is possible to use UV-curable gel electrolytes for 3D printed polymer batteries. In one study, Muench et al. [227] created inks made of an ionic liquid and a UV-curable binder and tested them with amorphous silica as the ceramic filler. The use of the UV-curable binder eliminated the need for high temperatures during the printing process. The resulting ionic conductivity of the gel electrolyte was very high (0.74 S·cm^{-1}), but it was used in an all-organic battery rather than a Li-ion battery. In another study, Cheng et al. [228] used the DIW method to print PEO-based UV-curable polymer-ionic-liquid composite electrolytes with added silane-treated hexagonal boron nitride thermal conductors to mitigate thermal runaway issues. The Li-ion battery using this printed electrolyte had a high specific discharge capacity and a stable Coulombic efficiency for 100 cycles at room temperature. Finally, Gambe et al. [229] demonstrated direct ink writing and curing using (LITFSI: EMI-TFSI): (HEMA: ethylene glycol Di-methacrylate) with Silica reinforcements as a rheological aid for shape retention post extrusion while exhibiting a room temperature ionic conductivity of 2×10^{-3} S·cm^{-1}. Other unconventional DIW methods that include the use of coextrusion carbon fiber composites with polyethylene shells for batteries and supercapacitors have also been reported [230–233].

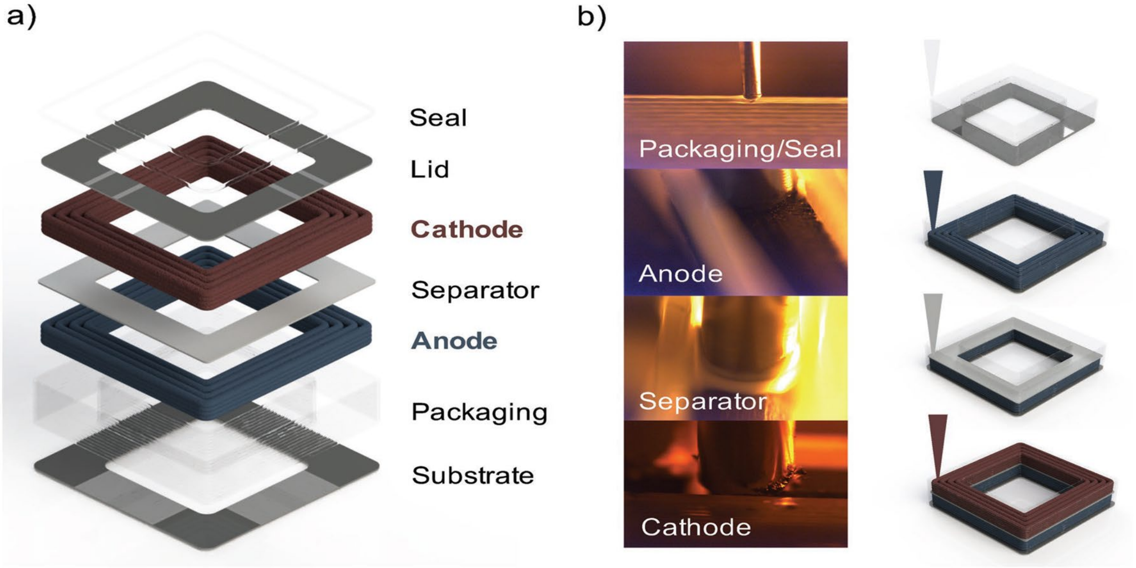


Fig. 5 Multilateral 3D printing using direct ink writing to print battery components: **a** schematic diagram of the design; **b** images and Process visualization of the 3D printed layers Reproduced with permission [226] Copyright 2018, Wiley

The FFF method, which includes solvent-free extrusion, is a promising method for manufacturing solid electrolytes for Li-ion batteries because it is fast, easily scalable, and environmentally friendly. However, it is currently not widely used in the continuous manufacturing of solid electrolytes for Li-ion batteries [234–236]. Most reports of FFF of electrolytes for Li-ion and lithium-metal batteries focus on the printing of polymer membranes, which are then infused with liquid electrolytes. One example is the use of polylactic acid (PLA) infused with $LiClO_4$ in PC/ethyl methyl carbonate (ECM), which had an ionic conductivity of 8.5×10^{-5} S·cm^{-1} and was able to retain its mechanical integrity and ionic conductivity after 24 h. The FFF technique has also been used to print a complete 3D battery with all components based on PLA. However, conduction in PLA is a major issue, and more research is needed to improve the dispersion of fillers in the PLA matrix [237].

SiO_2/PLA-based membranes containing 40% polyethylene glycol dimethyl ether with a molecular weight (MW) of 500 were 3D-printed by Maurel et al. [238]. They found that through SiO_2 addition, a more porous structure is formed, and this contributes to faster electrolyte uptake. The most conductive membrane was the one containing 7% SiO_2, and the ion transport was believed to occur mainly via polyethylene glycol dimethyl ether domains. Plasticizer addition was used to increase the lithium iron phosphate loading within the positive electrode to 52% wt while still maintaining flexibility for the filament to be printed. A 3D-printed lithium iron phosphate/carbon battery with a two-layered separator delivered a reversible capacity of 30 mAh/g at C/40, corresponding to 15 mAh/g for the total composite (or 6.5 mAh/cm^2 when considering the total volume of the battery).

Vinegrad et al. [239] used FFF to print polylactide-PEO blended membranes, in which the influence of the relative content of polymers on the ease of extrusion and printing were studied. Four polymer samples containing PLA: $_h$PEO (High MW = 5×10^6): $_l$PEO (Low MW = 2×10^3) of different proportions (25:40:35, 40:30:30, 40:40:20, 0:50:50)%(w/w) were extruded, and the sample containing 25% PLA appeared to be the most ductile and had sufficient mechanical strength. The calculated porosity of the neat PLA and the blended polymer samples was very low (not exceeding 4% for all compositions), indicating that the films are dense and contain few to no pores. In addition, ionic liquid N-butyl-N-methyl pyrrolidinium bis(trifluoromethanesulfonyl)imide with dissolved LiTFSI salt was infused into the membranes under vacuum, forming free-standing plasticized quasi-solid films. A complex interplay was found between the ionic liquid uptake and the Li-PEO/Li-PLA interactions, which influenced phase transitions, conductivity, and self-diffusion coefficients. The quasi-solid printed electrolyte showed higher bulk-conductivity values with increased PEO content and increased concentration of lithium imide salt. A comparison of the ionic conduction properties of quasi-solid-state 3D-printed electrolyte plasticized by ionic liquid of pristine PLA and PLA-PEO printed blends showed that neat PLA has a low bulk-conductivity of 7×10^{-6} S·cm^{-1}; upon being mixed with PEO, the ionic conductivity increased by almost 1.5 orders of magnitude, approaching 2×10^{-4} S·cm^{-1} at 60 °C.

FFF printing of all-solid-state electrolytes composed of LiTFSI, PEO, and PLA was performed in an inert atmosphere by Ragones et al. [240, 241]. In these studies, the printing process was complicated by the high ductility of the filament, but this was resolved by the addition of 1% SiO_2 or Al_2O_3 nanoparticles. It was found that the printing of the filament lowered the degree of crystallinity of the composite electrolytes but induced order in the crystalline entities. Time-of-flight secondary ion mass spectrometry analysis was used to find the lateral distribution of PEO, PLA, lithium salt, and ceramic additive. The conductivity values of the printed LiTFSI:(PLA-PEO) Al_2O_3- or SiO_2-containing electrolytes were found to be lower than those reported for neat LiTFSI:PEO electrolytes, but the bulk conductivity of the 3D-printed electrolytes was more than one order of magnitude higher than that of the cast electrolytes. In both these studies, the group was able to demonstrate a fully 3D printed solid state battery. Katcharava et al. [242] studied the influence of nano SiO_2 fillers at different concentrations to explore the 3D printability; however, the filler modified filaments showed very low ionic conductivity 10^{-5} S·cm^{-1} at 80 °C.

Maurel et al. [243] created a 3D-printable solid lithium-ion PEO filament using a PEO:LiTFSI mixture and fed it through a FFF 3D printer (Fig. 6). However, the filament exhibited poor mechanical properties, and the process of printing using this filament required modifications to be made to the printer. The extrusion process resulted in a compact filament microstructure, in contrast to the large spherulites and visible pores seen in cast films. The researchers used different sample holders to measure the conductivity, and they found that the conductivity varied depending on the orientation of the PEO chains, which was attributed to the extrusion process. This data is consistent with the results reported in previous studies.

High-performance solid-polymer and composite electrolytes that can be printed using the FFF method may open up new possibilities for creating a bipolar multilayer battery using a one-shot co-extrusion technique [244, 245]. This method could be applied in flexible batteries for electronic and wearable applications as well as for electric vehicles (where it would allow for effective integration of the battery not only at the bottom of the car but in other locations as well). The Bolloré Group (France) is mentioned as a pioneer in the production of thin-film electrodes using extrusion

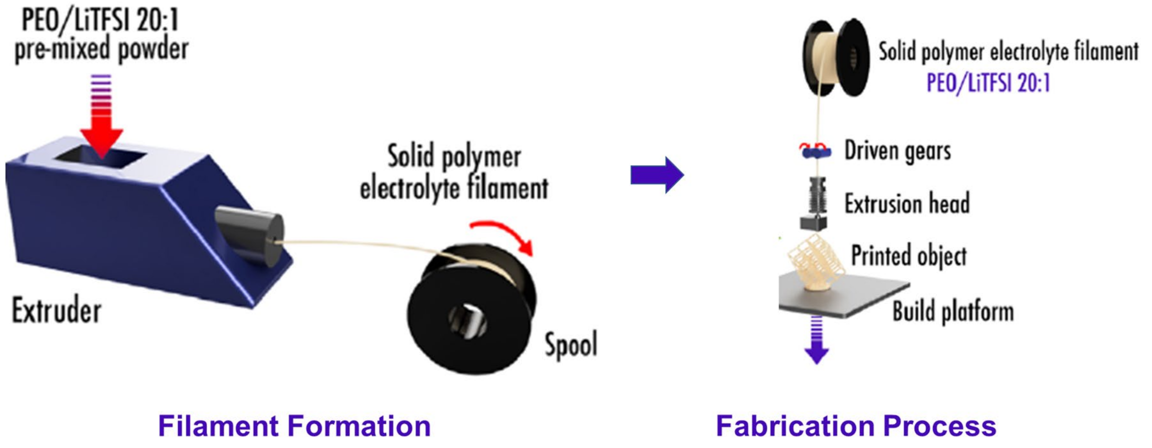


Fig. 6 Fused filament fabrication of a PEO/LiTFSi solid polymer electrolyte Reproduced with Permission [243] Copyright 2020, IOP science

Table 3 Quantitative comparison of 3D printed Solid Polymer Electrolyte Systems and their respective ionic conductivity values (RT: Room Temperature)

Formulation	Ionic Conductivity (S/cm)	Printing process	References
PEGDA/SCN/LiTFSI	3.7×10^{-4} @RT	SLA	[223]
	4.8×10^{-3} @RT	Micro SLA	[224]
Poly Urethane Acrylate: Lithium Perchlorate: Dimethyl Formamide	1.2×10^{-3} @RT	Aerosol Jet printing	[225]
PEO/LiDFOB/Al2O3(EO:Li = 10:1)	$>10^{-5}$ @ 45 °C	Aerosol Jet Printing	[226]
PVDF-Co-HFP + TiO_2 | Pyr13TFSi + LiTFSI	5.81×10^{-5} @RT	Direct Ink Write	[199]
PVDF + 1butyl-3-methylimidazolium-TFSI	0.74×10^{-3} @RT	Direct Ink Write	[227]
S-hBN/TEGDME | LiTFSI | 1-methyl-1-propylpyrrolidinium bis(trifluoromethyl sulfonyl)imide	0.47×10^{-3} @RT	Direct Ink Writing	[228]
(LITFSI: EMI-TFSI): (HEMA: ethylene glycol dimethacrylate) @ 7% SiO_2	2×10^{-3} @RT	Direct Ink Writing	[229]
PLA: $LiPF_6$:PC/EMC	1.7×10^{-3} @20 °C	Fused Filament Fabrication	[237]
PLA: $LiClO_4$:PC/EMC	8.5×10^{-5} @RT	Fused Filament Fabrication	[237]
PLA: LiTMFS: PC/EMC	3.9×10^{-5} @RT	Fused Filament Fabrication	[237]
(PLA: PEGDME 500): EC/DMC: $LiPF_6$	1.2×10^{-4} @RT	Fused filament fabrication	[238]
PLA (MW = 5×10^{6}): PEO(MW = 2×10^{3}): Pyr14TFSI + LiTFSI	0.2×10^{-3} @RT	Fused Filament Fabrication	[243]
PLA: PEO: LiTFSI	2.18×10^{-3} @90 °C	Fused Filament Fabrication	[247]

techniques. It is expected that such one-shot extruded/printed batteries will show significant improvement in discharge capacity, cycle life, and internal resistance as compared to mechanically assembled cells. To summarize Table 3 shows the quantitative comparison of different 3D printed solid polymer electrolytes.

The field of using printing techniques for advanced non-lithium batteries (sometimes referred to a *beyond-lithium batteries*) is a relatively new field and is still under development. Researchers have been exploring the use of printing techniques for different types of electrodes for non-lithium batteries but have yet to fully utilize printed solid electrolytes for this purpose. Studies have shown that it is possible to print high-performance sodium ion (Na-ion) batteries using methods such as drop-on-demand inkjet printing [246] and FFF [247], but challenges remain in terms of improving the robustness of the printing process and the electrochemical performance of the batteries produced using this method. Further research on battery design and ink compositions may lead to better results without the need to make significant changes to the printing system.

3.2 Challenges and Limitations Associated with Additive Manufacturing of SPEs

Despite the considerable research efforts in this field, there is a lack of comprehensive publications that thoroughly investigate the intricate interplay between composition, properties, ion transport, and interfacial phenomena associated with printing methods. One notable printing technique is Direct

Ink Writing (DIW), which utilizes a viscous gel ink fed from a pressurized reservoir to a needle for continuous filament writing. The ink must possess specific rheological characteristics that enable it to flow within the printing head components while rapidly solidifying on the substrate. Another technique, Stereolithography (SLA), employs photopolymerization to construct solid objects layer-by-layer using a container of liquid resin ink and a laser. SLA requires a light-curable binder system, and the inks used must incorporate both electrochemically active materials and printable resin. Aerosol Jet Printing (AJP) involves atomizing droplets from a fluid reservoir and depositing them as an unordered jet of minuscule droplets onto the substrate. Developing inks for AJP poses challenges in terms of droplet formation, viscosity, surface tension, and particle distribution. Additionally, the widely used Fused Deposition Modeling (FDM) or Fused Filament Fabrication (FFF) technique relies on heating and extruding thermoplastic materials in a semi-molten state, layer-by-layer, to fabricate solid products.

Traditional liquid electrolytes employed in Lithium-ion Batteries (LiBs) consist of Li salts dissolved in aprotic organic solvents, but their flammability and vulnerability to moisture pose safety concerns. As a result, researchers are exploring alternative electrolyte options, including liquid, polymeric, and solid-based electrolytes. Polymer electrolytes offer advantages such as ease of processing, resistance to mechanical deformation, and low flammability. They also exhibit improved interfacial contact and compatibility compared to inorganic solid electrolytes. However, polymer electrolytes generally demonstrate relatively low ionic conductivity at room temperature and exhibit enhanced performance at higher temperatures. Gel polymer electrolytes combine the mechanical stability of polymers with the ionic conductivity of liquid electrolytes. On the other hand, solid electrolytes offer enhanced safety, broader operating voltages, and high energy density for various applications. Nevertheless, achieving a uniform interface and minimizing contact resistance between electrodes and solid-state electrolytes remains a challenge. While gel electrolytes can achieve high ionic conductivity, solid-state, polymer-based electrolytes are preferred for improved durability. These solid-state electrolytes maintain their structure and exhibit good lithium-ion conductivity even after exposure to UV radiation, making them suitable for 3D-printed solid-state Lithium-ion Batteries (LIBs). Finally in terms of large-scale technology adoption, current state of the art both for 3D printing and solid state batteries needs optimization in ease of material processing and improvement of cycle time to compete with conventional batteries available in the market.

3.3 Opportunities

To enhance solid polymer electrolytes, there is potential in investigating alternative structures such as cross-linked polymers or unconventional polymer electrolytes that offer higher room temperature conductivities or improved mechanical strength compared to standard PEO electrolytes. Another unexplored area is the utilization of ceramic-reinforced polymer matrix composites and hybrid polymer/ceramic materials for 3D printing of solid-state lithium-ion batteries. Like nontraditional polymers, these composite materials aim to retain the favorable processability of polymers while enhancing their conductive properties and mechanical strength. Additionally, there is an opportunity to delve into the 3D printing of ceramic-based electrolytes, particularly in terms of post-deposition processing, enabling their incorporation into battery cells. Advancing knowledge in 3D printing and processing of ceramic-based electrolytes would open doors to exploring printed battery systems combining ceramic electrolytes with lithium metal anodes. Lastly, beyond the need to identify printing parameters and processes for a wider range of electrolyte materials, the ultimate challenge lies in pushing digitally printed solid-state batteries beyond the demonstration stage to showcase their performance and integration benefits. These benefits are expected to arise through micro- and mesoscale structuring of battery cell components and the macro-scale integration of printed batteries into the structural elements of the devices they power.

4 Commercial Landscape of Additive Manufacturing of Solid-State Batteries

Battery technologies can be classified liquid state battery and solid-state battery. The 3D printing battery technology is a subset of solid-state battery. After 2018, four new companies that produce 3D printed batteries have been established: Sakuú (United States), Blackstone Technology (Germany), Blue Spark Technologies (United States), and Photocentric (United Kingdom) [198, 248]. All of these companies are unique in their own way. With the advent of the fourth industrial revolution, innumerable battery technologies are being developed, with diversity in terms of the type of electrolyte (solid or liquid), the chemistry of the battery elements (cathode, anode, and electrolytes), and the manufacturing techniques (jelly roll construction, coin cell construction, or 3D printing). Surveying the competitive landscape is particularly important for identifying any unmet needs and for finding ways to develop disruptive technologies. In this case, it would be naïve to say that all or none of today's

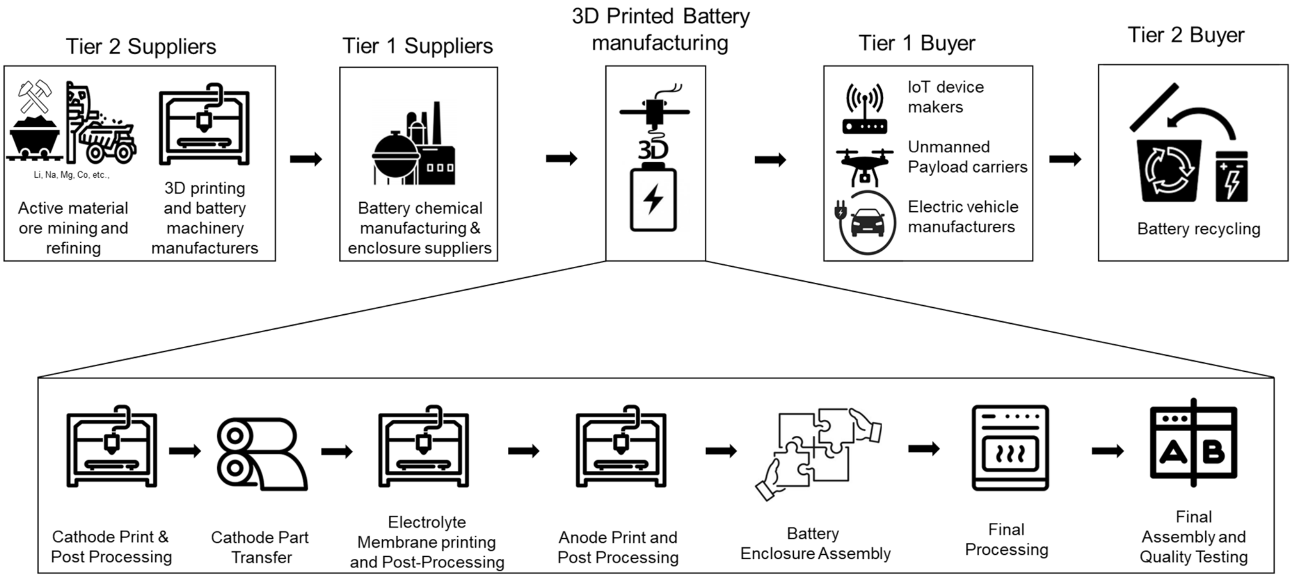


Fig. 7 An Illustration of the 3D printed battery manufacturing ecosystem.

battery technologies can be considered as competitors to the proposed technology. An outlook illustration of the 3D printed battery manufacturing ecosystem is shown in Fig. 7 and the following sections explain in detail about the key players and market potential surrounding this sector of battery manufacturing.

4.1 Additive Manufacturing

The 3D printing/rapid prototyping services industry has experienced rapid growth over the past five years, driven by technological advancements and increased applications of 3D printing technology. Although some companies have bypassed industry services by purchasing their own machines, the demand for these services has continued to grow due to the normalization of 3D printing. Over the course of the COVID-19 pandemic, the industry was able to continue operations without disruption due to remote development and design services and automated physical printing, which led to solid growth in 2020. In 2021, the market size was \$14 billion, which represents a growth of 15.6% in total over the five years leading up to 2021, with a growth of 8.6% in 2021, as the economy recovered from the pandemic. The performance of the 3D printing and rapid prototyping services industry is closely linked to R&D expenditures in the manufacturing sector, as 3D printing a cost effective, rapid, and less costly way to create prototypes than using traditional manufacturing processes for this purpose. In total, 25% of industry revenue was derived from outsourcing of the development/design of custom prototypes by their clients. As the growth of 3D printing brought down the costs for prototyping, operators were able to raise prices for their skilled services and target the consumer market with cost-effective customized 3D-printed products. Over the next five years, the revenue in the industry is expected to continue to grow as 3D printing and rapid prototyping become more accessible and the price of 3D printers decreases. The market size is forecasted to grow 21% from years from 2021 to 2026 and is expected to reach \$77.83 billion by 2030 [249].

4.2 Lithium Battery Manufacturing

The lithium battery manufacturing industry experienced growth in the five years leading up to 2020 due to the growth of the EV industry. However, this growth was impacted by offshoring trends in electronics manufacturing, particularly when Apple moved its lithium-ion battery supplier to China in 2017. This has resulted in an estimated 6.8% decline in revenue (to \$1.5 billion over five years) and a 9.7% drop in 2020 alone. The lithium battery manufacturing industry has faced a sharp decline in revenue due to the impact of the COVID-19 pandemic on supply chains and EV sales. Despite this, the industry remains strong and is expected to continue growing in the future, driven by partnerships such as Tesla Inc. and Panasonic Holdings Corp. as well as joint ventures such as that between Panasonic and Toyota Motor Corp. The cost savings from vertical integration are expected to make EVs more competitive, but the average profit margin has fallen due to lower demand. Despite this, the industry's profit margin has increased overall during this period. The lithium battery manufacturing industry is expected to grow over the next five years as major automakers seek to enter the EV market. Joint ventures—such as the partnership between General Motors Company and LG Chem Ltd. and the

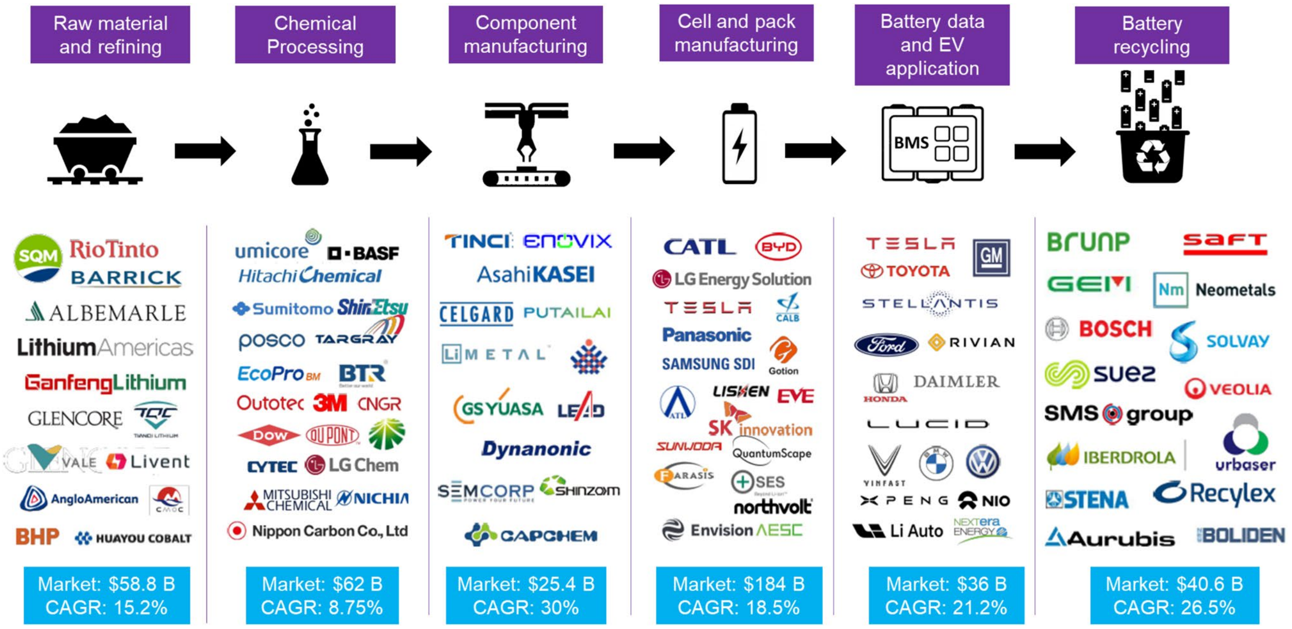


Fig. 8 EV battery manufacturing landscape of companies with a market capitalization greater than $1B Reproduced with permission [37] copyright 2023, Volta foundation

partnership between Panasonic and Toyota—are expected to contribute significantly to the industry's growth. The Volta foundation group summarized (Fig. 8) the supply chain, key players and corresponding market cap and growth rate. [37] However, lithium battery producers in the Asia–Pacific region are expected to maintain their dominance, limiting the growth of the domestic industry in the United States. Despite this, the industry is expected to see an annualized increase in revenue of 3.0%, reaching $1.8 billion over the next five years. The landscape of battery manufacturing sector consists of raw material mining, processing, component manufacturing, cell manufacturing, battery data and energy storage system and battery recycling ensuring a circular economy. The overall battery manufacturing industry revenue will also continue to increase, rising at an annualized 3.2% to reach $12.0 billion by the year 2026 [250, 251].

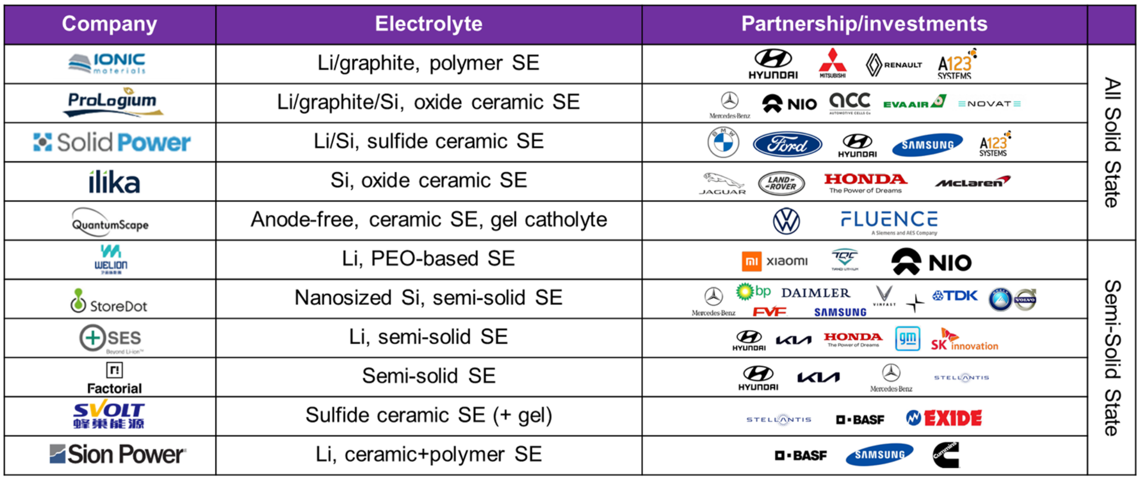
4.3 Production of Solid-State Batteries

The future production capacity of solid-state batteries is highly uncertain and depends on technological advancements and corporate strategies. Polymer solid-state batteries currently have a production capacity of less than 2 GWh, with expected growth to 2–15 GWh by 2025 and to 10–50 GWh by 2035. Sulfide solid-state batteries, which are not yet on the market at a large scale, are expected to have a capacity of 0–5 GWh in 2025, growing to 20–50 GWh by 2035. The oxide solid-state battery market is expected to be smaller in size, with a capacity of 0–1 GWh in 2025, growing to only 10–20 GWh by 2035. The global market for lithium-ion batteries is projected to expand from its current 400 GWh to 0.5–2 TWh by 2025, 1–6 TWh by 2030, and 2–8 TWh by 2035, primarily due to the use of conventional liquid electrolyte technology. Solid-state batteries currently make up less than 0.5% of the overall lithium-ion battery market but are predicted to make up more than 1% by 2035. Table 4 shows some of the major players in the solid-state battery ecosystem. Liquid electrolyte LIBs are expected to continue to dominate the market, with solid-state batteries needing time to establish themselves as a major technology. Polymer-based solid-state batteries are currently on the market, but their operating temperature of 50–80 °C limits their applications to systems that are in regular use with only small idling periods outside charging periods, such as electric buses. However, further applications are envisioned for use in automated guided vehicles or other industrial applications, with stationary storage also considered promising. By the end of the decade, with further developments in polymer solid-state battery technology, passenger cars and trucks are also considered promising applications [252].

4.4 Commercial Viability of 3D Printed Batteries

The global 3D printing market was valued at $14 billion in the year 2021 and is expected to grow to $77.83 billion by the year 2030, with a compound annual growth rate of 21% from 2021 to 2030. The market for 3D printed batteries is expected to grow at a compound annual growth rate of 19.53% between 2020 and 2030 [248]. The three major 3D-printed battery startups—Sakuú, Blackstone,

Table 4 Key players in the solid-state battery manufacturing industry Reproduced with Permission [37]



Company	Electrolyte	Partnership/investments	
IONIC materials	Li/graphite, polymer SE	HYUNDAI MITSUBISHI RENAULT A123 SYSTEMS	All Solid State
ProLogium	Li/graphite/Si, oxide ceramic SE	Mercedes-Benz NIO ACC AUTOMOTIVE CELLS Co EVA AIR NOVAT	
Solid Power	Li/Si, sulfide ceramic SE	BMW Ford HYUNDAI SAMSUNG A123 SYSTEMS	
ilika	Si, oxide ceramic SE	JAGUAR LAND ROVER HONDA The Power of Dreams McLaren	
QuantumScape	Anode-free, ceramic SE, gel catholyte	FLUENCE A Siemens and AES Company	
WELION	Li, PEO-based SE	mi xiaomi NIO	Semi-Solid State
StoreDot	Nanosized Si, semi-solid SE	Mercedes-Benz bp EVE DAIMLER SAMSUNG VINFAST TDK VOLVO	
SES Beyond Li-ion	Li, semi-solid SE	HYUNDAI KIA HONDA The Power of Dreams gm SK innovation	
Factorial	Semi-solid SE	HYUNDAI KIA Mercedes-Benz STELLANTIS	
SVOLT 蜂巢能源	Sulfide ceramic SE (+ gel)	STELLANTIS BASF EXIDE	
Sion Power	Li, ceramic+polymer SE	BASF SAMSUNG Cummins	

and Photocentric—have been making huge strides towards commercialization.

Several companies provide standard and customized printed batteries to meet a variety of application requirements. Leading producers of printed batteries include BrightVolt (United States), Power Paper Ltd. (United States), Enfucell Oy (Finland), Blue Spark Technologies, Imprint Energy Inc. (United States), and Prelonic Technologies (Austria) [198]. The most widely available printed batteries are non-rechargeable and feature zinc-manganese dioxide with $ZnCl_2$ as an electrolyte. These batteries are printed on plastic substrates and have an open circuit voltage of approximately 1.6 V, as seen in the batteries produced by Blue Spark and Enfucell. These batteries do not contain heavy metals and can be used in the temperature range from – 30 °C to 65 °C. BrightVolt, offers high-energy–density printed batteries based on solid-state electrolytes uses a proprietary polymer matrix electrolyte.

Sakuú developed Kavian printing platform 3D printers for solid-state batteries that are lighter and cheaper than traditional methods. They use a combination of 3D printing techniques to halve the battery manufacturing process steps. The Kavian platform reduces manufacturing costs by 33% and factory size by 44%, aiming for 100% recyclability with a powder-to-powder recycling system. Sakuú's Swift Print Battery Technology binder jetting and metal material jetting with AI-based quality control produces smaller and lighter batteries with twice the energy density of leading Li-ion batteries. This technology reduces waste and toxic materials during manufacturing and recycling, enabling high-volume, rapid battery production with fewer emissions [253].

Blackstone Technology is a European startup that uses a 3D printing method called Thick Layer Technology to manufacture batteries that are more environmentally friendly, cheaper, and have higher energy density. This technology aims to solve problems associated with current battery production methods, and it allows for printing batteries in any shape using a single system. The use of 3D printing in battery production can result in significant savings in capital and operational costs. Additionally, Blackstone's technology increases energy density by 20%, reduces energy consumption by 25%, and enables the addition of additives or the printing of thin current collectors or embedded electronic components. It also facilitates automatic high-speed production [254].

Photocentric has received significant investment from the government of the United Kingdom since 2020 to develop industrial 3D printers that can produce solid-state battery cells for EVs. Photocentric technology, still in the R&D stage, differs from the technology used by Sakuú and Blackstone, as it uses resin 3D printing with a photopolymer. So far, the company has developed printable photopolymer resin–containing polymer electrolyte binders as well as anode and cathode powders. Their patent-pending technology promises to enable low-cost mass production of lightweight batteries for the UK market. As companies worldwide work to improve battery manufacturing, all options are being considered to solve safety issues, support domestic production, secure material sourcing, and increase sustainability [248].

4.5 Circular Economy Through the Recycling of Batteries

The battery recycling industry collects and processes battery waste to facilitate safe disposal and to recover valuable materials. The growth in demand for industry services is due to regulations mandating or encouraging battery recycling as well as the increasing number of used batteries that are entering the waste stream. However, the battery recycling industry has been impacted by the COVID-19 pandemic, leading to a decrease in consumer recycling and a lower demand from downstream industries. Despite this, the industry was expected to realize an annualized increase in revenue of 5.6% to $765.0 million from 2016 to 2021, including a 7.7% increase in 2021 alone. Every year, the billions of batteries, both primary and secondary, that are discarded by consumers and businesses can pose a threat to the environment and public health if they are not properly disposed of. To address this issue, the Mercury-Containing Rechargeable Battery Management Act was passed by the United States Congress in 1996 to facilitate battery recycling. Since then, state and local governments in the United States have gone beyond this legislation by implementing laws that mandate or encourage battery recycling through subsidies, landfill fees, and extended producer responsibility programs. These laws have generated a stable demand for battery recycling services; as collection rates increase, the industry can benefit from economies of scale, resulting in increased profits. The consumption of consumer electronics is expected to increase in the coming years, resulting in a larger number of used batteries entering the waste stream. This will lead to an expansion of state and local regulations supporting the battery recycling industry and driving its growth. In addition, stability in the prices of virgin metals over the next five years will allow the industry to generate significant revenue through the sale of recovered materials during the recycling process (Fig. 9). The industry's revenue is expected to increase at an annualized rate of 1.8% to reach $835.5 million by 2026 [255].

4.5.1 Lithium-Ion Battery Reutilization Strategies:

The growth of electric vehicles will lead to a significant number of retired battery packs. The management of end-of-use (EOU) batteries involves the cooperation of governments, consortia, manufacturers, users, and recycling facilities. Governments play a crucial role in establishing infrastructure and regulations. Collection systems vary across countries, with Japan and Europe enforcing mandatory laws for battery recovery. Mandatory recovery regulations and a nationwide battery tracking system are essential for effective EOU battery utilization. The economic feasibility of battery refurbishment depends on pack architecture, which affects disassembly costs and state-of-health testing. Battery design impacts the complexity of testing and extracting functional components. Optimization tailored to specific applications maximizes economic returns. The chosen recycling route and technologies significantly influence economic feasibility and environmental impact, with cobalt-containing batteries being more profitable to recycle.[257]. The challenges and opportunities that have been recognized of through the review are as follows:

- Manual disassembly of large battery packs is labor-intensive and time-consuming, requiring dedicated machines and automated intelligent disassembly systems.
- Screening functional modules/cells is crucial for efficient battery refurbishment, reducing time and costs.
- Hydrometallurgical extraction can yield acceptable quality regenerated materials, but reducing wastewater, toxic gases, and cost is necessary for industrialization.
- Evaluating environmental impact and economic potential informs decision-making in battery recovery technologies.
- Collaborations among stakeholders are vital for the early-stage battery refurbishing industry, with a focus on business model innovation and knowledge exchange.
- Value recovery thinking should be integrated into the early design stage of batteries to facilitate recycling and improve the product life cycle.

Remanufacturing these lithium-ion battery packs can maximize their remaining value. A partial disassembly line

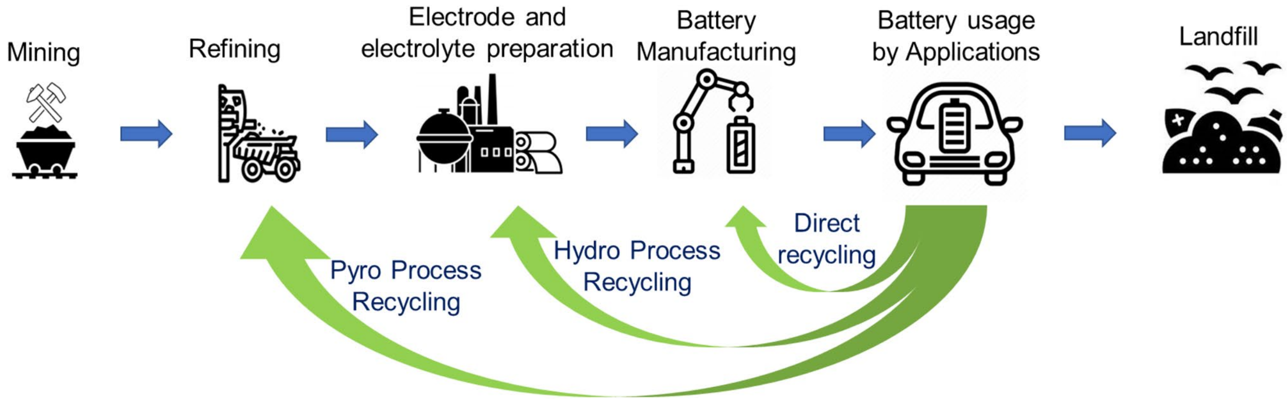


Fig. 9 Circular economy driven by the Li-ion battery recycling Reproduced with permission [256] Copyright 2019, Argonne National Laboratory

model and hybrid genetic-firework algorithm were developed to optimize the disassembly process, considering factors such as the number of stations, smoothness, profit, energy consumption, and safety. The hybrid algorithm outperformed traditional algorithms, and future work will focus on adjusting parameters and addressing uncertainties. Screening retired battery cells based on remaining capacity enables their reuse [258]. An adaptive genetic algorithm and neural network estimated battery capacity with high accuracy, showing potential for rapid screening of decommissioned power batteries [259]. Designing products for recyclability and recovery supports the circular economy, and strategies such as replacing screws with snap-fits and addressing coating obstacles can improve value recovery. This design method is applicable to various products, and future research will explore different curve types and establish capacity estimation models for diverse battery types [260].

5 Discussion

All of the research accomplishments in developing Li-ion batteries over past three decades have seen large-scale acceleration efforts in terms of commercialization. To produce commercially and environmentally viable 3D-printed batteries using solid polymer electrolyte, it is important to discuss how various technologies have emerged from the lab into the market. 3D printing/additive manufacturing technology alone has seen major growth since its inception. The past decade has witnessed new innovations and applications in these directions, which is evident by their increasing market growth and the exponential growth of the battery manufacturing field with the advent of EVs and Internet of Things–based wireless applications. That said, the case for a solid polymer electrolyte can be similar to that for solid-state electrolytes, which have gained a great deal of popularity due to their safety, reliability, and energy efficiency. Solid polymer electrolytes have the major upper hand when it comes shape customization because of their flexibility and other mechanical properties. Hence, 3D printing a solid polymer electrolyte could greatly make a compelling case with disruptive advantages surrounding ease of fabrication compared to other solid electrolytes because of easy processibility minimal need post processing steps. Companies such as Sakuú and Blackstone Technology have successfully commercialized the 3D printing of batteries that use inorganic solid electrolytes have been successful because of their 3D printability complemented with competitive battery properties. Hence 3D printed polymer battery can be competitive if they are able to demonstrate high ionic conductivity, and good solid electrolyte interface, without any compromise in the electrochemical properties like high cycle number, energy density etc. Additionally, with growing concerns over environmental impacts of the materials extraction and battery disposals, it is necessarily to make the electrolytes and electrodes degradable and recyclable, opening sustainable avenues for circular economy and contribute to sustainability of life on planet earth.

6 Summary

Since its introduction to the market in 1991, lithium-ion batteries have undergone significant evolution. The specific energy content of these batteries has nearly tripled, and their lifespan has more than tripled. Despite requiring advanced control systems and cell energy reserves, LIBs can now meet most automotive calendar and cycle life requirements while maintaining a high degree of safety. Moreover, the cost of LIBs has decreased significantly—by almost two orders of magnitude in recent years. However, while LIBs have made great strides in performance and cost, they still are not able to meet the energy targets required by the automotive industry. As a result, various Beyond Lithium Ion (BLI) research initiatives were established within the last decade or so. The three main technologies pursued in the initial BLI efforts were lithium-air (Li-O_2), lithium-sulfur, and lithium-metal. Of these, lithium metal has made the most progress in the past decade, and its potential impact is underscored by recent automotive lithium metal cell targets from the United States Advanced Battery Consortium.

In order to transition towards a society with zero carbon emissions, it is necessary to develop batteries that have better performance, improved ecological footprints, and enhanced safety and sustainability. The European battery community must engage in cross-disciplinary and cross-sectorial research efforts to achieve this goal. With its BATTERY 2030 + initiative, the community has adopted a chemistry-neutral approach to create a generic toolbox for the development, design, and manufacture of batteries that will lead to the development of specific battery chemistries and technologies. They intend to promote the development of diverse battery technologies and build synergies in their understanding by addressing three themes. The first theme relates to the speedy discovery of battery materials by focusing on designing and improving critical battery components. The second theme focuses on integrating smart features (such as self-healing mechanisms) into batteries to improve safety, reliability, and cycle life. Finally, the third theme recommends that basic research efforts must consider the manufacturability and recyclability of batteries. Over the next ten years, the BATTERY 2030 + initiative intends to develop a circular model by incorporating specific R&D actions that are based on the considerations developed in the initial roadmap.

The use of polymer electrolytes in alkaline metal batteries has gained attention, as it can minimize safety hazards. Polymer electrolytes can combine the benefits of inorganic electrolytes and other components to achieve desirable physicochemical properties. However, challenges remain in achieving high ionic conductivity and addressing interfacial issues between the polymer and inorganic components of the battery. Thin-film fabrication techniques, such as atomic/molecular layer deposition, can improve ionic conductance and increase the energy density of these batteries. Strategies such as introducing organic species on the surfaces of inorganic particles and lithophilic layers can help address interfacial issues, but uniform lithium plating is necessary for stabilizing the solid-state battery interface.

Solid-state electrolytes offer a solution to safety issues in using alkali-ion and alkali-metal batteries while providing high energy density and a long calendar lifetime. However, the challenge is to create custom-shaped solid-state batteries using innovative fabrication methods, and additive manufacturing is one of the most promising approaches for this purpose. Polymer and composite polymer electrolytes have been printed by different 3D printing processes that have varying ionic conductivity, and gel electrolytes have been produced that have shown higher ionic conductivity. Solid-state lithium-ion batteries with printed electrolytes have shown a high level of performance. PLA and PLA-PEO solid electrolytes and membranes infused with organics and ionic liquids have been successfully 3D-printed. It is expected that much research will be accomplished on the sustainability of 3D-printed solid polymer electrolyte batteries in the next decades, with enhanced productivity, a more reliable manufacturing process, higher conductivity, and greater energy density.

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Data availability Not available.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

References

1. Masias, A., Marcicki, J., & Paxton, W. A. (2021). Opportunities and challenges of Lithium ion batteries in automotive applications. *ACS Energy Letters, 6*(2), 621–630. https://doi.org/10.1021/acsenergylett.0c02584
2. Li, M., Lu, J., Chen, Z., & Amine, K. (2018). 30 Years of Lithium-ion batteries. *Advanced Materials, 30*(33), 1800561. https://doi.org/10.1002/adma.201800561
3. He, W., Guo, W., Wu, H., Lin, L., Liu, Q., Han, X., & Peng, D. L. (2021). Challenges and recent advances in high capacity Li-rich cathode materials for high energy density lithium-ion batteries. *Advanced Materials, 33*(50), 2005937. https://doi.org/10.1002/adma.202005937
4. Emon, M. O. F., & Choi, J. W. (2017). Flexible piezoresistive sensors embedded in 3D printed tires. *Sensors (Switzerland), 17*(3), 656. https://doi.org/10.3390/s17030656
5. Jo, Y., Park, H. J., Kim, Y. B., Lee, S. S., Lee, S. Y., Kim, S. K., & Jeong, S. (2020). Form-factor free 3D copper circuits by surface-conformal direct printing and laser writing. *Advanced Functional Materials, 30*(45), 2004659. https://doi.org/10.1002/adfm.202004659
6. Ponnada, S., Babu Gorle, D., Chandra Bose, R. S., Sadat Kiai, M., Devi, M., Raju, C. V., Baydogan, N., Nanda, K. K., Marken, F., & Sharma, R. K. (2022). Current insight into 3d printing in solid-state lithium-ion batteries: a perspective. *Batteries & Supercaps, 5*(8), e202200223. https://doi.org/10.1002/batt.202200223
7. Zhang, B., Guo, Y. X., Sun, H., & Wu, Y. (2017). Metallic, 3D-printed, K-band-stepped, double-ridged square horn antennas. *Applied Sciences (Switzerland), 8*(1), 33. https://doi.org/10.3390/app8010033
8. Sverdrup, H. U., Olafsdottir, A. H., & Ragnarsdottir, K. V. (2019). "On the long-term sustainability of copper, zinc and lead supply, using a system dynamics model", resources. *Conservation and Recycling: X*. https://doi.org/10.1016/j.rcrx.2019.100007
9. Cong, L., Zhao, F., & Sutherland, J. W. (2017). Integration of dismantling operations into a value recovery plan for circular economy. *Journal of Cleaner Production, 149*, 378–386. https://doi.org/10.1016/j.jclepro.2017.02.115
10. Royo, P., Ferreira, V. J., Ure, Z., Gledhill, S., López-Sabirón, A. M., & Ferreira, G. (2020). Multiple-criteria decision Analysis and haracterization of phase change materials for waste heat recovery at high temperature for sustainable energy-intensive industry. *Materials and Design*. https://doi.org/10.1016/j.matdes.2019.108215
11. Opatokun, S. A., Lopez-Sabiron, A. M., Ferreira, G., & Strezov, V. (2017). Life cycle analysis of energy production from food waste through anaerobic digestion, pyrolysis and integrated energy system. *Sustainability (Switzerland), 9*(10), 1804. https://doi.org/10.3390/su9101804
12. López-Sabirón, A. M., et al. (2015). Refuse derived fuel (RDF) plasma torch gasification as a feasible route to produce low environmental impact syngas for the cement industry. *Waste Management and Research, 33*(8), 715–722. https://doi.org/10.1177/0734242X15586476
13. Kim, T., Song, W., Son, D. Y., Ono, L. K., & Qi, Y. (2019). Lithium-ion batteries: Outlook on present, future, and hybridized technologies. *Journal of Materials Chemistry A, 7*(7), 2942–2964. https://doi.org/10.1039/C8TA10513H
14. Winter, M., Barnett, B., & Xu, K. (2018). "Before Li Ion Batteries", chemical reviews. *American Chemical Society*. https://doi.org/10.1021/acs.chemrev.8b00422
15. Masias, A. (2018). Lithium-Ion Battery Design for Transportation. *Behaviour of Lithium-Ion Batteries in Electric Vehicles: Battery Health, Performance, Safety, and Cost*, 1–33. Doi: https://doi.org/10.1007/978-3-319-69950-9.
16. European Council for Automotive R&D, "Battery requirements for future automotive applications EG BEV&FCEV," 2019. [Online]. Available: https://www.dke.de/resource/blob/933404/fa7a24099c84ef613d8e7afd2c860a39/haracteri-li-ionen

17. 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://uscar.org/usabc/.
18. United States Advanced Battery Consortium (USABC). Goals for advanced high performance batteries for electric vehicle applications. April 2020. Retrieved from: https://uscar.org/usabc/
19. Tan, M. Y., Safanama, D., Goh, S. S., Lim, J. Y., Lee, C. H., Yeo, J. C. C., & Fam, D. W. H. (2022). Concepts and emerging trends for structural battery electrolytes. *ChemistryAn Asian Journal, 17*(21), e202200784.
20. Myung, S. T., Maglia, F., Park, K. J., Yoon, C. S., Lamp, P., Kim, S. J., & Sun, Y. K. (2017). Nickel-rich layered cathode materials for automotive lithium-ion batteries: Achievements and perspectives. *ACS Energy Letters, 2*(1), 196–223. https://doi.org/10.1021/acsenergylett.6b00594
21. Thackeray, M. M., Kang, S. H., Johnson, C. S., Vaughey, J. T., Benedek, R., & Hackney, S. A. (2007). Li2MnO3-stabilized LiMO2 (M = Mn, Ni, Co) electrodes for lithium-ion batteries. *Journal of Materials Chemistry, 17*(30), 3112–3125. https://doi.org/10.1039/b702425h
22. Chen, S., Dai, F., & Cai, M. (2020). Opportunities and challenges of high-energy Lithium metal batteries for electric vehicle applications. *ACS Energy Letters, 5*(10), 3140–3151. https://doi.org/10.1021/acsenergylett.0c01545
23. Choi, J. W., & Aurbach, D. (2016). Promise and reality of post-lithium-ion batteries with high energy densities. *Nature Reviews Materials*. https://doi.org/10.1038/natrevmats.2016.13
24. Shen, X., Tian, Z., Fan, R., Shao, L., Zhang, D., Cao, G., & Bai, Y. (2018). Research progress on silicon/carbon composite anode materials for lithium-ion battery. *Journal of Energy Chemistry, 27*(4), 1067–1090. https://doi.org/10.1016/j.jechem.2017.12.012
25. Salah, M., Murphy, P., Hall, C., Francis, C., Kerr, R., & Fabretto, M. (2019). Pure silicon thin-film anodes for lithium-ion batteries: a review. *Journal of Power Sources, 414*, 48–67. https://doi.org/10.1016/j.jpowsour.2018.12.068
26. Wold, J., Marcicki, J., & Masias, A. (2017). Derived Quantities uncertainty propagation in high precision battery testing. *Journal of the Electrochemical Society, 164*(9), A2131–A2137. https://doi.org/10.1149/2.1461709jes
27. Belt, J., Utgikar, V., & Bloom, I. (2011). Calendar and PHEV cycle life aging of high-energy, lithium-ion cells containing blended spinel and layered-oxide cathodes. *Journal of Power Sources, 196*(23), 10213–10221. https://doi.org/10.1016/j.jpowsour.2011.08.067
28. Smith, A. J., Burns, J. C., Xiong, D., & Dahn, J. R. (2011). Interpreting high precision coulometry results on Li-ion Cells. *Journal of the Electrochemical Society, 158*(10), A1136–A1142. https://doi.org/10.1149/1.3625232
29. Burns, J. C., Kassam, A., Sinha, N. N., Downie, L. E., Solnickova, L., Way, B. M., & Dahn, J. R. (2013). Predicting and extending the lifetime of Li-ion batteries. *Journal of The Electrochemical Society, 160*(9), A1451. https://doi.org/10.1149/2.060309jes
30. Ma, X., Harlow, J. E., Li, J., Ma, L., Hall, D. S., Buteau, S., & Dahn, J. R. (2019). Hindering rollover failure of Li [Ni0. 5Mn0. 3Co0.2] O2/graphite pouch cells during long-term cycling. *Journal of The Electrochemical Society*. https://doi.org/10.1149/2.0801904jes
31. Burns, J. C., Stevens, D. A., & Dahn, J. R. (2015). In-situ detection of lithium plating using high precision coulometry. *Journal of the Electrochemical Society, 162*(6), A959–A964. https://doi.org/10.1149/2.0621506jes
32. Bach, T. C., Schuster, S. F., Fleder, E., Müller, J., Brand, M. J., Lorrmann, H., & Sextl, G. (2016). Nonlinear aging of cylindrical lithium-ion cells linked to heterogeneous compression. *Journal of Energy Storage, 5*, 212–223. https://doi.org/10.1016/j.est.2016.01.003
33. Sauerteig, D., Hanselmann, N., Arzberger, A., Reinshagen, H., Ivanov, S., & Bund, A. (2018). Electrochemical-mechanical coupled modeling and parameterization of swelling and ionic transport in lithium-ion batteries. *Journal of Power Sources, 378*, 235–247. https://doi.org/10.1016/j.jpowsour.2017.12.044
34. Spingler, F. B., Wittmann, W., Sturm, J., Rieger, B., & Jossen, A. (2018). Optimum fast charging of lithium-ion pouch cells based on local volume expansion criteria. *Journal of Power Sources, 393*, 152–160. https://doi.org/10.1016/j.jpowsour.2018.04.095
35. Severson, K. A., Attia, P. M., Jin, N., Perkins, N., Jiang, B., Yang, Z., Chen, M. H., Aykol, M., Herring, P. K., Fraggedakis, D., Bazant, M. Z., Harris, S. J., Chueh, W. C., & Braatz, R. D. (2019). Data-driven prediction of battery cycle life before capacity degradation. *Nature Energy, 4*(5), 383–391. https://doi.org/10.1038/s41560-019-0356-8
36. Attia, P. M., Grover, A., Jin, N., Severson, K. A., Markov, T. M., Liao, Y. H., & Chueh, W. C. (2020). Closed-loop optimization of fast-charging protocols for batteries with machine learning. *Nature*. https://doi.org/10.1038/s41586-020-1994-5
37. Yen. T. Teh, Aubrey Demaray, Kent Griffith, Charlie Parker, Josh Stilling, Sriram Bharath, Andrew Weng, Eric Zheng, Jeffrey Bell, Mouda Abusukheila, Nick Albanese, Nicholas Yu, Katherine He, Rohit Kumar, & Jhalak Sharma. (2023). *The battery Report 2022*. https://www.volta.foundation/annual-battery-report
38. Duffner, F., Wentker, M., Greenwood, M., & Leker, J. (2020). Battery cost modeling: A review and directions for future research. *Renewable and Sustainable Energy Reviews*. https://doi.org/10.1016/j.rser.2020.109872
39. J. Frith, "Battery Price Declines Slow Down in Latest Pricing Survey," *Bloomberg Finance*, Nov. 30, 2021. https://www.bloomberg.com/news/articles/2021-11-30/battery-price-declines-slow-down-in-latest-pricing-survey#xj4y7vzkg
40. Ahmed, S., Nelson, P. A., Gallagher, K. G., Susarla, N., & Dees, D. W. (2017). Cost and energy demand of producing nickel manganese cobalt cathode material for lithium ion batteries. *Journal of Power Sources, 342*, 733–740. https://doi.org/10.1016/j.jpowsour.2016.12.069
41. Schmuch, R., Wagner, R., Hörpel, G., Placke, T., & Winter, M. (2018). Performance and cost of materials for lithium-based rechargeable automotive batteries. *Nature Energy, 3*(4), 267–278. https://doi.org/10.1038/s41560-018-0107-2
42. Turcheniuk, K., Bondarev, D., Amatucci, G. G., & Yushin, G. (2021). Battery materials for low-cost electric transportation. *Materials Today, 42*, 57–72. https://doi.org/10.1016/j.mattod.2020.09.027
43. Chen, M., Ma, X., Chen, B., Arsenault, R., Karlson, P., Simon, N., & Wang, Y. (2019). Recycling end-of-life electric vehicle lithium-ion batteries. *Joule, 3*(11), 2622–2646. https://doi.org/10.1016/j.joule.2019.09.014
44. Borlaug, B., Salisbury, S., Gerdes, M., & Muratori, M. (2020). Levelized cost of charging electric vehicles in the United States. *Joule, 4*(7), 1470–1485. https://doi.org/10.1016/j.joule.2020.05.013
45. Kim, H. C., Wallington, T. J., Arsenault, R., Bae, C., Ahn, S., & Lee, J. (2016). Cradle-to-gate emissions from a commercial electric vehicle Li-Ion battery: a comparative analysis. *Environmental Science and Technology, 50*(14), 7715–7722. https://doi.org/10.1021/acs.est.6b00830
46. Tong, F., & Azevedo, I. M. L. (2020). "What are the best combinations of fuel-vehicle technologies to mitigate climate change

and air pollution effects across the United States. *Environmental Research Letters*. https://doi.org/10.1088/1748-9326/ab8a85

47. Wu, D., Guo, F., Field, F. R., III., De Kleine, R. D., Kim, H. C., Wallington, T. J., & Kirchain, R. E. (2019). Regional heterogeneity in the emissions benefits of electrified and lightweighted light-duty vehicles. *Environmental science & technology, 53*(18), 10560–10570. https://doi.org/10.1021/acs.est.9b00648
48. Liu, X., Stoliarov, S. I., Denlinger, M., Masias, A., & Snyder, K. (2015). Comprehensive calorimetry of the thermally-induced failure of a lithium ion battery. *Journal of Power Sources, 280*, 516–525. https://doi.org/10.1016/j.jpowsour.2015.01.125
49. Liu, X., Wu, Z., Stoliarov, S. I., Denlinger, M., Masias, A., & Snyder, K. (2018). A Thermo-kinetic model of thermally-induced failure of a lithium ion battery: development, validation and application. *Journal of the Electrochemical Society, 165*(11), A2909–A2918. https://doi.org/10.1149/2.0111813jes
50. Sahraei, E., Meier, J., & Wierzbicki, T. (2014). Characterizing and modeling mechanical properties and onset of short circuit for three types of lithium-ion pouch cells. *Journal of Power Sources, 247*, 503–516. https://doi.org/10.1016/j.jpowsour.2013.08.056
51. Sahraei, E., Hill, R., & Wierzbicki, T. (2012). Calibration and finite element simulation of pouch lithium-ion batteries for mechanical integrity. *Journal of Power Sources, 201*, 307–321. https://doi.org/10.1016/j.jpowsour.2011.10.094
52. B. Xia, Z. Chen, C. Mi, and B. Robert, “External short circuit fault diagnosis for lithium-ion batteries,” in *2014 IEEE Transportation Electrification Conference and Expo: Components, Systems, and Power Electronics – From Technology to Business and Public Policy, ITEC 2014*, Jul. 2014 https://doi.org/10.1109/itec.2014.6861806.
53. B. Xia, C. Mi, Z. Chen, and B. Robert, “Multiple cell lithium-ion battery system electric fault online diagnostics,” in *2015 IEEE Transportation Electrification Conference and Expo, ITEC 2015*, Jul. 2015. https://doi.org/10.1109/ITEC.2015.7165777.
54. A. Masias, “Ford Safety Performance of Rechargeable Energy Storage Systems (RESS),” Report No. DOT HS 812 756. Final report. Washington, D.C.: National Highway Traffic Safety Administration. July 2019. [Online]. Available: www.ntis.gov.
55. Deng, J., Bae, C., Marcicki, J., Masias, A., & Miller, T. (2018). Safety modelling and testing of lithium-ion batteries in electrified vehicles. *Nature Energy, 3*(4), 261–266. https://doi.org/10.1038/s41560-018-0122-3
56. United Nations, “Uniform provisions concerning the approval of vehicles with regard to specific requirements for the electric power train, Addendum 99: Regulation No. 100, rev. 2, New York: United Nations, Aug 12, 2013.
57. H. Jung, B. Moon, S. Lee, and J. Bae, “A STUDY ON THERMAL ENERGY AT FIRE RESISTANCE TEST FOR REESS.” 25th International Technical Conference on the Enhanced Safety of Vehicles (ESV), Detroit, Michigan. 2017.
58. United Nations Economic commission for Europe, “Manual of Tests and Criteria” Seventh revised edition. December 2019. https://doi.org/10.18356/390e2d1a-en
59. United Nations Economic Commission for Europe, “Global Technical Regulation on the Electric Vehicle Safety (EVS)” ECE/TRANS/180/Add.20. In: 1998 Agreement on Global Technical Regulations Addendum 20, 1998. https://unece.org/transport/standards/transport/vehicle-regulations-wp29/global-technical-regulations-gtrs
60. Wu, X., Song, K., Zhang, X., Hu, N., Li, L., Li, W., & Zhang, H. (2019). Safety issues in lithium ion batteries: Materials and cell design. *Frontiers in Energy Research, 7*, 65. https://doi.org/10.3389/fenrg.2019.00065
61. Liu, X., Ren, D., Hsu, H., Feng, X., Xu, G. L., Zhuang, M., Gao, H., Lu, L., Han, X., Chu, Z., Li, J., He, X., Amine, K., & Ouyang, M. (2018). Thermal runaway of lithium-ion batteries without internal short circuit. *Joule, 2*(10), 2047–2064. https://doi.org/10.1016/j.joule.2018.06.015
62. Liu, K., Liu, Y., Lin, D., Pei, A., & Cui, Y. (2018). Materials for lithium-ion battery safety. *Science Advances*. https://doi.org/10.1126/sciadv.aas9820
63. Ahmed, S., Bloom, I., Jansen, A. N., Tanim, T., Dufek, E. J., Pesaran, A., & Zhang, J. (2017). Enabling fast charging–A battery technology gap assessment. *Journal of Power Sources*. https://doi.org/10.1016/j.jpowsour.2017.06.055
64. Bommier, C., Chang, W., Lu, Y., Yeung, J., Davies, G., Mohr, R., & Steingart, D. (2020). In operando acoustic detection of lithium metal plating in commercial LiCoO2/graphite pouch cells. *Cell Reports Physical Science*. https://doi.org/10.1016/j.xcrp.2020.100035
65. Amici, J., Asinari, P., Ayerbe, E., Barboux, P., Bayle-Guillemaud, P., Behm, R. J., & Edström, K. (2022). A roadmap for transforming research to invent the batteries of the future designed within the european large scale research initiative battery 2030+. *Advanced Energy Materials, 12*(17), 2102785. https://doi.org/10.1002/aenm.202102785
66. Clean Energy Materials Innovation Challenge Expert Workshop. (2020). Materials Acceleration Platform Accelerating Advanced Energy Materials Discovery by Integrating High-Throughput Methods with Artificial Intelligence. UC Office of the President: UC-Mexico Initiative. Retrieved from https://escholarship.org/uc/item/3926s13w
67. Vegge, T., Tarascon, J. M., & Edström, K. (2021). Toward better and smarter batteries by combining ai with multisensory and self-healing approaches. *Advanced Energy Materials, 11*(23), 2100362. https://doi.org/10.1002/aenm.202100362
68. Seifrid, M., Hickman, R. J., Aguilar-Granda, A., Lavigne, C., Vestfrid, J., Wu, T. C., & Aspuru-Guzik, A. (2022). Routescore: punching the ticket to more efficient materials development. *ACS Central Science, 8*(1), 122–131. https://doi.org/10.1021/acscentsci.1c01002
69. Atkins, D., Ayerbe, E., Benayad, A., Capone, F. G., Capria, E., Castelli, I. E., Cekic-Laskovic, I., Ciria, R., Dudy, L., Edstrom, K., Johnson, M. R., Li, H., Lastra, J. M. G., De Souza, M. L., Meunier, V., Morcrette, M., Reichert, H., Simon, P., Rueff, J. P., … Grimaud, A. (2022). Understanding battery interfaces by combined characterization and simulation approaches: challenges and perspectives. *Advanced Energy Materials, 12*(17), 2102687. https://doi.org/10.1002/aenm.202102687
70. Philippot, M., Alvarez, G., Ayerbe, E., van Mierlo, J., & Messagie, M. (2019). Eco-efficiency of a lithium-ion battery for electric vehicles: Influence of manufacturing country and commodity prices on GHG emissions and costs. *Batteries, 5*(1), 23. https://doi.org/10.3390/batteries5010023
71. Narayan, R., Laberty-Robert, C., Pelta, J., Tarascon, J. M., & Dominko, R. (2022). Self-healing: an emerging technology for next-generation smart batteries. *Advanced Energy Materials, 12*(17), 2102652. https://doi.org/10.1002/aenm.202102652
72. Senyshyn, A., Mühlbauer, M. J., Nikolowski, K., Pirling, T., & Ehrenberg, H. (2012). ‘In-operando’ neutron scattering studies on Li-ion batteries. *Journal of Power Sources, 203*, 126–129. https://doi.org/10.1016/j.jpowsour.2011.12.007
73. Bergman, S. D., & Wudl, F. (2008). Mendable polymers. *Journal of Materials Chemistry, 18*(1), 41–62. https://doi.org/10.1039/b713953p
74. Wang, H., Wang, P., Feng, Y., Liu, J., Wang, J., Hu, M., & Huang, Y. (2019). Recent advances on self-healing materials and batteries. *ChemElectroChem, 6*(6), 1605–1622. https://doi.org/10.1002/celc.201801612
75. Kwon, T. W., Choi, J. W., & Coskun, A. (2019). Prospect for supramolecular chemistry in high-energy-density rechargeable

batteries. *Joule, 3*(3), 662–682. https://doi.org/10.1016/j.joule.2019.01.006

76. Sharpe, R., van Lopik, K., Neal, A., Goodall, P., Conway, P. P., & West, A. A. (2019). An industrial evaluation of an Industry 4.0 reference architecture demonstrating the need for the inclusion of security and human components. *Computers in Industry, 108*, 37–44. https://doi.org/10.1016/j.compind.2019.02.007
77. Negri, E., Fumagalli, L., & Macchi, M. (2017). A review of the roles of digital Twin in CPS-based production systems. *Procedia Manufacturing, 11*, 939–948. https://doi.org/10.1016/j.promfg.2017.07.198
78. Liu, K., Wei, Z., Yang, Z., & Li, K. (2021). Mass load prediction for lithium-ion battery electrode clean production: a machine learning approach. *Journal of Cleaner Production*. https://doi.org/10.1016/j.jclepro.2020.125159
79. The European Parliament And The Council Of The Euro-Pean Union. (2006). DIRECTIVE 2006/66/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCILof 6 September 2006on batteries and accumulators and waste batteries and accumulators and repealing Directive91/157/EEC. http://data.europa.eu/eli/dir/2006/66/oj
80. Yao, W., Zhang, Z., Gao, J., Li, J., Xu, J., Wang, Z., & Yang, Y. (2009). Vinyl ethylene sulfite as a new additive in propylene carbonate-based electrolyte for lithium ion batteries. *Energy & Environmental Science, 2*(10), 1102–1108. https://doi.org/10.1039/b905162g
81. Wang, R., Li, X., Wang, Z., & Zhang, H. (2017). Electrochemical analysis graphite/electrolyte interface in lithium-ion batteries: p-Toluenesulfonyl isocyanate as electrolyte additive. *Nano Energy, 34*, 131–140. https://doi.org/10.1016/j.nanoen.2017.02.037
82. Ushirogata, K., Sodeyama, K., Okuno, Y., & Tateyama, Y. (2013). Additive effect on reductive decomposition and binding of carbonate-based solvent toward solid electrolyte interphase formation in lithium-ion battery. *Journal of the American Chemical Society, 135*(32), 11967–11974. https://doi.org/10.1021/ja405079s
83. Leggesse, E. G., & Jiang, J. C. (2012). Theoretical study of the reductive decomposition of ethylene sulfite: a film-forming electrolyte additive in lithium ion batteries. *Journal of Physical Chemistry A, 116*(45), 11025–11033. https://doi.org/10.1021/jp3081996
84. Goodenough, J. B., & Kim, Y. (2010). Challenges for rechargeable Li batteries. *Chemistry of Materials, 22*(3), 587–603. https://doi.org/10.1021/cm901452z
85. Armand, M. B. (1986). Polymer electrolytes. *Ann Rev Mater*. https://doi.org/10.1146/annurev.ms.16.080186.001333
86. Scrosati, B. (1995). Challenge of portable power. *Nature, 373*, 557–558. https://doi.org/10.1038/373557a0
87. Behera, Ajit, and Ajit Behera. "Energy Harvesting and Storing Materials." Advanced Materials: An Introduction to Modern Materials Science (2022): 507–555.
88. Boz, B., Ford, H. O., Salvadori, A., & Schaefer, J. L. (2021). Porous polymer gel electrolytes influence lithium transference number and cycling in lithium-ion batteries. *Electronic Materials, 2*(2), 154–173. https://doi.org/10.3390/electronicmat2020013
89. Fenton, D. E. (1973). Complexes of alkali metal ions with poly (Haracte oxide). *Polymer, 14*, 589.
90. Armand, M. B., Chabagno, J. M., & Duclot, M. J. (1979). Fast ion transport in solids. Electrodes and Electrolytes, 131.
91. Bouchet, R., Maria, S., Meziane, R., Aboulaich, A., Lienafa, L., Bonnet, J. P., & Armand, M. (2013). Single-ion BAB triblock copolymers as highly efficient electrolytes for lithium-metal batteries. *Nature materials, 12*(5), 452–457. https://doi.org/10.1038/nmat3602
92. Imanishi, N., & Yamamoto, O. (2013). Polymer electrolytes for Lithium-air batteries. *Lithium Batteries Advanced Technologies and Applications*. https://doi.org/10.1002/9781118615515.ch10
93. Meyer, W. H. (1998). Polymer electrolytes for lithium-ion batteries. *Advanced Materials, 10*(6), 439–448. https://doi.org/10.1002/(SICI)1521-4095(199804)10:6%3c439::AID-ADMA439%3e3.0.CO;2-I
94. Yue, L., Ma, J., Zhang, J., Zhao, J., Dong, S., Liu, Z., & Chen, L. (2016). All solid-state polymer electrolytes for high-performance lithium ion batteries. *Energy Storage Materials, 5*, 139–164. https://doi.org/10.1016/j.ensm.2016.07.003
95. Ahmad, S. (2009). Polymer electrolytes: Characteristics and peculiarities. *Ionics (Kiel), 15*(3), 309–321. https://doi.org/10.1007/s11581-008-0309-x
96. Hallinan, D. T., & Balsara, N. P. (2013). Polymer electrolytes. *Annual Review of Materials Research*. https://doi.org/10.1146/annurev-matsci-071312-121705
97. D. E. Fenton, M. Parker, and P. v Wright, "A-l) 1966, 4, 1563," 1964.
98. Scrosati, B., & Vincent, C. A. (2000). Polymer electrolytes: the key to lithium polymer batteries. *Mrs Bulletin, 25*(3), 28–30.
99. G. G. Cameron, "Polymer electrolyte reviews-l edited by J. R. MacCallum and C. A. Vincent, Elsevier Applied Science Publishers, London, 1987. Pp. x + 351, price £48.00. ISBN 1–85166–07 1–2," *British Polymer Journal*, 20 (3) 299–300, 1988
100. Ib, I., Olsen, R. K., & Skou, E. (1995). Transference Number Measurements on a Hybrid Polyer Electrolyte. *Electrochimica Acta, 40*(11), 1701–1706.
101. Agrawal, R. C., & Pandey, G. P. (2008). Solid polymer electrolytes: materials designing and all-solid-state battery applications: an overview. *Journal of Physics. D. Applied Physics*. https://doi.org/10.1088/0022-3727/41/22/223001
102. Echeverri, M., Kim, N., & Kyu, T. (2012). Ionic conductivity in relation to ternary phase diagram of poly(ethylene oxide), succinonitrile, and lithium bis(trifluoromethane)sulfonyl imide blends. *Macromolecules, 45*(15), 6068–6077. https://doi.org/10.1021/ma3008509
103. Scorsati, B. (2000). Polymer electrolytes: the key to Lithiumpolymer batteries. *MRS Bulletin*. https://doi.org/10.1557/mrs2000.15
104. Petersen, G., Jacobson, P., & Torell, L. M. (1992). A raman study interactions of Ion-polymer and Ion-Ion interactions in low molecular weight polyether–$LiCF_3SO_3$ complexes. *Electrochimica Acta, 37*(9), 1495–1497.
105. Bruce, P. G., & Vincent, C. A. (1989). Effect of Ion association on transport in polymer electrolytes. *Faraday Discussions of the Chemical Society, 88*, 43–54. https://doi.org/10.1039/DC9898800043
106. Lightfoot, P., Mehta, M. A., & Bruce, P. G. (1993). Crystal structure of the polymer electrolyte poly (ethylene oxide) 3: LiCF3SO3. *Science, 262*(5135), 883–885.
107. Benrabah, D., Baril, D., Sanchez, J.-Y., Armand, M., & Gard, G. G. (1993). Comparative electrochemical study of new poly(oxyethy1ene)-Li salt complexes. *Faraday Discussions of the Chemical Society, 89*, 355–359. https://doi.org/10.1039/FT9938900355
108. Wilson, D. J., Nicholas, C. V., Mobbs, R. H., Booth, C., & Giles, J. R. M. (1990). "Synthesis of block copolymers based on oxyethylene chains and their use as polymer electrolytes. *British Polymer Journal*. https://doi.org/10.1002/pi.4980220206
109. Chintapalli, S., & Frech, R. (1996). Effect of plasticizers on Ionic association and conductivity in the (PEO) 9 $LiCF_3SO_3$ system. *Macromolecules, 29*(10), 3499–3506. https://doi.org/10.1021/ma9515644

110. Kim, Y.-T., & Smotkin, E. S. (2002). The effect of plasticizers on transport and electrochemical properties of PEO-based electrolytes for lithium rechargeable batteries. *Solid State Ionics, 149*(1–2), 29–37.
111. Das, S., & Ghosh, A. (2015). Ionic conductivity and dielectric permittivity of PEO-$LiClO_4$ solid polymer electrolyte plasticized with propylene carbonate. *AIP Advances*. https://doi.org/10.1063/1.4913320
112. Frech, R., & Chintapalli, S. (1996). Effect of propylene carbonate as a plasticizer in high molecular weight PEO $LiCF_3SO_3$ electrolytes. *Solid State Ionics, 85*(1–4), 61–66. https://doi.org/10.1016/0167-2738(96)00041-0
113. Kelly, I. E., Owen, J. R., & Steele, B. C. H. (1985). Poly (ethylene oxide) electrolytes for operation at near room temperature. *Journal of Power Sources, 14*(1–3), 13–21. https://doi.org/10.1016/0378-7753(85)88004-6
114. Wang, C., Liu, Q., Cao, Q., & Meng, Q. (1992). Investigation on the structure and the conductivity of plasticized polymer electrolytes. *Solid State Ionics*. https://doi.org/10.1016/0167-2738(92)90298-4
115. Porcarelli, L., Gerbaldi, C., Bella, F., & Nair, J. R. (2016). Super Soft All-ethylene oxide polymer electrolyte for safe all-solid lithium batteries. *Science and Reports, 6*, 19892. https://doi.org/10.1038/srep19892
116. Michael, M. S., Jacob, M. M. E., Prabaharan, S. R. S., & Radhakrishna, S. (1997). Enhanced lithium-ion transport in PEO-based solid polymer electrolytes employing a novel class of plasticizers. *Solid State Ionics, 98*(3–4), 167–174. https://doi.org/10.1016/S0167-2738(97)00117-3
117. Nicotera, I., Ranieri, G. A., Terenzi, M., Chadwick, A. V., & Webster, M. I. (2002). A study of stability of plasticized PEO electrolytes. *Solid State Ionics, 146*(1–2), 143–150. https://doi.org/10.1016/S0167-2738(01)01003-7
118. Angell, C. A., Liu, C., & Sanchez, E. (1993). Rubbery solid electrolytes with dominant cationic transport and high ambient conductivity. *Nature, 362*(6416), 137–139.
119. Ferry, A., Edman, L., Forsyth, M., MacFarlane, D. R., & Sun, J. (1999). Connectivity, ionic interactions, and migration in a fast-ion-conducting polymer-in-salt electrolyte based on poly(acrylonitrile) and $LiCF_3SO_3$. *Journal of Applied Physics, 86*(4), 2346–2348. https://doi.org/10.1063/1.371053
120. Bushkova, O. V., Zhukovsky, V. M., Lirova, B. I., & Kruglyashov, A. L. (1999). Fast ionic transport in solid polymer electrolytes based on acrylonitrile copolymers. *Solid State Ionics, 119*(1–4), 217–222. https://doi.org/10.1016/S0167-2738(98)00506-2
121. Mishra, R., Baskaran, N., Ramakrishnan, P. A., & Rao, K. J. (1998). Lithium-ion conduction in extreme polymer in salt regime. *Solid State Ionics, 112*(3–3), 261–273. https://doi.org/10.1016/S0167-2738(98)00209-4
122. Croce, F., Appetecchi, G. B., Persi, L., & Scrosati, B. (1998). Nanocomposite polymer electrolytes for lithium batteries. *Nature, 394*(6692), 456–458.
123. Appetecchi, G. B., Scaccia, S., & Passerini, S. (2000). Investigation on the stability of the lithium-polymer electrolyte interface. *Journal of the Electrochemical Society, 147*(12), 4448–4452. https://doi.org/10.1149/1.1394084
124. Bronstein, L. M., Karlinsey, R. L., Ritter, K., Joo, C. G., Stein, B., & Zwanziger, J. W. (2004). Design of organic-inorganic solid polymer electrolytes: synthesis, structure, and properties. *Journal of Materials Chemistry, 14*(12), 1812–1820. https://doi.org/10.1039/b401826e
125. Itoh, T., Miyamura, Y., Ichikawa, Y., Uno, T., Kubo, M., & Yamamoto, O. (2003). Composite polymer electrolytes of poly(ethylene oxide)/BaTiO3/Li salt with hyperbranched polymer. *Journal of Power Sources, 119–121*, 403–408. https://doi.org/10.1016/S0378-7753(03)00261-1
126. Skaarup, S., West, K., & Zachau-Christiansen, B. (1988). Mixed phase solid electrolytes. *Solid State Ionics*. https://doi.org/10.1016/0167-2738(88)90314-1
127. Wieczorek, W. (1992). Temperature dependence of conductivity of mixed-phase composite polymer solid electrolytes. *Materials Science and Engineering B, 15*(2), 108–114. https://doi.org/10.1016/0921-5107(92)90041-7
128. Capuano, F., Croce, F., & Scrosati, B. (1991). Composite polymer electrolytes. *Journal of the Electrochemical Society, 138*(7), 1918.
129. Forsyth, M., MacFarlane, D. R., Best, A., Adebahr, J., Jacobsson, P., & Hill, A. J. (2002). The effect of nano-particle TiO_2 fillers on structure and transport in polymer electrolytes. *Solid State Ionics, 147*(3–4), 203–211. https://doi.org/10.1016/S0167-2738(02)00017-6
130. Liu, Y., Lee, J. Y., & Hong, L. (2002). Functionalized SiO_2 in poly(ethylene oxide)-based polymer electrolytes. *Journal of Power Sources, 109*(2), 507–514. https://doi.org/10.1016/S0378-7753(02)00167-2
131. Croce, F., Settimi, L., & Scrosati, B. (2006). Superacid ZrO_2-added, composite polymer electrolytes with improved transport properties. *Electrochemistry Communications, 8*(2), 364–368. https://doi.org/10.1016/j.elecom.2005.12.002
132. Croce, F., Sacchetti, S., & Scrosati, B. (2006). Advanced, lithium batteries based on high-performance composite polymer electrolytes. *Journal of Power Sources, 162*(1), 685–689. https://doi.org/10.1016/j.jpowsour.2006.07.038
133. Xi, J., Qiu, X., Zheng, S., & Tang, X. (2005). Nanocomposite polymer electrolyte comprising PEO/LiClO4 and solid super acid: Effect of sulphated-zirconia on the crystallization kinetics of PEO. *Polymer (Guildf), 46*(15), 5702–5706. https://doi.org/10.1016/j.polymer.2005.05.051
134. S. Abbrent, S. Greenbaum, E. Peled, and D. Golodnitsky, "Polymer Electrolytes." In: *Handbook of Solid State Batteries*, 2nd ed. N.J. Dudney, W.C. West, and J. Nanda (eds.). Singapore: World Scientific. 2015 https://doi.org/10.1142/9789814651905_0016.
135. Varzi, A., Raccichini, R., Passerini, S., & Scrosati, B. (2016). Challenges and prospects of the role of solid electrolytes in the revitalization of lithium metal batteries. *J Mater Chem A Mater, 4*(44), 17251–17259. https://doi.org/10.1039/c6ta07384k
136. Bocharova, V., & Sokolov, A. P. (2020). Perspectives for polymer electrolytes: a view from fundamentals of ionic conductivity. *Macromolecules, American Chemical Society*. https://doi.org/10.1021/acs.macromol.9b02742
137. Strauss, E., Menkin, S., & Golodnitsky, D. (2017). On the way to high-conductivity single lithium-ion conductors. *Journal of Solid State Electrochemistry, 21*(7), 1879–1905. https://doi.org/10.1007/s10008-017-3638-8
138. Kerman, K., Luntz, A., Viswanathan, V., Chiang, Y.-M., & Chen, Z. (2017). Review—practical challenges hindering the development of solid state Li Ion batteries. *Journal of the Electrochemical Society, 164*(7), A1731–A1744. https://doi.org/10.1149/2.1571707jes
139. Golodnitsky, D., Ardel, G., Strauss, E., Peled, E., Lareah, Y., & Rosenberg, Y. (1997). Conduction mechanisms in concentrated Lu-polyethylene Oxide-A1203-based solid electrolytes. *Journal of the Electrochemical Society, 144*(10), 3484–3491. https://doi.org/10.1149/1.1838037
140. Wang, Y., Fan, F., Agapov, A. L., Saito, T., Yang, J., Yu, X., & Sokolov, A. P. (2014). Examination of the fundamental relation between ionic transport and segmental relaxation in polymer electrolytes. *Polymer, 55*(16), 4067–4076. https://doi.org/10.1016/j.polymer.2014.06.085
141. Golodnitsky, D., Strauss, E., Peled, E., & Greenbaum, S. (2015). Review—on order and disorder in polymer electrolytes. *Journal*

of the Electrochemical Society, 162(14), A2551–A2566. https://doi.org/10.1149/2.0161514jes

142. Blanga, R., Burstein, L., Berman, M., Greenbaum, S. G., & Golodnitsky, D. (2015). Solid polymer-in-ceramic electrolyte formed by electrophoretic deposition. *Journal of the Electrochemical Society, 162*(11), D3084–D3089. https://doi.org/10.1149/2.0221511jes
143. Menkin, S., Lifshitz, M., Haimovich, A., Goor, H., Blanga, R., Greenbaum, S. G., & Golodnitsky, D. (2019). Evaluation of ion-transport in composite polymer-in-ceramic electrolytes. case study of active and inert ceramics. *Electrochimica Acta, 304*, 447–455. https://doi.org/10.1016/j.electacta.2019.03.006
144. Kumar, B., & Scanlon, L. G. (1994). Polymer-ceramic composite electrolytes. *Journal of Power Sources, 52*(2), 261–268. https://doi.org/10.1016/0378-7753(94)02147-3
145. Abraham, K. M. (1993). Highly conductive polymer electrolytes. *Applications of electroactive polymer*. https://doi.org/10.1007/978-94-011-1568-1_3
146. Dai, Y., Wang, Y., Greenbaum, S. G., Bajue, S. A., Golodnitsky, D., Ardel, G., & Peled, E. (1998). Electrical, thermal and NMR investigation of composite solid electrolytes based on PEO, LiI and high surface area inorganic oxides. *Electrochimica acta, 43*(10–11), 1557–1561. https://doi.org/10.1016/S0013-4686(97)10053-6
147. Horowitz, Y., Lifshitz, M., Greenbaum, A., Feldman, Y., Greenbaum, S., Sokolov, A. P., & Golodnitsky, D. (2020). polymer/ceramic interface barriers: the fundamental challenge for advancing composite solid electrolytes for Li-ion batteries. *Journal of The Electrochemical Society*. https://doi.org/10.1149/1945-7111/abcd12
148. Tang, S., Guo, W., & Fu, Y. (2021). Advances in composite polymer electrolytes for lithium batteries and beyond. *Advanced Energy Materials, 11*(2), 2000802. https://doi.org/10.1002/aenm.202000802
149. Appetecchi, G. B., Croce, F., & Scrosati, B. (1995). Kinetics and stability of the lithium electrode in poly(methylmethacrylate)-based gel electrolytes. *Electrochimica Acta, 40*(8), 991–997. https://doi.org/10.1016/0013-4686(94)00345-2
150. Ahmed, H. T., Jalal, V. J., Tahir, D. A., Mohamad, A. H., & Abdullah, O. G. (2019). Effect of PEG as a plasticizer on the electrical and optical properties of polymer blend electrolyte MC-CH-$LiBF_4$ based films. *Results Phys*. https://doi.org/10.1016/j.rinp.2019.102735
151. Verdier, N., Lepage, D., Zidani, R., Prebe, A., Ayme-Perrot, D., Pellerin, C., & Rochefort, D. (2019). Cross-linked polyacrylonitrile-based elastomer used as gel polymer electrolyte in Li-Ion battery. *ACS Applied Energy Materials, 3*(1), 1099–1110. https://doi.org/10.1021/acsaem.9b02129
152. Tseng, Y. C., Hsiang, S. H., Lee, T. Y., Teng, H., Jan, J. S., & Kyu, T. (2021). In Situ polymerized electrolytes with fully cross-linked networks boosting high ionic conductivity and capacity retention for lithium Ion batteries. *ACS Applied Energy Materials, 4*(12), 14309–14322. https://doi.org/10.1021/acsaem.1c03011
153. Watanabe, M., Thomas, M. L., Zhang, S., Ueno, K., Yasuda, T., & Dokko, K. (2017). Application of ionic liquids to energy storage and conversion materials and devices. *Chemical Reviews, 117*(10), 7190–7239. https://doi.org/10.1021/acs.chemrev.6b00504
154. X. He, Y. Ni, Y. Hou, Y. Lu, S. Jin, H. Li, Z. Yan, K. Zhang, J. Chen, “Insights into the Ionic Conduction Mechanism of Quasi-Solid Polymer Electrolytes through Multispectral Characterization,” *Angewandte Chemie – International Edition*, 2021 https://doi.org/10.1002/anie.202107648.
155. Tripathi, A. K. (2021). Ionic liquid–based solid electrolytes (ionogels) for application in rechargeable lithium battery. *Materials Today Energy*. https://doi.org/10.1016/j.mtener.2021.100643
156. Li, W., Pang, Y., Liu, J., Liu, G., Wang, Y., & Xia, Y. (2017). A PEO-based gel polymer electrolyte for lithium ion batteries. *RSC Advances, 7*(38), 23494–23501. https://doi.org/10.1039/c7ra02603j
157. Zuo, X., Ma, X., Wu, J., Deng, X., Xiao, X., Liu, J., & Nan, J. (2018). Self-supporting ethyl cellulose/poly (vinylidene fluoride) blended gel polymer electrolyte for 5 V high-voltage lithium-ion batteries. *Electrochimica Acta, 271*, 582–590. https://doi.org/10.1016/j.electacta.2018.03.195
158. Zhang, M. Y., Li, M. X., Chang, Z., Wang, Y. F., Gao, J., Zhu, Y. S., & Huang, W. (2017). A sandwich PVDF/HEC/PVDF gel polymer electrolyte for lithium ion battery. *Electrochimica acta, 245*, 752–759. https://doi.org/10.1016/j.electacta.2017.05.154
159. Zhou, L., Wu, N., Cao, Q., Jing, B., Wang, X., Wang, Q., & Kuang, H. (2013). A novel electrospun PVDF/PMMA gel polymer electrolyte within situ TiO2 for Li-ion batteries. *Solid State Ionics, 249*, 93–97. https://doi.org/10.1016/j.ssi.2013.07.019
160. Kufian, M. Z., Aziz, M. F., Shukur, M. F., Rahim, A. S., Ariffin, N. E., Shuhaimi, N. E. A., & Arof, A. K. (2012). PMMA–LiBOB gel electrolyte for application in lithium ion batteries. *Solid State Ionics, 208*, 36–42. https://doi.org/10.1016/j.ssi.2011.11.032
161. Deng, X., Huang, Y., Song, A., Liu, B., Yin, Z., Wu, Y., & Cao, H. (2019). Gel polymer electrolyte with high performances based on biodegradable polymer polyvinyl alcohol composite lignocellulose. *Materials Chemistry and Physics, 229*, 232–241. https://doi.org/10.1016/j.matchemphys.2019.03.014
162. J. Thornton, “Environmental Impacts of Polyvinyl Chloride (PVC) Building Materials A briefing paper for the Healthy Building Network.” Available at: http://mts.sustainableproducts.com/SmaRT/ThorntonRevised.pdf
163. Huang, X. (2013). Cellular porous polyvinylidene fluoride composite membranes for lithium-ion batteries. *Journal of Solid State Electrochemistry, 17*(3), 591–597. https://doi.org/10.1007/s10008-012-1891-4
164. Gong, W., Wei, S., Ruan, S., & Shen, C. (2019). Electrospun coaxial PPESK/PVDF fibrous membranes with thermal shutdown property used for lithium-ion batteries. *Materials Letters, 244*, 126–129. https://doi.org/10.1016/j.matlet.2019.02.009
165. Sun, X., Liu, G., Li, R., Meng, Y., & Wu, J. (2021). Polyporous PVDF/TiO_2 photocatalytic composites for photocatalyst fixation, recycle, and repair. *Journal of the American Ceramic Society, 104*(12), 6290–6298. https://doi.org/10.1111/jace.18034
166. Zhao, Y., Fang, L. Z., Kang, Y. Q., Wang, L., Zhou, Y. N., Liu, X. Y., & Li, B. H. (2021). A novel three-step approach to separate cathode components for lithium-ion battery recycling. *Rare Metals, 40*, 1431–1436. https://doi.org/10.1007/s12598-020-01587-y
167. Xu, P., Chen, H., Zhou, X., & Xiang, H. (2021). Gel polymer electrolyte based on PVDF-HFP matrix composited with rGO-PEG-NH2 for high-performance lithium ion battery. *J Memb Sci*. https://doi.org/10.1016/j.memsci.2020.118660
168. Hosseinioun, A., Nürnberg, P., Schönhoff, M., Diddens, D., & Paillard, E. (2019). Improved lithium ion dynamics in crosslinked PMMA gel polymer electrolyte. *RSC Advances, 9*(47), 27574–27582. https://doi.org/10.1039/c9ra05917b
169. Kaminsky, W., & Franck, J. (1991). Monomer recovery by pyrolysis of poly(methyl methacrylate) (PMMA). *Journal of Analytical and Applied Pyrolysis, 19*, 311–318. https://doi.org/10.1016/0165-2370(91)80052-A
170. Chiellini, E., Corti, A., D’Antone, S., & Solaro, R. (2003). Biodegradation of poly (vinyl alcohol) based materials. *Progress in Polymer science, 28*(6), 963–1014.
171. Alipoori, S., Mazinani, S., Aboutalebi, S. H., & Sharif, F. (2020). Review of PVA-based gel polymer electrolytes in flexible

solid-state supercapacitors: Opportunities and challenges. *Journal of Energy Storage*. https://doi.org/10.1016/j.est.2019.101072

172. Piątek, J., Afyon, S., Budnyak, T. M., Budnyk, S., Sipponen, M. H., & Slabon, A. (2021). Sustainable Li-Ion batteries: chemistry and recycling. *Advanced Energy Materials, 11*(43), 2003456. https://doi.org/10.1002/aenm.202003456
173. Han, Y. H., Han, S. O., Cho, D., & Kim, H. I. L. (2007). Kenaf/polypropylene biocomposites: Effects of electron beam irradiation and alkali treatment on kenaf natural fibers. *Composite Interfaces*. https://doi.org/10.1163/156855407781291272
174. Wohlert, M., Benselfelt, T., Wågberg, L., Furó, I., Berglund, L. A., & Wohlert, J. (2022). Cellulose and the role of hydrogen bonds: not in charge of everything. *Cellulose, 29*, 1–23. https://doi.org/10.1007/s10570-021-04325-4
175. Du, Z., Su, Y., Qu, Y., Zhao, L., Jia, X., Mo, Y., Yu, F., Du, J., & Chen, Y. (2019). A mechanically robust, biodegradable and high performance cellulose gel membrane as gel polymer electrolyte of lithium-ion battery. *Electrochimica acta, 299*, 19–26. https://doi.org/10.1016/j.electacta.2018.12.173
176. Chun, S. J., Lee, S. Y., Doh, G. H., Lee, S., & Kim, J. H. (2011). Preparation of ultrastrength nanopapers using cellulose nanofibrils. *Journal of Industrial and Engineering Chemistry, 17*(3), 521–526. https://doi.org/10.1016/j.jiec.2010.10.022
177. Leo Edward, M., Dharanibalaji, K. C., Kumar, K. T., Chandrabose, A. R. S., Shanmugharaj, A. M., & Jaisankar, V. (2022). Preparation and haracterization of chitosan extracted from shrimp shell (Penaeus monodon) and chitosan-based blended solid polymer electrolyte for lithium-ion batteries. *Polymer Bulletin*. https://doi.org/10.1007/s00289-020-03472-1
178. Xu, D., Jin, J., Chen, C., & Wen, Z. (2018). From nature to energy storage: a novel sustainable 3D cross-linked chitosan-PEGGE-based gel polymer electrolyte with excellent lithium-ion transport properties for lithium batteries. *ACS Applied Materials & Interfaces, 10*(44), 38526–38537. https://doi.org/10.1021/acsami.8b15247
179. Osinska-Broniarz, M., Gieparda, W., Wesolek, D., Martyla, A., Przekop, R., & Sierczynska, A. (2015). Preparation and characterization of gel polymer electrolytes based on electrospun PLA/PHB membranes for Lithium-Ion batteries. *ECS Transactions, 70*(1), 79–88. https://doi.org/10.1149/07001.0079ecst
180. Zhu, M., Lan, J., Tan, C., Sui, G., & Yang, X. (2016). Degradable cellulose acetate/poly-l-lactic acid/halloysite nanotube composite nanofiber membranes with outstanding performance for gel polymer electrolytes. *J Mater Chem A Mater, 4*, 12136–12143. https://doi.org/10.1039/c6ta05207j
181. Piana, G., Ricciardi, M., Bella, F., Cucciniello, R., Proto, A., & Gerbaldi, C. (2020). Poly(glycidyl ether)s recycling from industrial waste and feasibility study of reuse as electrolytes in sodium-based batteries. *Chemical Engineering Journal*. https://doi.org/10.1016/j.cej.2019.122934
182. Kim, S. K., Yoon, Y., Ryu, J. H., Kim, J. H., Ji, S., Song, W., Myung, S., Lim, J., Jung, H. K., Lee, S. S., Lee, J., & An, K. S. (2020). Recyclable high-performance polymer electrolyte based on a modified methyl cellulose-lithium Trifluoromethanesulfonate salt composite for sustainable energy systems. *Chemsuschem, 13*(2), 376–384. https://doi.org/10.1002/cssc.201902756
183. Tenhaeff, W. E., & Kalnaus, S. (2015). Designing solid polymer composite electrolytes for facile lithium transport and mechanical strength. *Handbook of Solid State Batteries*. https://doi.org/10.1142/9789814651905_0007
184. Judez, X., Zhang, H., Li, C., Eshetu, G. G., González-Marcos, J. A., Armand, M., & Rodriguez-Martinez, L. M. (2017). Solid electrolytes for safe and high energy density lithium-sulfur batteries: Promises and challenges. *Journal of The Electrochemical Society, 165*(1), A6008. https://doi.org/10.1149/2.0041801jes
185. Huang, X., Liu, C., Lu, Y., Xiu, T., Jin, J., Badding, M. E., & Wen, Z. (2018). A Li-Garnet composite ceramic electrolyte and its solid-state Li-S battery. *Journal of Power Sources, 382*, 190–197. https://doi.org/10.1016/j.jpowsour.2017.11.074
186. Doux, J. M., Nguyen, H., Tan, D. H., Banerjee, A., Wang, X., Wu, E. A., & Meng, Y. S. (2020). Stack pressure considerations for room-temperature all-solid-state lithium metal batteries. *Advanced Energy Materials, 10*(1), 1903253. https://doi.org/10.1002/aenm.201903253
187. Doux, J. M., Yang, Y., Tan, D. H., Nguyen, H., Wu, E. A., Wang, X., & Meng, Y. S. (2020). Pressure effects on sulfide electrolytes for all solid-state batteries. *Journal of Materials Chemistry A, 8*(10), 5049–5055. https://doi.org/10.1039/c9ta12889a
188. Chen, A., Qu, C., Shi, Y., & Shi, F. (2020). Manufacturing strategies for solid electrolyte in Batteries. *Frontiers in Energy Research*. https://doi.org/10.3389/fenrg.2020.571440
189. Appetecchi, G. B., Alessandrini, F., Carewska, M., Caruso, T., Prosini, P. P., Scaccia, S., & Passerini, S. (2001). Investigation on lithium–polymer electrolyte batteries. *Journal of power sources, 97*, 790–794. https://doi.org/10.1016/S0378-7753(01)00609-7
190. Nguyen, H., Banerjee, A., Wang, X., Tan, D., Wu, E. A., Doux, J. M., Stephens, R., Verbist, G., & Meng, Y. S. (2019). Single-step synthesis of highly conductive Na3PS4 solid electrolyte for sodium all solid-state batteries. *Journal of Power Sources*. https://doi.org/10.1016/j.jpowsour.2019.05.031
191. Hafner, S., Guthrey, H., Lee, S. H., & Ban, C. (2019). Synchronized electrospinning and electrospraying technique for manufacturing of all-solid-state lithium-ion batteries. *Journal of Power Sources, 431*, 17–24. https://doi.org/10.1016/j.jpowsour.2019.05.008
192. Commarieu, B., Paolella, A., Daigle, J. C., & Zaghib, K. (2018). Toward high lithium conduction in solid polymer and polymer–ceramic batteries. *Current Opinion in Electrochemistry*. https://doi.org/10.1016/j.coelec.2018.03.033
193. Tan, S. J., Zeng, X. X., Ma, Q., Wu, X. W., & Guo, Y. G. (2018). Recent advancements in polymer-based composite electrolytes for rechargeable lithium batteries. *Electrochemical Energy Reviews, 1*(2), 113–138. https://doi.org/10.1007/s41918-018-0011-2
194. Li, S., Zhang, S. Q., Shen, L., Liu, Q., Ma, J. B., Lv, W., & Yang, Q. H. (2020). Progress and perspective of ceramic/polymer composite solid electrolytes for lithium batteries. *Advanced Science, 7*(5), 1903088. https://doi.org/10.1002/advs.201903088
195. Cavaliere, S., Subianto, S., Savych, I., Jones, D. J., & Rozière, J. (2011). Electrospinning: designed architectures for energy conversion and storage devices. *Energy and Environmental Science, 4*(12), 4761–4785. https://doi.org/10.1039/c1ee02201f
196. Pang, Y., Cao, Y., Chu, Y., Liu, M., Snyder, K., MacKenzie, D., & Cao, C. (2020). Additive manufacturing of batteries. *Advanced Functional Materials, 30*(1), 1906244. https://doi.org/10.1002/adfm.201906244
197. Strauss, E., Menkin, S., & Golodnitsky, D. (2018). Polymer electrolytes for printed batteries. *Printed Batteries: Materials, Technologies and Applications*, 80–111. DOI: https://doi.org/10.1002/9781119287902.ch4
198. Oliveira, J., Costa, C. M., & Lanceros-Méndez, S. (2018). Printed batteries: an overview printed batteries: materials. *Technologies and Applications*. https://doi.org/10.1002/9781119287902.ch1
199. Cheng, M., Jiang, Y., Yao, W., Yuan, Y., Deivanayagam, R., Foroozan, T., Huang, Z., Song, B., Rojaee, R., Shokuhfar, T., Pan, Y., Lu, J., & Shahbazian-Yassar, R. (2018). Elevated-temperature 3D printing of hybrid solid-state electrolyte for Li-ion batteries. *Advanced Materials, 30*(39), 1800615. https://doi.org/10.1002/adma.201800615

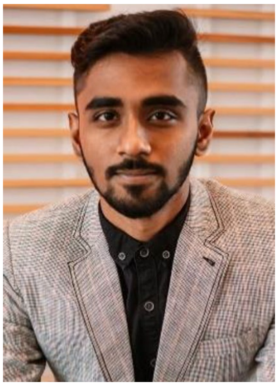
200. Yang, Y., Yuan, W., Zhang, X., Yuan, Y., Wang, C., Ye, Y., & Tang, Y. (2020). Overview on the applications of three-dimensional printing for rechargeable lithium-ion batteries. *Applied Energy*. https://doi.org/10.1016/j.apenergy.2019.114002
201. Costa, C. M., Gonçalves, R., & Lanceros-Méndez, S. (2020). Recent advances and future challenges in printed batteries. *Energy Storage Mater, 28*, 216–234. https://doi.org/10.1016/j.ensm.2020.03.012
202. Tehrani, Z., Korochkina, T., Govindarajan, S., Thomas, D. J., O'Mahony, J., Kettle, J., & Gethin, D. T. (2015). Ultra-thin flexible screen printed rechargeable polymer battery for wearable electronic applications. *Organic Electronics, 26*, 386–394. https://doi.org/10.1016/j.orgel.2015.08.007
203. Kim, S. H., Choi, K. H., Cho, S. J., Choi, S., Park, S., & Lee, S. Y. (2015). Printable Solid-state lithium-ion batteries: a new route toward shape-conformable power sources with aesthetic versatility for flexible electronics. *Nano Letters, 15*(8), 5168–5177. https://doi.org/10.1021/acs.nanolett.5b01394
204. D. Steingart, C. C. Ho, J. Salminen, J. W. Evans, and P. K. Wright, "Dispenser Printing of Solid Polymer-Ionic Liquid Electrolytes for Lithium Ion Cells," 6th International Conference on Polymers and Adnesives in Microelectronics and Photonics, Tokyo, Japan. 15–18 January, 2007. Doi: https://doi.org/10.1109/POLYTR.2007.4339179.
205. Cheng, M., Deivanayagam, R., & Shahbazian-Yassar, R. (2020). 3D Printing of electrochemical energy storage devices: a review of printing techniques and electrode/electrolyte architectures. *Batteries and Supercaps, 3*(2), 130–146. https://doi.org/10.1002/batt.201900130
206. Ponnada, S., Babu Gorle, D., Chandra Bose, R. S., Sadat Kiai, M., Devi, M., Venkateswara Raju, C., Baydogan, N., Nanda, K. K., Marken, F., & Sharma, R. K. (2022). Current insight into 3D printing in solid-state lithium-Ion batteries: a perspective. *Batteries & Supercaps*. https://doi.org/10.1002/batt.202200223
207. Shinde, V. V., Wang, Y., Salek, M. F., Auad, M. L., Beckingham, L. E., & Beckingham, B. S. (2022). Material design for enhancing properties of 3D printed polymer composites for target applications. *Technologies (Basel)*. https://doi.org/10.3390/technologies10020045
208. Zhu, C., Liu, T., Qian, F., Chen, W., Chandrasekaran, S., Yao, B., Song, Y., Duoss, E. B., Kuntz, J. D., Spadaccini, C. M., Worsley, M., & Li, Y. (2017). 3D printed functional nanomaterials for electrochemical energy storage. *Nano Today, 15*, 107–120. https://doi.org/10.1016/j.nantod.2017.06.007
209. Ngo, T. D., Kashani, A., Imbalzano, G., Nguyen, K. T. Q., & Hui, D. (2018). Additive manufacturing (3D printing): a review of materials, methods, applications and challenges. *Composites Part B: Engineering, 143*, 172–196. https://doi.org/10.1016/j.compositesb.2018.02.012
210. Chu, T., Park, S., & Fu, K. (2021). 3D printing-enabled advanced electrode architecture design. *Carbon Energy, 3*(3), 424–439. https://doi.org/10.1002/cey2.114
211. Kiran, A. S. K., Veluru, J. B., Merum, S., Radhamani, A. V., Doble, M., Kumar, T. S., & Ramakrishna, S. (2018). Additive manufacturing technologies: an overview of challenges and perspective of using electrospraying. *Nanocomposites, 4*(4), 190–214. https://doi.org/10.1080/20550324.2018.1558499
212. Pei, M., Shi, H., Yao, F., Liang, S., Xu, Z., Pei, X., & Hu, Y. (2021). 3D printing of advanced lithium batteries: a designing strategy of electrode/electrolyte architectures. *Journal of Materials Chemistry A, 9*(45), 25237–25257. https://doi.org/10.1039/d1ta06683h
213. McOwen, D. W., Xu, S., Gong, Y., Wen, Y., Godbey, G. L., Gritton, J. E., Hamann, T. R., Dai, J., Hitz, G. T., Hu, L., & Wachsman, E. D. (2018). 3D-printing electrolytes for solid-state batteries. *Advanced materials, 30*(18), 1707132. https://doi.org/10.1002/adma.201707132
214. Zhou, S., Usman, I., Wang, Y., & Pan, A. (2021). 3D printing for rechargeable lithium metal batteries. *Energy Storage Materials, 38*, 141–156. https://doi.org/10.1016/j.ensm.2021.02.041
215. Lewis, J. A. (2006). Direct ink writing of 3D functional materials. *Advanced Functional Materials, 16*(17), 2193–2204. https://doi.org/10.1002/adfm.200600434
216. Matsuzaki, R., Kanatani, T., & Todoroki, A. (2019). Multi-material additive manufacturing of polymers and metals using fused filament fabrication and electroforming. *Additive Manufacturing*. https://doi.org/10.1016/j.addma.2019.100812
217. Medellin, A., Du, W., Miao, G., Zou, J., Pei, Z., & Ma, C. (2019). Vat photopolymerization 3d printing of nanocomposites: A literature review. *Journal of Micro and Nano-Manufacturing*. https://doi.org/10.1115/1.4044288
218. Deiner, L. J., & Reitz, T. L. (2017). Inkjet and aerosol jet printing of electrochemical devices for energy conversion and storage. *Advanced Engineering Materials, 19*(7), 1600878. https://doi.org/10.1002/adem.201600878
219. He, Y., Chen, S., Nie, L., Sun, Z., Wu, X., & Liu, W. (2020). Stereolithography three-dimensional printing solid polymer electrolytes for all-solid-state lithium metal batteries. *Nano Letters, 20*(10), 7136–7143. https://doi.org/10.1021/acs.nanolett.0c02457
220. Chen, Q., Xu, R., He, Z., Zhao, K., & Pan, L. (2017). Printing 3D gel polymer electrolyte in Lithium-Ion microbattery using stereolithography. *Journal of the Electrochemical Society, 164*(9), A1852–A1857.
221. M. F. Norjeli, N. Tamchek, Z. Osman, I. S. Mohd Noor, M. Z. Kufian, and M. I. B. M. Ghazali, "Additive Manufacturing Polyurethane Acrylate via Stereolithography for 3D Structure Polymer Electrolyte Application," *Gels*, 2022 https://doi.org/10.3390/gels8090589.
222. Deiner, L. J., Jenkins, T., Howell, T., & Rottmayer, M. (2019). Aerosol Jet Printed Polymer Composite Electrolytes for Solid-State Li-Ion Batteries. *Advanced Engineering Materials, 21*(12), 1900952. https://doi.org/10.1002/adem.201900952
223. Blake, A. J., Kohlmeyer, R. R., Hardin, J. O., Carmona, E. A., Maruyama, B., Berrigan, J. D., Huang, H., & Durstock, M. F. (2017). 3D printable ceramic–polymer electrolytes for flexible high-performance li-ion batteries with enhanced thermal stability. *Advanced energy materials, 7*(14), 1602920. https://doi.org/10.1002/aenm.201602920
224. Kohlmeyer, R. R., Buskohl, P. R., Deneault, J. R., Durstock, M. F., Vaia, R. A., & Chen, J. (2014). Shape-reprogrammable polymers: Encoding, erasing, and re-encoding. *Advanced Materials, 26*(48), 8114–8119. https://doi.org/10.1002/adma.201402901
225. Fu, K., Wang, Y., Yan, C., Yao, Y., Chen, Y., Dai, J., & Hu, L. (2016). Graphene oxide-based electrode inks for 3D-printed lithium-ion batteries. *Advanced Materials*. https://doi.org/10.1002/adma.201505391
226. Wei, T. S., Ahn, B. Y., Grotto, J., & Lewis, J. A. (2018). 3D Printing of customized Li-Ion batteries with thick electrodes. *Advanced Materials, 30*(16), 1703027. https://doi.org/10.1002/adma.201703027
227. Muench, S., Burges, R., Lex-Balducci, A., Brendel, J. C., Jäger, M., Friebe, C., Wild, A., & Schubert, U. S. (2020). Printable ionic liquid-based gel polymer electrolytes for solid state all-organic batteries. *Energy Storage Materials, 25*, 750–755. https://doi.org/10.1016/j.ensm.2019.09.011
228. Cheng, M., Ramasubramanian, A., Rasul, M. G., Jiang, Y., Yuan, Y., Foroozan, T., Deivanayagam, R., Saray, M. T., Rojaee, R., Song, B., Yurkiv, V. R., Pan, Y., Mashayek, F., & Shahbazian-Yassar, R. (2021). Direct ink writing of polymer composite

electrolytes with enhanced thermal conductivities. *Advanced Functional Materials, 31*(4), 2006683. https://doi.org/10.1002/adfm.202006683

229. Gambe, Y., Kobayashi, H., Iwase, K., Stauss, S., & Honma, I. (2021). A photo-curable gel electrolyte ink for 3D-printable quasi-solid-state lithium-ion batteries. *Dalton Transactions, 50*(45), 16504–16508. https://doi.org/10.1039/D1DT02918E
230. Cho, K. G., Jang, S. S., Heo, I., Kyung, H., Yoo, W. C., & Lee, K. H. (2022). 3D printed solid-state composite electrodes and electrolytes for high-energy-density flexible microsupercapacitors. *J Energy Storage*. https://doi.org/10.1016/j.est.2022.105206
231. Jackson, S., & Dickens, T. (2021). Rheological and structural characterization of 3D-printable polymer electrolyte inks. *Polymer Testing*. https://doi.org/10.1016/j.polymertesting.2021.107377
232. Thakur, A., & Dong, X. (2020). Printing with 3D continuous carbon fiber multifunctional composites via UV-assisted coextrusion deposition. *Manuf Lett, 24*, 1–5. https://doi.org/10.1016/j.mfglet.2020.02.001
233. Thakur, A., & Dong, X. (2020). Additive manufacturing of 3D structural battery composites with coextrusion deposition of continuous carbon fibers. *Manuf Lett, 26*, 42–47. https://doi.org/10.1016/j.mfglet.2020.09.007
234. Seeba, J., Reuber, S., Heubner, C., Müller-Köhn, A., Wolter, M., & Michaelis, A. (2020). Extrusion-based fabrication of electrodes for high-energy Li-ion batteries. *Chemical Engineering Journal*. https://doi.org/10.1016/j.cej.2020.125551
235. Cook, J. A., Park, G. B., & McLoughlin, R. (1989). *MATERIALS FOR ELECTRICAL DEVICES* (Patent No. 4818643). United States Patent Office.
236. Duval, M., & Quebec, H. (1993). Process of coating by melt extrusion a solid polymer electrolyte on positive electrode of lithium battery (patent no. 5348824). united states patent office.
237. Reyes, C., Somogyi, R., Niu, S., Cruz, M. A., Yang, F., Catenacci, M. J., & Wiley, B. J. (2018). Three-dimensional printing of a complete lithium ion battery with fused filament fabrication. *ACS Applied Energy Materials, 1*(10), 5268–5279. https://doi.org/10.1021/acsaem.8b00885
238. Maurel, A., Grugeon, S., Fleutot, B., Courty, M., Prashantha, K., Tortajada, H., Armand, M., Panier, S., & Dupont, L. (2019). Three-dimensional printing of a LiFePO4/graphite battery cell via fused deposition modeling. *Scientific reports, 9*(1), 1–14. https://doi.org/10.1038/s41598-019-54518-y
239. Vinegrad, A., Ragones, H., Jayakody, N., Ardel, G., Goor, M., Kamir, Y., Dorfman, M. M., Gladkikh, A., Burstein, L., Horowitz, Y., Greenbaum, S., & Golodnitsky, D. (2021). Plasticized 3D-printed polymer electrolytes for lithium-ion batteries. *Journal of The Electrochemical Society*. https://doi.org/10.1149/1945-7111/ac39d5
240. Ragones, H., Menkin, S., Kamir, Y., Gladkikh, A., Mukra, T., Kosa, G., & Golodnitsky, D. (2018). Towards smart free form-factor 3D printable batteries. *Sustainable Energy & Fuels, 2*(7), 1542–1549. https://doi.org/10.1039/c8se00122g
241. Ragones, H., Vinegrad, A., Ardel, G., Goor, M., Kamir, Y., Dorfman, M. M., Gladkikh, A., & Golodnitsky, D. (2019). On the road to a multi-coaxial-cable battery: development of a novel 3D-printed composite solid electrolyte. *Journal of The Electrochemical Society*. https://doi.org/10.1149/2.0032007jes
242. Katcharava, Z., Marinow, A., Bhandary, R., & Binder, W. H. (1859). 3D Printable composite polymer electrolytes: influence of SiO_2 nanoparticles on 3D-printability. *Nanomaterials, 12*(11), 2022. https://doi.org/10.3390/nano12111859
243. Maurel, A., Armand, M., Grugeon, S., Fleutot, B., Davoisne, C., Tortajada, H., Courty, M., Panier, S., & Dupont, L. (2020). Poly (ethylene oxide)− LiTFSI solid polymer electrolyte filaments for fused deposition modeling three-dimensional printing. *Journal of The Electrochemical Society*. https://doi.org/10.1149/1945-7111/ab7c38
244. Verdier, N., Foran, G., Lepage, D., Prébé, A., Aymé-Perrot, D., & Dollé, M. (2021). Challenges in solvent-free methods for manufacturing electrodes and electrolytes for lithium-based batteries. *Polymers, 13*(3), 323. https://doi.org/10.3390/polym13030323
245. Jung, K. N., Shin, H. S., Park, M. S., & Lee, J. W. (2019). Solid-state lithium batteries: bipolar design, fabrication, and electrochemistry. *ChemElectroChem, 6*(15), 3842–3859. https://doi.org/10.1002/celc.201900736
246. Brown, E., Yan, P., Tekik, H., Elangovan, A., Wang, J., Lin, D., & Li, J. (2019). 3D printing of hybrid MoS2-graphene aerogels as highly porous electrode materials for sodium ion battery anodes. *Materials & Design*. https://doi.org/10.1016/j.matdes.2019.107689
247. Down, M. P., Martínez-Periñán, E., Foster, C. W., Lorenzo, E., Smith, G. C., & Banks, C. E. (2019). Next-generation additive manufacturing of complete standalone sodium-ion energy storage architectures. *Advanced Energy Materials, 9*(11), 1803019. https://doi.org/10.1002/aenm.201803019
248. Schwaar, C. (2023, January 31). *Additive manufacturing for batteries of the future: Will 3D printing transform battery making?* Forbes. Retrieved April 12, 2023, from https://www.forbes.com/sites/carolynschwaar/2023/01/30/additive-manufacturing-for-batteries-of-the-future-will-3d-printing-transform-battery-making/?sh=5bb74f063795
249. Vlad Khaustovich, “3D Printing & Rapid Prototyping Services Breaking the mold: Developing technologies and a growing customer base are expected to boost revenue,” 2023.,Report no.: OD 4581.
250. Arielle Rose, “Battery Manufacturing in the US,” IBIS World, September 2022., Report No.: 33591.
251. Matt Pantalon, “Lithium Battery Manufacturing. In the US”, IBIS World, January 2023, OD4499.
252. Schmaltz, T., Wicke, T., Weymann, L., Voß, P., Neef, C., & Thielmann, A. (2022). Solid-State Battery Roadmap 2035+.
253. *Sakuu to become publicly traded company Via Business Combination with plum acquisition corp.. I.* Business Wire. (2023, March 2). Retrieved April 12, 2023, from https://www.businesswire.com/news/home/20230302005883/en/
254. *Blackstone's major milestone in 3D-printing solid-state batteries*. Business Wire. (2021, April 12). Retrieved April 12, 2023, from https://www.businesswire.com/news/home/20210412005715/en/Blackstone%E2%80%99s-major-milestone-in-3D-printing-solid-state-batteries
255. Arielle Rose, “Battery Recycling in the US.”, IBIS World, November 2022, Report No; OD4511
256. Kunz, T. (2019). DOE launches its first lithium-ion battery recycling R&D center: ReCell. Retrieved April 12, 2023, from Argonne National Laboratory. Available at https:// www. anl.gov/article/doe-launches-its-first-lithiumion-battery-recycling-rd-center-recell.
257. Cong, L., Liu, W., Kong, S., Li, H., Deng, Y., & Ma, H. (2021). A review on end-of-use management of spent lithium-ion batteries from sustainability perspective. *Journal of Manufacturing Science and Engineering*, 1–35. DOI: https://doi.org/10.1115/1.4050925
258. Cong, L., Zhou, K., Liu, W., & Li, R. (2023). Retired lithium-ion battery pack disassembly line balancing based on precedence graph using a hybrid genetic-firework algorithm for

remanufacturing. *Journal of Manufacturing Science and Engineering*. https://doi.org/10.1115/1.4056572

259. Li, H., Cong, L., Ma, H., Liu, W., Deng, Y., & Kong, S. (2022). Screening of retired lithium-Ion batteries using incremental capacity charging curve-based residual capacity estimation method for facilitating sustainable circular lithium-ion battery system. *Journal of Manufacturing Science and Engineering*. https://doi.org/10.1115/14051677
260. Cong, L., Zhao, F., & Sutherland, J. W. (2019). A design method to improve end-of-use product value recovery for circular economy. *Journal of Mechanical Design, 10*(1115/1), 4041574.



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