



## HIGHLIGHTED PAPER



# Structural batteries: Advances, challenges and perspectives

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The development of light-weight batteries has a great potential value for mobile applications, including electric vehicles and electric aircraft. Along with increasing energy density, another strategy for reducing battery weight is to endow energy storage devices with multifunctionality – e.g., creating an energy storage device that is able to bear structural loads and act as a replacement for structural components such that the weight of the overall system is reduced. This type of batteries is commonly referred to as “structural batteries”. Two general methods have been explored to develop structural batteries: (1) integrating batteries with light and strong external reinforcements, and (2) introducing multifunctional materials as battery components to make energy storage devices themselves structurally robust. In this review, we discuss the fundamental rules of design and basic requirements of structural batteries, summarize the progress made to date in this field, examine potential avenues and sources of inspiration for future research, and touch upon challenges remaining in this field such as safety, costs, and performance stability. Though more fundamental and technical research is needed to promote wide practical application, structural batteries show the potential to significantly improve the performance of electric vehicles and devices.

**Keywords:** Structural batteries; Transportation electrification; Lithium batteries; Mechanical properties; Simulation

## Introduction

Electrification of transportation is one of the key technologies to reduce CO<sub>2</sub> emissions and address the imminent challenge of climate change [1,2]. Currently, lithium-ion batteries (LIBs) are widely adopted for electrification, such as in electric vehicles (EV) and electric aircraft, due to their attractive performance among various energy storage devices [3–6]. Nevertheless, the energy density of state-of-the-art (SOA) LIBs is still not satisfactory which limits the mileage of electric transportation. To solve this issue, tremendous efforts have been devoted to developing batteries with higher energy density, such as high-voltage cathodes [7] and high-specific-capacity electrodes [8–13]. Along with this important direction, an alternative strategy is to develop

multifunctional batteries. If batteries are capable of carrying appreciable mechanical loads and serving as structural components, the overall system weight will be reduced and the mileage can be extended (Fig. 1(a)) [14,15]. For example, it has been demonstrated that by replacing structural parts such as roofs with structural batteries in electric vehicles, 20% of the corresponding mass can be saved, which further allows more batteries to increase mileages (Fig. 1(b)) [16,17]. Moreover, as structural batteries can distribute across the entire body of a system instead of concentrating at one location (e.g. chassis of electric vehicles), such distributed energy storage designs could enhance the safety and resilience of the entire system. This concept of “structural batteries” has drawn increasing interest among academia and industry in recent years [18].

The cardinal requirements of structural batteries are adequate energy density and strong mechanical properties. However, SOA LIBs, consisting of alternative stacks of electrode and separator

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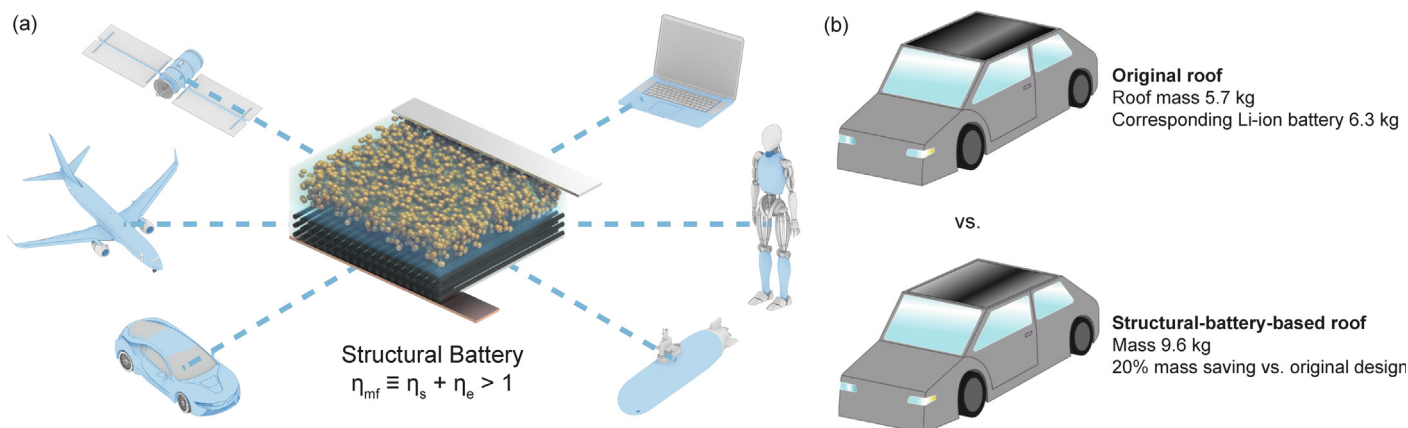


FIGURE 1

(a) Various applications of structural batteries to save weight or increase energy storage at the system levels. Examples include: electric vehicles, consumer electronics, robotics, satellites, aircraft, and marine systems. (b) Schematic of mass saving results from using structural batteries in the roof of an electric vehicle. Reproduced with permission [16]. Copyright © 2019 IOP Publishing.

layers filled with liquid electrolytes and sealed inside a pouch bag or a metal case, do not satisfy the mechanical demands because they are not built for load carrying [19]. In a commercial LIB, the electrode is made of active material particles, carbon additives, and polymer binders coated onto both sides of a metal foil. As the active material layer in an electrode is made of granular particles with a porosity of ca. 30%, the compressive properties of electrodes are much lower than conventional structural materials [20], and the tensile properties are also compromised by the small portions of binders and the weak adhesion among active material particles [21–23]. The separator is a porous polymer film that has good electrolyte permeability, but poor mechanical properties for the load-carrying purpose [24]. In addition, the electrode and separator layers are stacked together without binding, indicating a poor load transfer between layer components which also degrades the mechanical properties of batteries such as flexural stiffness [25]. Consequently, new designs are in request to strengthen batteries mechanically, and two categories of strengthening methods have been proposed.

The first one is at the cell-level, focusing on sandwiching batteries between robust external reinforcement composites such as metal shells and carbon fabric sheets (Fig. 2(a)). In such designs, the external reinforcement is mainly responsible for the load-carrying without contributions to energy storage, and the battery mainly functions as a power source and bears limited loads [26–28]. Therefore, the weight saving from this strategy is limited. On the other side, out of its good compatibility for mass production, this strategy draws increasing attention among electric vehicle companies with more and more cells directly integrated with vehicle parts such as chassis and frames, which is called the cell-to-chassis (CTC) approach. The performance can be further improved by better spatial arrangement and by developing new materials and manufacturing processes.

The second one is at the material-level, where multifunctional materials are developed to serve as both load-carrying components and functional battery components (Fig. 2(b)). Several notable strategies include using carbon fibers as strong current collectors and electrodes, solid electrolytes to enhance mechanical properties and load transfer, and mechanically robust separa-

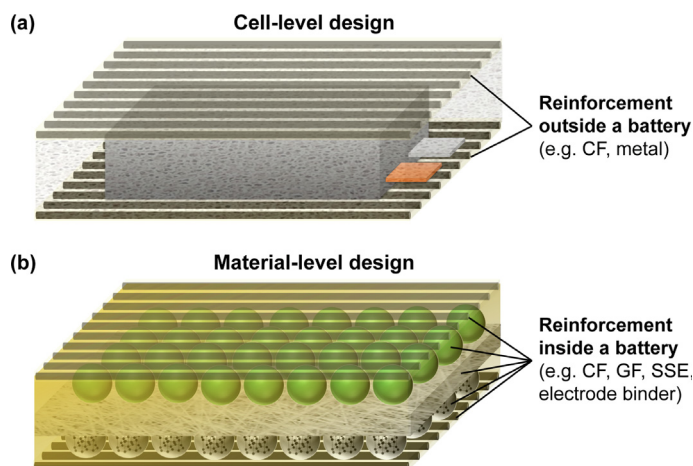


FIGURE 2

The schematic of structural batteries with (a) cell-level designs and (b) material-level designs, where different parts of a battery can be reinforced. Examples of reinforcements include carbon fiber (CF), glass fiber (GF), solid-state electrolyte (SSE), or electrode binder.

tors and binders. The material development can help enhance the intrinsic mechanical properties of batteries for structural applications but require careful designs so that electrochemical performance is not compromised.

In this review, we target to provide a comprehensive summary of recent developments in structural batteries and our perspectives. In the following sections, we will first discuss the fundamentals of structural batteries, including how to evaluate their multifunctionalities, key requirements on properties, and potential applications. Then we will focus on recent experimental and computational advances in structural batteries. Finally, key challenges and prospects of structural batteries towards practical applications will be discussed.

## Fundamentals of structural energy storage devices

Structural energy storage devices function as both a structural component and an energy storage device simultaneously. There-

fore, a system (e.g. a vehicle) with such multifunctional devices can have better mass efficiency and longer operating time. However, such a benefit is not always guaranteed by simply endowing energy storage function to a structural component or mechanically enhancing an energy storage device. A successful structural energy storage device must meet various property requirements based on its targeted application. Therefore, it is important to first understand how to evaluate a structural energy storage device design and what properties to be considered.

### Assessment methods on the multifunctionality of structural energy storage devices

In 2000, Ashby proposed a value function to evaluate the multifunctional performance of a general system as a combination of different user-defined performance metrics which are related to material properties [29]. The approach can be applied to any multifunctional system and is the foundation of most works on structural energy storage assessment. Thomas and Qidwai later extended Ashby's methodology to demonstrate the feasibility of improving the flight endurance time of unmanned air vehicles (UAVs) by integrating lithium batteries into structures as multifunctional devices [30,31].

To evaluate the mass or volume saving of structural composite capacitors, in 2010 O'Brien et al. brought up the term "multifunctional efficiency", which can be generalized to all energy storage systems and is defined as [32,33]:

$$\eta_{mf} \equiv \eta_s + \eta_e > 1 \quad (1)$$

This criterion is an analogy to Ashby's, where the multifunctional efficiency  $\eta_{mf}$  is the sum of structural and energy performance metrics. Here structural efficiency  $\eta_s$  and energy efficiency  $\eta_e$  are defined as the ratio of specific stiffness and specific energy of multifunctional systems versus those of SOA monofunctional counterparts, respectively. Apparently,  $\eta_s$  and  $\eta_e$  can be generalized to other interested specific structural and energy parameters, such as specific shear modulus and specific power. Therefore, a mass reduction is expected in a vehicle if a structural energy storage device is utilized with an  $\eta_{mf} > 1$ . The equation also indicates that simply combining individual structural and energy storage components into one device will practically not result in a beneficial multifunctional efficiency due to the extra mass and volume of parts like junctions and packages [34]. So rational designs to integrate monofunctional components or developments of components that can considerably carry loads and provide power simultaneously are the key to realizing practical structural energy storage devices.

The details of multifunctional efficiency were later further generalized by Snyder and coworkers [35]. They pointed out that the performance metrics could be more complicated in practice as multiple mechanical and electrochemical properties need to be satisfied simultaneously. For example, Young's modulus  $\bar{E}_{mf}$ , shear modulus  $\bar{G}_{mf}$ , tensile strength  $\bar{\sigma}_{mf}$ , energy  $\bar{w}_{mf}$  and power  $\bar{p}_{mf}$  per mass are common key parameters for a structural energy storage device. Then considering possible limiting factors, the structural efficiency  $\eta_s$  and energy efficiency  $\eta_e$  will have a more complex but rigorous definition to better evaluate the multifunctional efficiency of the proposed device:

$$\eta_s \equiv \min \left\{ \frac{\bar{E}_{mf}}{E}, \frac{\bar{G}_{mf}}{G}, \frac{\bar{\sigma}_{mf}}{\sigma} \right\} \quad (2)$$

$$\eta_e \equiv \min \left\{ \frac{\bar{w}_{mf}}{w}, \frac{\bar{p}_{mf}}{p} \right\} \quad (3)$$

where symbols with no subscripts are corresponding values of monofunctional counterparts in a multifunctional device. With this new universal notion, one can comprehensively assess a structural energy storage device based on practical structural and power requirements.

Recently, different views are presented to interpret the benefits of a structural energy storage device. Johannisson and coworkers first directly focused on the mass comparison between a multifunctional device and equivalent monofunctional ones [16]. Then they further proposed an idea of residual specific properties to avoid overlooking the gain of a multifunctional system, which allows people to evaluate its benefits in all aspects instead of focusing on only the most significant parameters [36]. Zhou et al. also developed a more elaborate model of multifunctional efficiency to encompass the effects of materials anisotropy, complex geometries, and diverse loading conditions [37].

Besides discussions on multifunctionality assessment above, it is crucial to point out that practical designs of structural energy storage devices can be more complex. Multiple properties usually need to be satisfied simultaneously given that the practical serving environment and economic considerations can be intricate. Accordingly, more performance metrics need to be considered for  $\eta$  based on the demands in specific cases, which will be discussed in the next section.

### Potential applications and corresponding key requirements

Structural energy storage devices can serve as various components in a system to enable more efficient designs, and their best solutions are system and application-specific. Therefore, it is important to first understand potential applications and corresponding required performance metrics.

Currently, structural batteries are attractive for automotive [16,17], aerospace [38–40], and marine systems [41]. For electric vehicles, their roofs, door panels, boot lids, chassis, and even seat frames can be integrated with structural batteries. The former three parts are not designed to have appreciable load-carrying capability compared to the latter two. Hence adequate bending rigidity is the key parameter for structural batteries used in roofs, doors, and boot lids, which maintains their morphology against external pressures. In contrast, high stiffness and strength are necessary for those used in chassis and seats, which bear loads in practice [16]. Similarly, floors, interior wall panels, and wings of aerospace systems, and floors and hull skins of marine systems can also adopt structural energy storage devices. In aerospace applications, mass is the critical factor to consider, whereas volume is the constraint in marine systems, hence their performance metrics should focus on mass-based and volume-based ones, respectively.

In addition, structural batteries can also serve as cases and other mechanical parts in devices with limited space to enhance their operational time and/or reduce weight, such as consumer electronics, robots, and medical microelectronics [42,43]. In these applications, developers need to first assess their key

requirements as the aforementioned analysis on vehicles to guide designs. Moreover, the designs of structural energy storage devices should not compromise performance other than mechanical ones, which include but are not limited to impact and chemical resistance, fatigue, safety, operating temperature windows, and costs. Besides, structural energy storage could also affect the performance at the system level, e.g. its consequent distributed energy storage may enhance the safety and resilience of systems while bringing more weight of battery management systems. Overall, comprehensive considerations about what properties to include in multifunctional efficiencies are vital in structural energy storage device designs and evaluations.

### Development of structural batteries

Mechanical properties of batteries are often 2–3 orders of magnitude lower than load-bearing structural components for aircraft or ground transportation [26]. Hence, to develop structural batteries, strategies for mechanical reinforcement are required. They can be divided into two main categories: (1) cell-level designs, where the reinforcement does not play a major role in energy storage (Fig. 2(a)), and (2) material-level designs, where multifunctional materials are used for both energy storage and structural reinforcement (Fig. 2(b)).

In cell-level designs, batteries are reinforced by external components, such as carbon fiber fabric-based laminates, or the cell configuration is modified to enhance mechanical properties, as reviewed in section *Cell-level designs*. On the other side, in material-level designs, one or multiple battery components are designed with improved mechanical performance, such as electrodes, current collectors, electrolytes, and even interfaces. Material-level designs and material developments for the reinforcement of different cell components are discussed in sections *CF-based material-level designs*, *Structural separators, binders, and lamination designs*, and *Solid-state electrolytes (SSE)*, followed by a summary of simulation progress in section *Simulations*.

#### Cell-level designs

One practical example of cell-level designs is the structural battery pack of the new EV model Y from Tesla (Fig. 3(a)) [44], which leads to a 10% mass reduction, a 14% range increase, and fewer parts [45]. The battery pack acts as a body structure, that links the front and rear underbody parts of the EV due to its improved mechanical properties by implementing 4680-type cylindrical battery cells into a lightweight polyurethane (PU) honeycomb design, which is encapsulated between aluminum and steel face sheets, enabling the transfer of shear stress to provide an overall improved torsional rigidity.

Besides this commercialized strategy by Tesla, one future strategy in cell-level designs is to encapsulate batteries in a carbon fiber (CF) composite [28,41,46–52]. Batteries embedded in CF laminates can be manufactured with conventional composite manufacturing techniques at lower temperatures and pressures since temperature-sensitive liquid electrolytes are involved. For instance, a wet hand-layup and the vacuum bag resin infusion (VBRI) technique was used for liquid electrolyte containing Li-polymer cells, embedded in cut-outs of CF laminates and cured at 20 °C for 24 hours to avoid any damage to the batteries due

to high pressure or elevated temperature [28], as shown in Fig. 3(b).

The same technique can be used for the preparation of a CF laminate sandwich with cells embedded in a foam core [47,48]. For example, Thomas et al., put LIB pouch cells and a closed-cell styrene acrylonitrile (SAN) foam between CF laminate face sheets and obtained a specific energy of 45 Wh kg<sup>-1</sup> and a flexural rigidity of 985 N m<sup>2</sup> [41]. Similarly, 94 Wh kg<sup>-1</sup> was demonstrated by this strategy with five pouch cells embedded in a comparable sandwich structure [28]. A slightly modified version of this encapsulation strategy is to use CF face sheets as both reinforcement and battery packaging, which saves 5–10 wt.% of the battery as no extra packaging is needed. This was realized by embedding thin-film lithium batteries (TFLB) in cut-outs of composite laminates [49,50]. The principle of this strategy is shown in Fig. 3(c). However, the long-term stability of CF composites against air permeation should be carefully examined as ultra-low permeation of air (moisture, oxygen) less than 20 ppm [53] is required for LIBs.

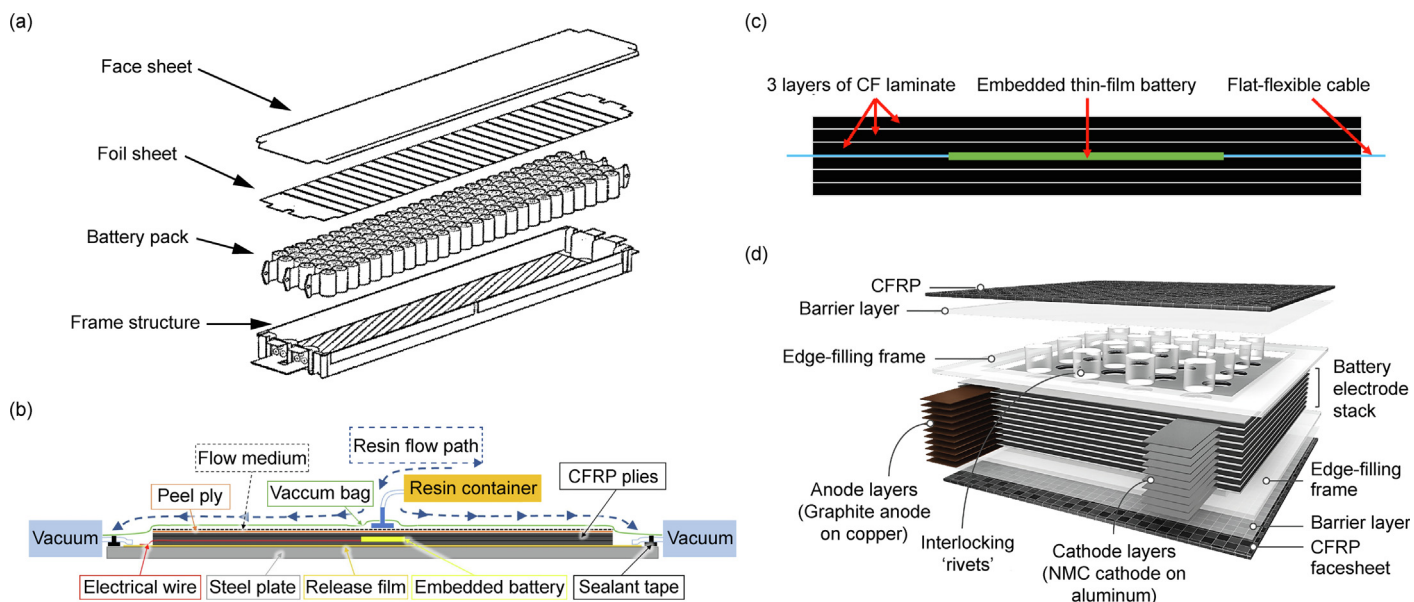
Structural batteries with embedded TFLBs can withstand higher processing temperatures and pressures in an autoclave. TFLBs have a thickness of < 300 μm (typically ca. 150 μm) and can be inserted between laminate layers [49,54]. A polymer-sealed all-solid-state TFLB pack has been embedded between two layers of CF prepregs and cured in an autoclave at 120 °C without any electrochemical performance loss or cell damage [50]. Besides the possibility of autoclave manufacturing, another advantage of TFLBs is their mechanical flexibility which allows the manufacturing of curved or more complex shapes of structural batteries.

The encapsulation strategy can greatly enhance the tensile properties of batteries but has limited improvement on the flexural properties of batteries, since different layers in batteries can slide over each other and dissipate stress. To avoid such sliding and enhance flexural rigidity, Fu-Kuo Chang developed an interlocking rivet strategy, where interlocking polymer rivets perpendicular to the plane were inserted in a perforated LIB stack and sandwiched between CF laminate face sheets, as shown in Fig. 3(d) [46]. A graphite/NMC cell with such a design shows a high specific energy of 131 Wh kg<sup>-1</sup> with 80% capacity retention after 800 cycles, and the flexural rigidity increased from 3 N m<sup>2</sup> to 12 N m<sup>2</sup> due to the improved load transfer between layers. Later, their group proposed a method to resolve the high impedance issue in their strengthened batteries [55].

Although the flexural rigidity could be improved by the interlocking rivets, the addition of its mass and reduction of active material reduced the energy density by about 40%. Zhang et al. [52] stacked batteries with NMC cathode and mesocarbon microbeads (MCMB) anode transversally between two Al face sheets and C-channels as structural components, in order to improve the bending stiffness. High-performance epoxy adhesive was used to enclose the battery core and bond it to the face sheets. With this approach, a structural battery with a specific energy of 102 Wh kg<sup>-1</sup> and a high flexural rigidity of 781 N m<sup>2</sup> was achieved, that could still be operated under a bending force of 1060 N without significant electrochemical performance loss.

Mechanical properties of CF-reinforced structural batteries with a cell-level design were reported with an elastic modulus





**FIGURE 3**

(a) The design of a structural battery pack from Tesla [44]. (b) A Schematic illustration of the vacuum bag resin infusion (VBRI) technique for the manufacturing of a structural battery. Reproduced with permission [28]. Copyright © 2020 Elsevier. (c) The principle of embedding a thin-film battery between CF laminates. (d) Reinforcement of a battery stack with interlocking rivets and CF face sheets. Reproduced with permission [46]. Copyright © 2019 Elsevier.

of up to 11.6 GPa and a flexural strength of 113 MPa [47]. Compared to high-performance carbon fiber reinforced polymers (CFRPs), which exhibit a modulus of 60–70 GPa and strength between 600–900 MPa for laminates with  $[0^\circ/90^\circ]$  fiber orientation [56,57], there is still a large room for improvement.

Generally, it is believed that cell-level designs have limited improvement in the multifunctional efficiency because both batteries and reinforcements mainly perform monofunctionally [58]. However, such a strategy may be a good answer for the near future because it is closer to the practical application compared to material-level designs which adopt multifunctional materials and need more tests to prove their benefits and long-term performance. We believe that better engineering designs such as directly integrating cells into vehicle parts, as demonstrated in industry, can take more advantage of cell-level designed structural batteries. For the next step, simpler manufacturing processes, high safety, low costs, and easy maintenance are the key targets for a practical cell-level designed structural battery, with more details discussed in section *Challenges and outlooks*.

### Carbon fiber-based material-level designs

#### CF-based structural electrodes

Carbon fibers (CFs) can act as both active materials and current collectors in battery electrodes. When used as the active material, CFs can serve as the anode for LIBs, and need to be supported and surrounded by an ion conductive electrolyte. The turbostratic microstructure of CFs, consisting of both amorphous and graphitic carbon, allows the intercalation of Li-ions under radial (8–13%) and longitudinal (1%) expansions, comparable to those occurring in graphite [59]. Similarly, insertion of K-ions [60] and Na-ions [61] in CFs represent further potential cell chemistries for CF-based structural batteries but have not been reported yet.

Modern polyacrylonitrile (PAN)-based CFs exhibit high intrinsic mechanical properties, with Young's moduli of 250–600 GPa and tensile strength of 4–7 GPa for an individual fiber [62]. During lithiation of the fiber in one study, the longitudinal elastic modulus was reduced from 290 GPa to 255 GPa (-12%) which fully recovered after delithiation, while the transversal modulus of the CF reversibly doubled from 7.6 GPa to 15 GPa [63]. The lithiation has a limited effect on the elastic tensile properties during cycling (e.g. 9% loss after 1,000 cycles), however, the ultimate tensile strength can be reduced by up to 20%, due to remaining Li-ions in the fiber [64].

Induced stresses and deformations due to CF electrode expansion should be taken into account in the design of structural batteries [65]. Regarding electrochemical performance, the specific capacity of different grades of commercial CFs varies greatly during delithiation (22–358 mAh g<sup>-1</sup>) [66,67] and can approach the theoretical capacity of graphite (372 mAh g<sup>-1</sup>) at low C-rates [59]. It should be noted that most reports showed that CFs have a low initial coulombic efficiency (ICE) of 40–89% [68–70], which increases to > 99% after 5–10 cycles.

Asp et al. utilized the CF with natural surface as an active structural anode for structural batteries. With polymer electrolytes, a specific energy of 24 Wh kg<sup>-1</sup> (or 106 Wh kg<sup>-1</sup> based on active material only) is achieved together with a relatively high modulus of 25 GPa and tensile strength of 300 MPa [14]. The specific energy can be further enhanced by reducing the thickness of polymer electrolytes. Moyer et al., reported graphite coating on CF to improve the capacity of carbon anode and reached a specific energy of 35 Wh kg<sup>-1</sup> with a liquid electrolyte. However, due to the use of liquid electrolyte, its mechanical performance was significantly reduced, and the Young's modulus and tensile strength were only 1.8 GPa and 213 MPa, respectively [71].

CFs can also act as the structural current collector in a battery, as they have a reasonable electrical conductivity of  $10^2$ – $10^3$  S  $\text{cm}^{-1}$  [72]. For instance,  $\text{LiFePO}_4$  (LFP) coatings on CFs with good adhesion can be produced by electrophoretic deposition [73,74] or coextrusion of CFs with an LFP-doped matrix [75]. Interfacial adhesion of LFP particles on the CF surface can be improved by an additional PAN layer to bridge the active material and the polymer matrix [76]. Other materials, such as nickel sulfides [77], polysulfides [78], or  $\text{LiMn}_{0.97}\text{Al}_{0.03}\text{O}_2$  [79] on CFs have been described. A combination of CF anode and LFP-coated CF cathode was successfully demonstrated [14,73,78] with a specific energy of up to  $222 \text{ Wh kg}^{-1}$  based on active material mass [73]. The electrodeposition of polysulfide onto CFs enables improved adhesion and reduces the risk of deterioration or delamination of active materials from the substrate under external mechanical load. Huang et al., demonstrated a structural Li-S battery with molten Li-infused CF anode and  $\text{Li}_2\text{S}_8/\text{CF}$  cathode, which achieved a specific energy of  $43 \text{ Wh kg}^{-1}$  based on the total mass of battery components [78].

Besides Li-ion batteries (LIB), CF-based current collectors have also been used in structural Zn-ion batteries (ZIB). Using a  $\text{MnO}_2/\text{CF}$  cathode, specific energy of  $182 \text{ Wh kg}^{-1}$  with Zn@CF anode [80] and  $205 \text{ Wh kg}^{-1}$  with a Zn sheet anode [81], based

on the mass of active materials, have been reported recently. The combination of internal ( $\text{MnO}_2/\text{CF}$ , GF separator, Zn sheet anode) and external reinforcement (CF and GF laminates), shown in Fig. 4(a), lead to a high flexural strength of 230 MPa and reduced cell capacity under tensile stress (Fig. 4(b)). Designs of structural batteries with alternative chemistries are also discussed in recent review articles [58,82].

#### CF as internal electrodes and current collectors vs as external reinforcement

The mechanical performance of a composite is influenced by the design and properties of its components. For structural batteries, the rigidity under bending is an important engineering criterion, which depends on whether the reinforcing components are placed inside or outside the battery. However, the potential gain in energy density of externally reinforced structural batteries is limited by the additional mass of reinforcement and its mechanical properties, whereas integrated multifunctional structural components inside the battery ideally do not add extra weight to it.

Multiple studies suggest that external reinforcement strategies – embedding the battery between structural layers in a sandwich design – tend to achieve higher specific energy and mechanical performance, since the two different components both keep

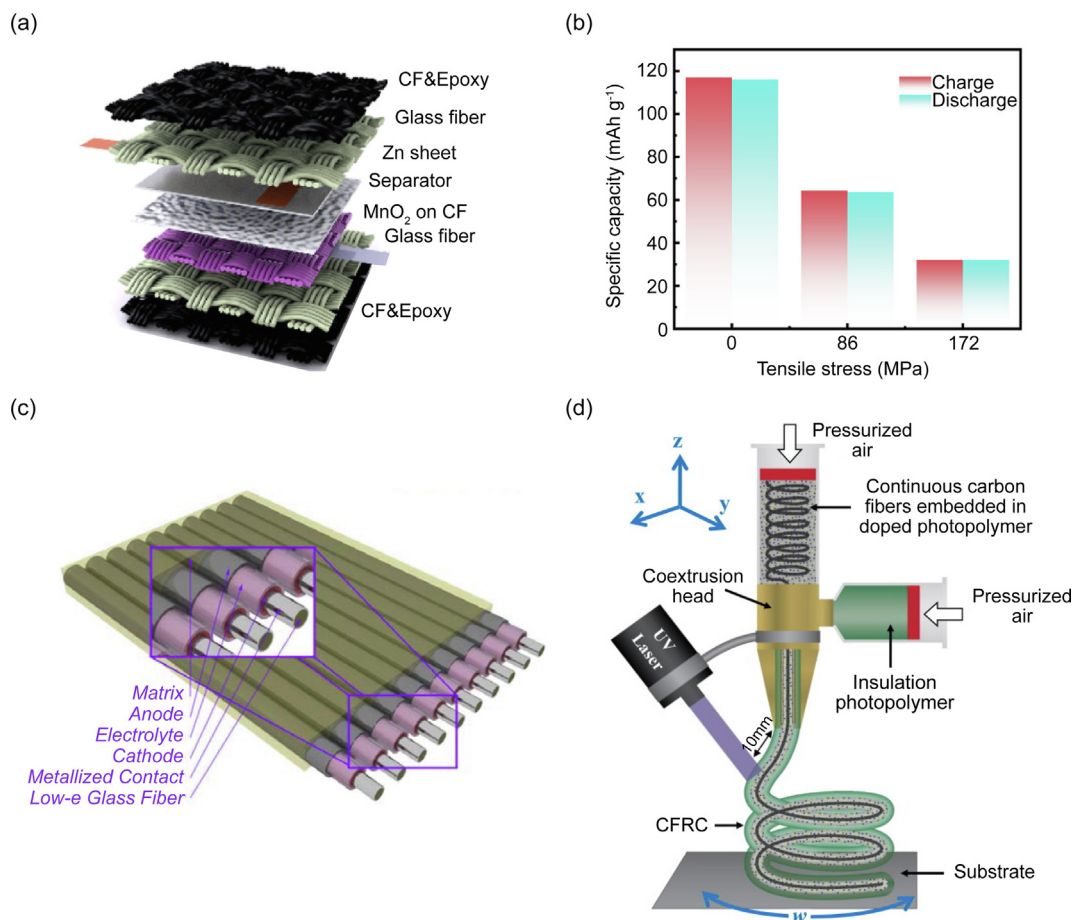


FIGURE 4

(a) A material-level design of a Zn-ion structural battery and (b) its specific capacity under tensile stress. Reproduced with permission [81]. Copyright © 2022 Elsevier. (c) Individual CFs with thin-film battery coating for a structural battery design [83]. (d) Coextrusion deposition of CF and photopolymer resin to produce CF-reinforced composite structures. Reproduced with permission [84]. Copyright © 2020 Elsevier.

their main functions. They were optimized for i.e. energy storage in batteries and mechanical stability of structural materials for multiple decades. Hopkins et al. [85] published a meta-analysis of studies, comparing specific energy and elastic modulus of reported structural batteries, indicating that decoupled approaches of cell integration outperform coupled versions. However, the novel development of multifunctional materials, which combine electrochemical and mechanical performance, drives the improvement of internally reinforced batteries to a competitive level. Table 1 provides a summary of different strategies for structural batteries and their performance achievements.

#### Individual CFs as micro-battery cells

The principle of an individual CF as a load-bearing substrate with a thin-film battery coating was first introduced in 2001 and referred to as PowerFibers [83]. The separate layers of battery components, shown in Fig. 4(c), were applied with various techniques, including vacuum deposition, magnetron sputtering, and electron-beam evaporation. Multiple fibers can be embedded in an adhesive matrix and sandwiched between two poly(ethylene terephthalate) (PET) foils. In this study, the CF is electrochemically inactive and only acts as structural support.

CFs can also be used as the active electrode material to manufacture a structural battery by coating the surface with polymer electrolytes. A 470 nm thick Li-triflate-containing polymethacrylate coating on CF tows was demonstrated with the electrocoating technique [86]. The coated CF tow was assembled with a glass microfiber separator and Li metal to demonstrate the electrochemical performance. In another study, UV-assisted coextrusion deposition of CFs and doped functional photopolymer resin were utilized to print free-standing CF-reinforced composite structures that were used as fully functional structural LIBs, where each individual fiber acts as a micro-battery cell [84] (Fig. 4(d)).

#### Structural separators, binders, and lamination designs

Similar to active materials and current collectors, conventional separators and binders in SOA batteries are also not designed to have outstanding mechanical properties. Therefore, their mechanical properties need to be significantly enhanced by rational designs in multifunctional energy storage without remarkably compromising electrochemical performance, so there is a gain in their multifunctional efficiencies. In this section, representative works on structural separators and binders are summarized together with their valuable inspirations, and key factors for a successful design are discussed. To avoid redundancy, the separators covered in this section are limited to non-ion-conducting membranes. Solid-state electrolytes (SSEs), which are also regarded as separators sometimes, will be discussed separately in section *Solid-state electrolytes (SSE)*.

Aramid nanofibers (ANFs) have been widely used to strengthen membranes in various devices since Ming et al. developed a direct and controllable method to synthesize ultrastrong ANFs in 2011 [90–97]. In LIBs, ANF-based separators were first proposed to suppress lithium dendrite growth thanks to their excellent mechanical properties [98]. Young's moduli of 1–10 GPa were realized via layer-by-layer dip or spin-coating method, and the batteries with such ANF-based separators all

showed similar or better cycling performance compared to conventional ones [99,100].

Patel and coworkers first suggested that ANF-based separators are also promising component candidates in structural batteries. To avoid drawbacks in previous ANF-based separator studies such as intensive and complicated synthesis, they adopted a straightforward vacuum filtration technique to prepare ANF separators without modification or additives, as shown in Fig. 5(a) [88]. The as-prepared separator had a high Young's modulus of 8.8 GPa (Fig. 5(b)), but it significantly decreased the battery's capacity and cycling stability due to its small pore size (0.8 nm on average) which impedes ion transport (Fig. 5(c)). Later, Wang et al. modified the process to enlarge the pores in ANF separators to 20–60 nm and the separator's Young's modulus remained at 2.8 GPa. The Li/LiCoO<sub>2</sub> and Na/Na<sub>2</sub>Ti<sub>3</sub>O<sub>7</sub> cells with such ANF separators overperform same cells with Celgard 2400 separators in cycling [101].

Besides separators, new binders have also been designed to enhance the mechanical properties of structural batteries. For example, various binders have been developed to better adhere electrode particles together and to current collectors in past decades [102,103]. Although not intentionally designed for structural batteries, some of them showed potential applications in structural energy storage. For example, Kovalenko and coworkers developed a sodium alginate binder with a Young's modulus of ca. 4.3 GPa [104], and Dang et al. proposed lithium substituted poly(acrylic acid) (PAA) as a binder whose modulus is 10–22 GPa [105]. Together with other robust binders like gelatin and konjac glucomannan [106–110], those materials can be employed to enhance the modulus and strength of electrodes. Also, some binders such as polyacrylonitrile (PAN) and sodium alginate can endow electrode composites with higher adhesion strength to current collectors compared to conventional binders [23,25,111], which is also vital for electrodes to stay intact and thus electrochemically functional under external loads.

Besides adopting ANF as a separator material, the Lutkenhaus group proposed that ANF could also be a good binder candidate for structural electrodes. In 2017, they designed a reduced graphene oxide (rGO)/ANF composite supercapacitor electrode with a tensile modulus of 13 GPa [112]. The achieved robustness was ascribed to the high intrinsic mechanical properties of graphene and ANF individually, and their extensive hydrogen bonding and  $\pi$ - $\pi$  interaction which enabled effective load transfer. Later, they found that chemical functionalization on rGO with carboxyl (–COOH) and amine (–NH<sub>2</sub>) groups or tannic acid could further enhance the bonding and consequently improve the electrode modulus to up to 25 GPa [113,114]. Based on this progress, the Lutkenhaus group demonstrated that via vacuum filtration of a DMSO solution of GO, branched ANF (BANF), and active materials such as Si or LFP and further reduction of GO, strong electrodes with moduli of 5–7 GPa could be realized (Fig. 5(d)) [89]. The as-prepared rGO/80 wt% LFP/5 wt% BANF electrode could deliver a specific capacity of 158 mAh g<sup>-1</sup> at 0.3C (Fig. 5(e)) and had a capacity retention of 66% after 200 cycles at 0.6C (Fig. 5(f)). The authors believed that the achieved high mechanical properties and cycling stability were attributed to the BANF that acted as a binder to adhere active materials to strong rGO substrates, which also allowed higher active material

TABLE 1

Summary of strategies for structural batteries and performance achievements. Young's modulus (E), ultimate tensile strength (UTS), flexural modulus ( $E_f$ ), flexural strength ( $\sigma_f$ ), flexural rigidity (D). Specific energy was calculated based on active materials (active) or all cell components (cell).

Strategy	Reinforcement type	Cell type	Specific energy (Wh kg <sup>-1</sup> )	Mechanical performance	Cycle life	Year	Reference
Embedding 5 LIBs between foam structure in CF laminate sandwich	external CF	LiPo pouch cell	ca. 94.4* (cell)	E = 9.1 GPa, UTS = 95.5 MPa	not tested	2020	[28]
LIB sheets with interlocking rivets between CF laminate sandwich	external CF, interlocking rivets	Graphite/NMC (LIB)	131 (active)	D = 12.1 N m <sup>2</sup>	80% capacity retention after 800 cycles	2019	[46]
CF anode and current collector, structural electrolyte, GF separator	internal CF, GF, electrolyte	CF/LFP (LIB)	23.6 (cell)	E = 25 GPa, UTS = 300 MPa	High capacity retention for 60 cycles	2021	[14]
Graphite@CF anode, LFP@CF cathode between CF laminate sandwich	internal CF, external CF	Graphite/LFP (LIB)	106 (active) 37 (cell)	E = 1.8 GPa, UTS = 213 MPa	High capacity retention for 50 cycles	2020	[71]
Embedding LIBs between foam structure in CF laminate sandwich	external CF	Li pouch cell	ca. 45 (cell)	D = 985 N m <sup>2</sup>	not tested	2012	[41]
Embedding LIBs transversal stacking between CF laminate sandwich	external CF, internal stacking	MCMB/NMC (LIB)	102 (cell)	D = 781 N m <sup>2</sup>	86% capacity after 190 cycles	2017	[52]
Embedding 2 LIBs between foam in CF laminate sandwich	external CF	LiPo pouch cell	ca. 72* (cell)	$E_f = 11.6$ GPa, $\sigma_f = 101$ MPa	not tested	2020	[47]
LFP@CF cathode, CF anode	internal CF	CF/LFP (LIB)	222 (active)	not tested	88% capacity retention after 300 cycles	2021	[73]
PS@CF cathode, Li@CF anode	internal CF	Li/PS (LIB)	43 (cell)	Electrode: E = 9.2 GPa, separator: E = 1.2 GPa	81% capacity retention after 200 cycles	2020	[78]
MnO <sub>2</sub> @CF cathode, Zn@CF anode	internal CF, external CF	Zn/MnO <sub>2</sub> (ZIB)	182 (active)	E = 12.5 GPa, UTS = 293 MPa, $E_f = 4.4$ GPa, $\sigma_f = 181$ MPa	88% capacity retention after 100 cycles	2021	[80]
MnO <sub>2</sub> @CF cathode, Zn sheet	internal CF, GF, external CF, GF	Zn/MnO <sub>2</sub> (ZIB)	205 (active)	E = 3 GPa, UTS = 180 MPa, $E_f = 12.9$ GPa, $\sigma_f = 230$ MPa	85% capacity retention after 300 cycles	2022	[81]
NH <sub>4</sub> V <sub>4</sub> O <sub>10</sub> @CF cathode, GF separator, SPE/kaolin, CF laminate sandwich	internal CF, GF, SPE, external CF	Zn/NH <sub>4</sub> V <sub>4</sub> O <sub>10</sub> (ZIB)	159 (active)	E = 5.7 GPa, UTS = 166 MPa, $E_f = 52.4$ GPa, $\sigma_f = 584$ MPa	95% capacity retention after 500 cycles	2022	[87]

(\*The value was calculated from the reported volumetric energy density and mass density).



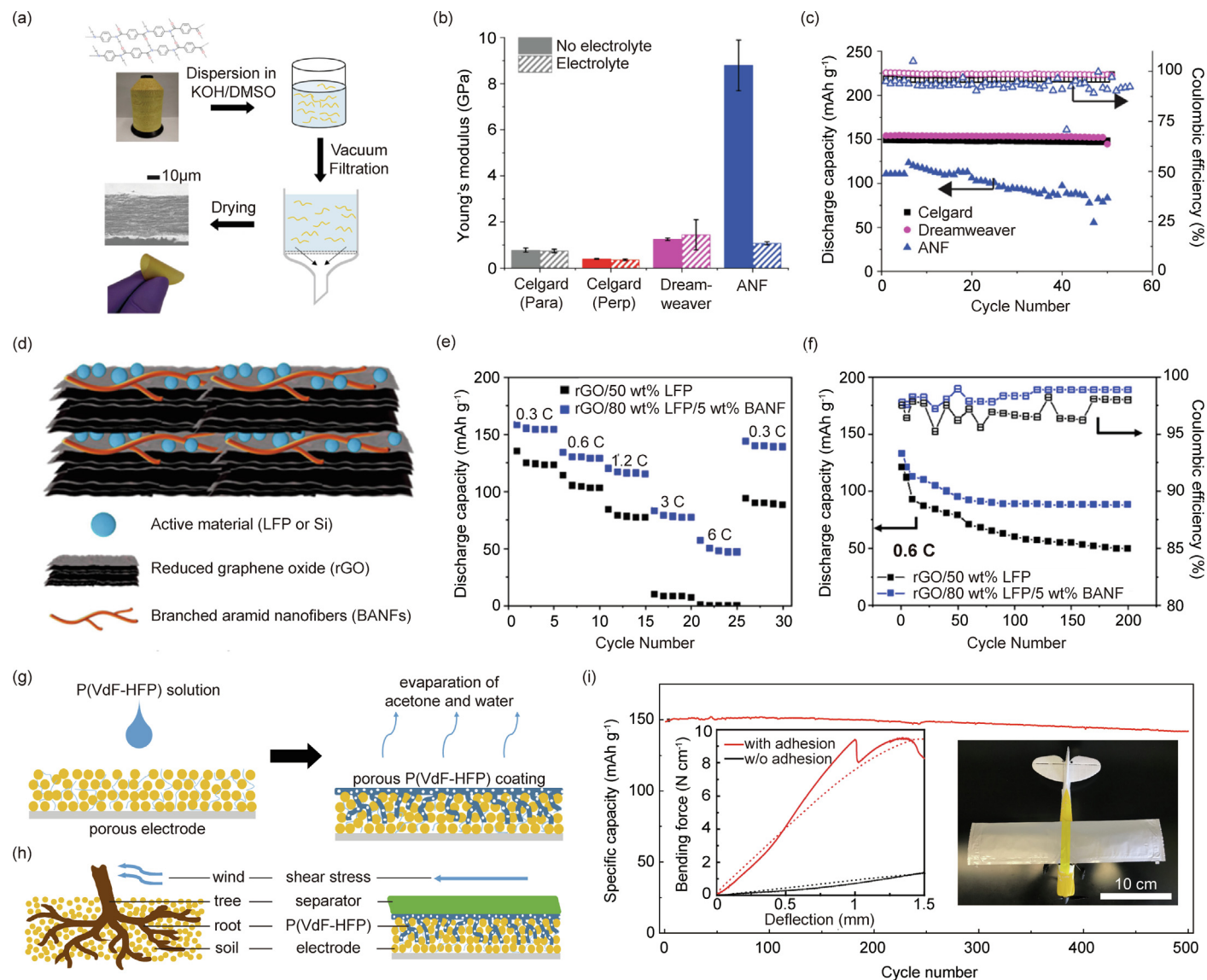


FIGURE 5

(a) Schematic representing the formation of ANF separators. (b) Young's modulus for Celgard (parallel and perpendicular directions), Dreamweaver, and ANF separators when dry and wet. (c) Cycle stability of batteries assembled with ANF, Celgard, and Dreamweaver separators. Reproduced with permission [88]. Copyright © 2020 American Chemical Society. (d) Schematic representation for rGO/BANF lithium-ion battery electrodes. (e) Galvanostatic cycling at different C-rates (0.3–6C), and (f) prolonged galvanostatic cycling at 0.6C for 200 cycles for rGO/50 wt % LFP and rGO/80 wt % LFP/5 wt % BANF in a lithium metal half-cell. Reproduced with permission [89]. Copyright © 2021, American Chemical Society. (g) A schematic of the fabrication process of an electrode with tree-root-like structure, followed by hot pressing to adhere to separators. (h) The analogy between a tree against a strong wind (left) and the electrode/separator adhesion against the shear stress introduced by bending (right). (i) Cycling performance of graphite/NMC532 full cells with electrode/separator adhesion. Left inset: Experimental bending force per width–deflection curves of 2.1 mm thick graphite/NMC pouch cells with and without electrode/separator adhesion (solid lines) and corresponding FE simulation results (dotted lines). Right inset: Top view of an airplane model with laminated pouch cells as “electric wings.” One wing has a capacity of 780 mAh, and the video of steady flying of the plane can be found in the original paper. Reproduced with permission [25]. Copyright © 2021 Wiley-VCH GmbH.

loadings than CF-based electrodes. Those works indicate that clever choices on binders can also create new strong substrates other than CFs to realize structural electrodes.

All those developments on structural electrodes, separators, and binders indicate that higher mechanical properties of batteries can be achieved by enhancing individual layer components. Furthermore, Jin and coworkers pointed out the significance of the interaction between each part in structural batteries to maximize the contribution of individual components, especially for high flexural and shear stiffness. In analogy to the case that bind-

ing pages together may make a thick book more rigid than making each thin page strong, they developed a bio-inspired tree-root-like poly(vinylidene fluoride-co-hexafluoropropylene) (PVdF-HFP) binder to adhere separator and porous electrode layers tightly via scalable coating and hot-pressing, as shown in Fig. 5(g-h) [25]. The interfacial PVdF-HFP binders were highly porous for electrolyte permeation and could enhance the flexural modulus of a graphite/NMC532 battery by 11 times to 3.1 GPa (Fig. 5(i) left inset) as all layers bend as a whole rather than separately. Such mechanically enhanced batteries with high flexural

modulus were further demonstrated to serve as both the only power source and wings of a UAV model (Fig. 5(i) right inset). The introduced interfacial binder layer only increased battery weight by ca. 3% and the corresponding battery maintained a high capacity retention of 95.5% after 500 cycles (Fig. 5(i)). Later, Ransil et al. reported a sodium silicate waterglass binder with a Young's modulus > 50 GPa based on nanoindentation in liquid electrolytes to adhere electrode and separator layers, and then proposed a graphite/LFP structural battery with a specific energy of 94 Wh kg<sup>-1</sup>, which maintained at 80 Wh kg<sup>-1</sup> after 50 cycles [115].

All the inspiring works discussed above, either bringing up new materials or new concepts, not only reflect the importance of structural separators and binders, but also provide valuable takeaway points to future researchers. One is that successful structural separators and binders should not lose mechanical strength when exposed to electrolytes, and not block ion transport severely during cycling. For example, Patel et al. observed that the modulus of their ANF separators would decrease by 8 times in liquid electrolytes and became comparable to commercial ones [88]. Secondly, batteries using structural components can be further strengthened by laminating due to better load transfer among components under shear or flexural loads, which highlights the significance of solid-state electrolytes for multifunctional energy storage, but the electrochemical properties such as ionic conductivity should not be sacrificed significantly in order to have a multifunctional efficiency of > 1.

#### *Solid-state electrolytes (SSE)*

Solid-state batteries use solid electrolytes, such as polymers, ceramics, and their composites, instead of conventional liquid electrolytes. SSEs are attractive for batteries since they have better thermal stability and can improve safety by replacing flammable liquid components [116–118]. In terms of electrochemical properties, their ionic conductivities are typically 10<sup>-6</sup>–10<sup>-3</sup> S cm<sup>-1</sup> in solid polymer electrolytes and 10<sup>-4</sup>–10<sup>-2</sup> S cm<sup>-1</sup> in ceramic electrolytes. For structural batteries, the solid nature indicates that they can enhance not only the tensile and compressive properties of a battery, but also load-transfer between different layers and thus improve flexural properties. In this section, we will discuss the merits and challenges of different types of solid electrolytes and solid-state batteries for the application in structural batteries and recent progress. We will focus on mechanical properties but less on electrochemical characteristics, which have been well discussed in a number of other reviews [119–122].

#### *Structural batteries with solid polymer electrolytes*

A solid polymer electrolyte (SPE) is a mixture of polymers and salts. The polar groups in a polymer can dissociate salts and allow ions to move in an electrical field. Additives, such as ceramic fillers, liquids, or solid plasticizers are often added to enhance ionic conductivity. Poly(ethylene oxide) (PEO), poly(acrylonitrile) (PAN), poly(vinyl alcohol) (PVA), poly(vinylidene fluoride) (PVdF) or polycarbonate (PC) are common polymer matrix materials among others, with PEO being the most widely used [119,122].

The intrinsic moduli of these polymers range from 290–330 MPa for PEO [123] up to 2.6 GPa for PC/PVdF [124,125] which can incrementally enhance the structural properties of a

battery. The addition of salts and plasticizers can greatly enhance the relative mobility of polymer chains, but reduce mechanical properties, such as modulus and strength, significantly. Such correlation has been studied in the pioneering work by Snyder et al., investigating the correlation between mechanical properties and ion conductivity for structural electrolytes [126,127]. It was found that the addition of salt to the polymer increased the T<sub>g</sub> and reached the highest ion conductivity between 9–12 wt% in PEG [126]. Further, it was observed that co-polymerization of different monomers exhibited improved electrochemical-mechanical multifunctionality compared to homopolymers [127]. For example, common polymer electrolytes with reasonable conductivities often have elastic moduli < 100 MPa. Moreover, gel polymers are sometimes referred to as solid electrolytes and can exhibit very high ionic conductivities and good flexibility, but usually their modulus is in the range of several 100 kPa [128–131]. Therefore, to take advantage of polymer electrolytes in structural batteries, it is important to further enhance their mechanical properties such as using a mechanically strong backbone network.

On the other side, polymer electrolytes can also bind different components in a battery together, which enhances load transfer between different layers and thus improve the flexural modulus and strength [25]. The enhancement in flexural properties is affected by both the intrinsic mechanical properties of the polymer electrolyte phase and the interfacial adhesion between the polymer electrolyte and other battery components.

In recent years, various groups have explored polymer electrolytes to enhance the mechanical performance of structural batteries. Polymer electrolytes can be directly deposited onto CFs which act as structurally strong substrates for structural batteries [86,132,133]. For the fabrication of lightweight primary load-bearing structural composites with CF reinforcement, epoxy adhesives are extensively used due to their unique characteristics compared to other polymers [134], which motivates the development of epoxy-based electrolytes for structural batteries. A common approach is to create a porous epoxy matrix, penetrated with liquid electrolytes as an ion-conducting phase, leading to sufficient ion conductivity, while maintaining good mechanical performance [135–138]. These bi-continuous multifunctional electrolytes, sometimes referred to as structural battery electrolytes (SBEs) [139,140], can be used to manufacture CF-reinforced structural batteries with high tensile modulus (25–50 GPa) and good cycling performance [14,141]. Willgert et al. investigated UV-polymerized acrylate-based electrolytes with high storage moduli [142–145], however, it should be noted that for the practical fabrication of structural batteries a thermal polymerization is needed [140].

#### *Structural batteries with ceramic/inorganic electrolytes*

In contrast to SPEs, ceramic electrolytes exhibit 2–3 orders of magnitude higher intrinsic ion conductivity (e.g. LLZO: 10<sup>-4</sup>–10<sup>-3</sup> S cm<sup>-1</sup>, sulfides: 10<sup>-3</sup>–10<sup>-2</sup> S cm<sup>-1</sup>) [146] and their moduli are also much higher (e.g. 115–200 GPa for oxides [147,148] and 15–37 GPa for sulfides [149,150]). Therefore, if liquid electrolytes can be completely replaced by ceramic electrolytes and the interface between ceramic electrolytes and electrodes/current

collectors is coherent, the modulus of a battery can be enhanced dramatically.

Unfortunately, such a coherent interface is very difficult to realize. Sintering and applying high pressure have been explored to enhance interfacial adhesion and coherence, but mechanical properties have not been extensively studied. On the other side, oxide ceramics are well known to be fragile. This means that the toughness and strength of oxide ceramic-based solid electrolytes may be compromised. Their lack of flexibility, sometimes referred to as the “mismatch problem”, can lead to high interfacial electrode/electrolyte resistance, and various defects and failure mechanisms (e.g. microcracking) may occur during cycling caused by internal stresses [151]. Given this issue, sulfide electrolytes may be more attractive as they are softer and have more ductility compared to oxides. These potential issues need to be carefully examined when constructing a ceramic electrolyte-based structural battery.

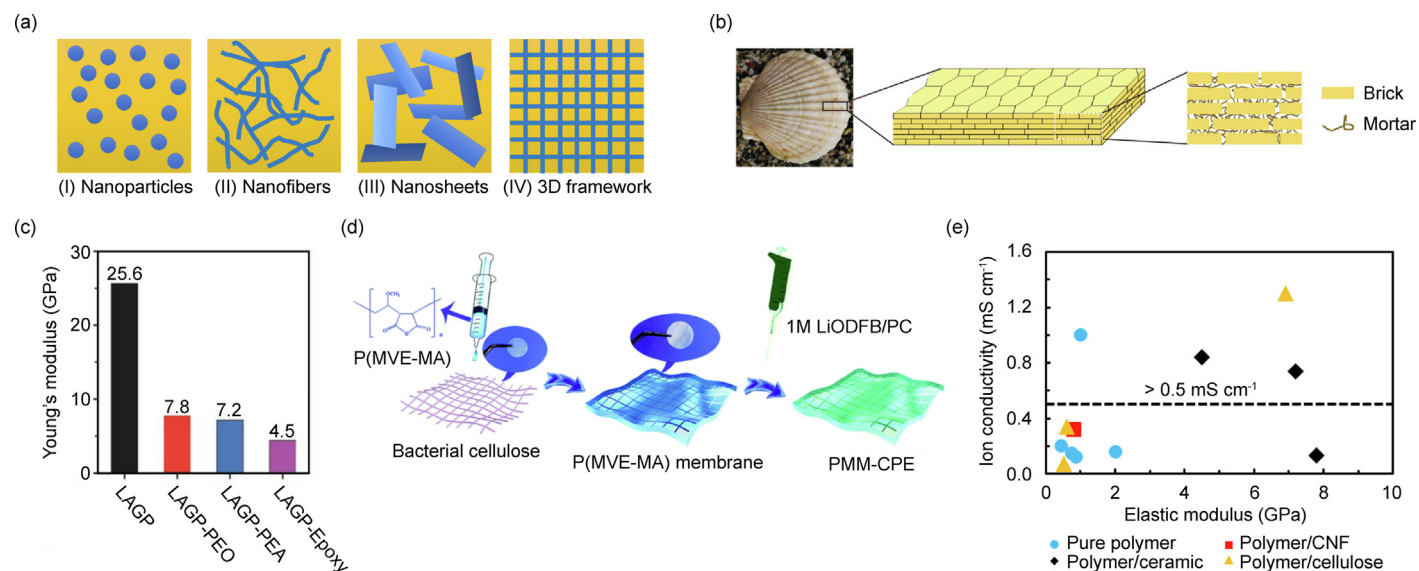
Structural batteries with integrated thin-film lithium batteries (TFLBs) containing ceramic electrolytes have been demonstrated [49,50]. However, such a design does not take advantage of the high mechanical properties of the ceramic phase. In general, oxide ceramic electrolytes demonstrate good performance under compression/tension but can fail at relatively low bending stress due to their brittle characteristics [49,54,152]. For a better understanding of oxide/sulfide electrolytes for structural batteries, more studies on mechanical characterizations at the microscale (e.g. nano/micro-indentation) and macroscale (e.g. tensile and bending tests) are necessary.

#### Structural batteries with composite electrolytes

Composite electrolytes, which integrate ceramics into polymer electrolytes, may combine their advantages, such as the high modulus and ion conductivity of ceramics with the high tough-

ness and flexibility of polymers. Nanocomposite polymer electrolytes (NCPE) with ionically conductive ceramic nanoparticles embedded in a polymer matrix can reach ion conductivities up to  $10^{-4}$ – $10^{-3}$  S  $\text{cm}^{-1}$  [153–155]. Detailed information and emerging trends can be found in recent reviews of polymer-based composite electrolytes [120,121,156–158]. In such composite electrolytes, the geometry and topology of the two phases are critical to both ion conductivity and mechanical properties. In general, nanofibers/nanowires lead to improved ion conductivity due to more connected pathways than isolated spherical nanoparticles. Similarly, the mechanical properties are typically enhanced by different shapes in the following order: nanoparticle < nanowire < nanosheet < 3D framework [159] (Fig. 6(a)).

Good interfacial adhesion is an essential requirement for the transfer of mechanical load and electrochemical performance. This was demonstrated in a nacre-inspired design of a ceramic/polymer solid composite electrolytes with a “brick-and-mortar” microstructure (Fig. 6(b)). The improved adhesion strength leads to an elastic modulus of up to 7.8 GPa (Fig. 6(c)) [160]. Zekoll et al. used a 3D-printed template to create an epoxy-infused LAGP structural framework with different designs [161]. The gyroid LAGP-epoxy electrolyte had an ion conductivity of  $1.6 \times 10^{-4}$  S  $\text{cm}^{-1}$  and a high modulus of 44 GPa in the micro-compression test with a sample size of ca. 1 mm  $\times$  1 mm  $\times$  2 mm. Another study demonstrated a solid poly(propylene carbonate) (PPC)/LLZTO composite electrolyte with a tensile strength of 6.8 MPa and  $5.2 \times 10^{-4}$  S  $\text{cm}^{-1}$  with good electrochemical performance and stability [162]. Implementing a macroscopic fibrous structural network to a polymer matrix was demonstrated with bacterial cellulose-supported poly(methyl vinyl ether-alt-maleic anhydride), P(MVE-MA), resulting in a high modulus of 6.9 GPa. After soaking in a liquid



**FIGURE 6**

(a) Different shapes of particles/structures for the improvement of ion conductivity and mechanical properties of SSEs. (b) The design of a nacre-inspired “brick-and-mortar” microstructure with (c) improved Young’s modulus. Reproduced with permission [160]. Copyright © 2020 Wiley - VCH GmbH. (d) A Scheme of the fabrication steps of a cellulose-reinforced polymer electrolyte membrane soaked with liquid electrolyte. Reproduced with permission [163]. Copyright © 2018 Royal Society of Chemistry. (e) Ion conductivity vs elastic modulus plot of different solid-state electrolytes reported in the literature from Table 2. The ion conductivity limit of  $0.5 \text{ mS cm}^{-1}$  is marked in the diagram as a threshold for good electrochemical performance.



electrolyte, the ion conductivity reached  $1.3 \times 10^{-3} \text{ S cm}^{-1}$  (Fig. 6(d)) [163]. A high strength of 29 MPa with a tensile modulus of about 600 MPa was achieved with a Cu-coordinated cellulose-based electrolyte, and the ion conductivity was  $1.5 \times 10^{-3} \text{ S cm}^{-1}$  along the molecular chain direction ( $3.4 \times 10^{-4} \text{ S cm}^{-1}$  through-plane) after soaking in liquid electrolyte [164].

A summary of representative solid-state electrolytes with their elastic moduli and ion conductivities is presented in Fig. 6(e) and Table 2. We only chose SPEs with moduli higher than 100 MPa. Accordingly, an elastic modulus of a few GPa with reasonably high ion conductivity of  $> 0.5 \text{ mS cm}^{-1}$ , which is a desirable combination of properties for electrolytes in structural batteries, was achieved with composite electrolytes consisting of polymers with ceramic plates [160] or with a cellulose fiber-support [163].

### Simulations

As summarized above, the number of reports on experimental progress in structural energy storage have increased fast since 2000. Those works comprehensively cover designs of both individual components and integrated systems, which enhance the mechanical and electrochemical properties of batteries and advance fundamental knowledge of structural batteries. On the other side, simulation works are not comparably thriving to provide theoretical insights into experimental designs where strong coupling of electrochemistry and mechanics across multiple length scales needs to be considered. On the other side, such complex coupling is also a key issue hindering the simulation progress. In this section, progress on the simulation of structural batteries is summarized, together with their inspirations for structural energy storage designs and future simulation modeling.

Most of the simulation works focus on the behaviors of CF-based batteries due to CFs' excellent mechanical properties and capability of serving as anodes or current collectors in structural batteries. Dionisi et al. presented a method to track the global deformations and interlaminar stresses in a multi-layer CF anode/separator/CF reinforced cathode structural battery laminate immersed in a structural battery electrolyte (SBE) [65]. Their results indicated that the volume changes of the CF anodes might cause interfacial delamination in batteries, and smart designs should be adopted to suppress this effect. To better understand the processes occurring, Xu and coworkers developed

physics-based electrochemical models consisting of micro-battery units where electrolyte-coated CFs embedded in a cathode particle-doped polymer matrix (Fig. 7(a)) [167,170]. They found that both the non-uniform Li concentration distribution in the CFs and the mechanical interaction between the CFs and the matrix governed the stress within the cell, which would cause CF cracks and matrix failure. As shown in Fig. 7(b), Li-ion inhomogeneity in CFs during 1C cycling together with a stiff matrix would lead to a high and nonuniform internal stress, which will become more severe at 10C (Fig. 7(c)). Therefore, tuning current densities and matrix properties could optimize the battery design. They further demonstrated that a cross-ply lay-up structural battery would have different debonding behavior from a UD laminate battery, which still required experimental evidence [171].

In the following studies, Carlstedt and Asp emphasized that thermal effects should also be considered in simulation. Their finite element (FE) models clearly disclosed that the internal stresses were the results of both heat generation and volume changes of CFs during cycling, and current densities and lamina dimensions mainly determined the level of heat generation [172]. Together with other coworkers, they later pointed out that the voltage profile of a structural battery would be different when CFs and electrolytes are under non-negligible mechanical loads (Fig. 7(d)). For example, Fig. 7(e) showed that a tensile strain of 0.01 along the fiber direction can lead to a 5 mV difference in the potential of CF electrode during lithiation, suggesting that electro-chemo-mechanical coupling played a pronounced role and it was important for the simulation of structural batteries [168]. The group also numerically demonstrated that electrode thicknesses, transport properties of SBEs, and current densities would appreciably affect the cycling performance of CF-based batteries [173], and that transverse elastic properties  $E_2$  and  $G_{23}$  and in-plane shear modulus  $G_{12}$  of a battery are strongly affected by the state of charge while the longitudinal stiffness  $E_1$  is not [174]. All those works indicate that coupling different physical and chemical processes is vital in simulation in order to better analyze the performance and failure of structural energy storage devices.

Simulation is also a powerful tool to better understand the behaviors of components such as CFs and SBEs themselves in

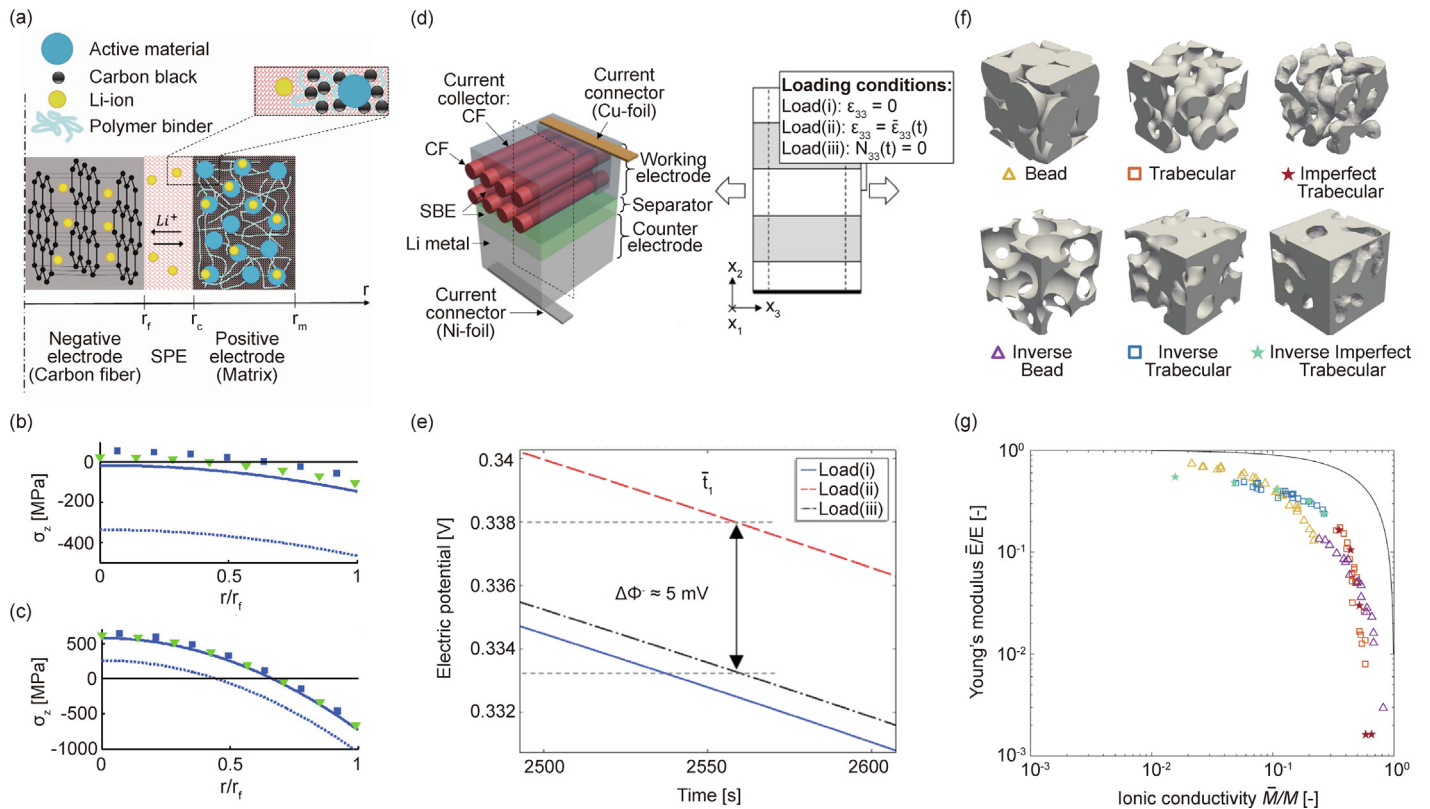
TABLE 2

Different types of solid-state electrolytes (SSEs) with their reported elastic moduli and ion conductivities which are around  $0.1 \text{ mS cm}^{-1}$  or above.

Type	Elastic modulus (MPa)	Ion conductivity ( $\text{mS cm}^{-1}$ )	Year	Reference
Pure polymer	ca. 1000	ca. 1	2014	[165]
	750	0.15	2017	[139]
	2000	0.16	2017	[166]
	ca. 540	ca. 0.2	2019	[140]
	865	0.12	2022	[138]
Polymer/ceramic	7800	0.13	2020	[160]
	7200	0.74	2020	[160]
	4500	0.84	2020	[160]
Polymer cellulose	520	0.07	2014	[144]
	6900	1.3	2018	[163]
	ca. 600	0.34 (1.5*)	2021	[164]
Polymer/CNF	805	0.32	2021	[135]

(\*Ion conductivity in fiber direction).



**FIGURE 7**

(a) Schematics of a structural battery model for (b) and (c). (b-c) Stress distribution along fiber radial direction at fully charged state with (b) 1C and (c) 10C, where the blue solid line corresponds to a model with Young's modulus of matrix  $E_m = 3$  GPa and expansion coefficient of matrix  $\beta_m = 0$ , blue dotted line indicates  $E_m = 3$  GPa and  $\beta_m = 0.05$ , blue square dots indicates  $E_m = 0.3$  GPa and  $\beta_m = 0$ , and green triangle dots indicates  $E_m = 0.3$  GPa and  $\beta_m = 0.05$ . Reproduced with permission [167]. Copyright © 2018 SAGE Publications. (d) Schematics of a structural battery model for (e). For Load (i) and (ii), the plane strain  $\epsilon_{33}$  is respectively set to 0 and 0.01. For load (iii), the normal force  $N_{33}$  is set to be 0. (e) Effects of external loads on the voltage profiles during the discharge process. Reproduced with permission [168]. Copyright © 2020 IOP Publishing. (f) Solid polymer matrix of various artificially generated microstructures. (g) Trade-off between stiffness and ionic conductivity. Reproduced with permission [169]. Copyright © 2020 IOP Publishing.

batteries. Pupurs and Varna built an FE model to track the origin of CF damage [175]. This model successfully revealed that delithiation might lead to radial cracks in a fiber, which might deflect into arc cracks during the subsequent lithiation cycles, together with details about the crack propagations which will not be all summarized here due to space limitations. Carlstedt and coworkers numerically studied the SBEs which consist of strong solid polymer skeletons and conductive liquid electrolytes, and the results showed that convection within the SBEs would only significantly influence the battery performance when external loads caused severe deformation of SBEs or large current pulses were applied [176]. To better understand the designing rules of SBEs, Tu et al. first summarized and studied the structures of eight reported solid polymer matrices, and consequently proposed six general topologies (Fig. 7(f)). By artificially generating a series of SBEs with different porosities and solid skeleton structures, they found a trade-off relation between Young's modulus and ionic conductivity for SBEs and different topologies behave distinctly (Fig. 7(g)) [169]. Though more characteristics need to be coupled to understand the components more thoroughly, computation is an appealing method due to its more inexpensive cost and higher freedom than experiments.

Simulation has also been adopted to evaluate the performance of structural batteries at the system level. In 2010, Roberts and Aglietti experimentally designed a multifunctional panel used in spacecraft by incorporating a polymer lithium-ion battery into structural sandwich panels, and then built an FE model to analyze experimental results to further guide experimental designs [177]. The model disclosed that optimally redistributing the battery mass, reducing the parasitic mass and the acceleration rate could further suppress the deformation of the multifunctional panels during launch. Capovilla and coworkers later developed a structural battery as an external face of a 1U CubeSat, and also conducted FE analysis to prove the stability of the proposed batteries under launch and find optimizing methods [178]. Carlstedt and Asp developed a performance analysis framework to study the benefits of using structural battery composites in EVs [17]. Their case study manifested that the driving range could be increased by 70% for lightweight vehicles with feasible structural battery designs. In addition, the performance analysis showed that separator thicknesses, electrolyte conductivities, and current collector designs are critical to the electrical performance. Kambampati et al. performed a topology optimization of the structural battery packs in aircraft wings, which was numerically

proven to be able to reduce the battery temperature via effective heat transfer while sustaining a given load [179]. Those system-level simulation works also imply that structural energy storage devices need to have different properties and targeted designs to satisfy specific applications.

Besides, it is worth mentioning that there are lots of simulation works on conventional lithium batteries, covering the mechanical and electrochemical behaviors of individual components and whole battery packs [180–186]. Though not systematically summarized here, those works can be of great benefit to the field of structural energy storage to better understand how a component or a device responds to a certain stimulation such as current or mechanical impact, and thus to better design devices with higher performance and safety. In the future, developing models with a more comprehensive coupling of multiple characteristics across multiple scales is the key to advance simulations for guiding experimental designs. In addition, more experimental data is also in demand for building such advanced models.

### Challenges and outlooks

As summarized above, significant progress has been made in the field of structural batteries in past years, but there is still a lot to be further improved. To implement structural batteries in systems such as vehicles, several key points must be satisfied first, including mechanical and electrochemical performance, safety, and costs, as summarized in Fig. 8. In this section, these points will be briefly discussed, covering current challenges and future development directions.

#### *Fundamental knowledge of electro-chemo-mechanical coupling*

Electro-chemo-mechanical coupling is a major topic in batteries where stress/strain and electrochemical processes in components interact with each other, especially for studies of decay and failure mechanisms, and it is more critical in structural batteries. Therefore, understanding such coupling is fundamental to addressing potential issues in structural batteries which need to bear appreciable loads during cycling. For example, as mentioned above, SOC will change the mechanical properties of CFs, and external loads may also shift the potential of electrodes. Diffusion of Li ions may induce a local stress, and conversely, a stress in materials can change the ionic diffusion properties. Besides, there will inevitably be propagation and aggregation of point defects and dislocations in both electrodes and electrolyte matrices during cycling, which involves rearrangement of atoms and consequent performance decays, and such processes will be influenced by the applied loads thermodynamically and kinetically. Volume changes of electrodes during cycling may cause debonding of electrode/electrolyte interfaces, inducing extra impedance and more heterogeneity of reactions and stresses.

In addition to these effects, there can be cascading cracks in various parts of batteries which exacerbate the performance degeneration. Stress accumulation and inhomogeneity can incur cracks in SEI or even electrodes, which are usually accompanied by more electrolyte decomposition and capacity decay. For more details about the electro-chemo-mechanical coupling in batteries, there are some comprehensive reviews where readers can find lots of valuable takeaways, including the experimental and

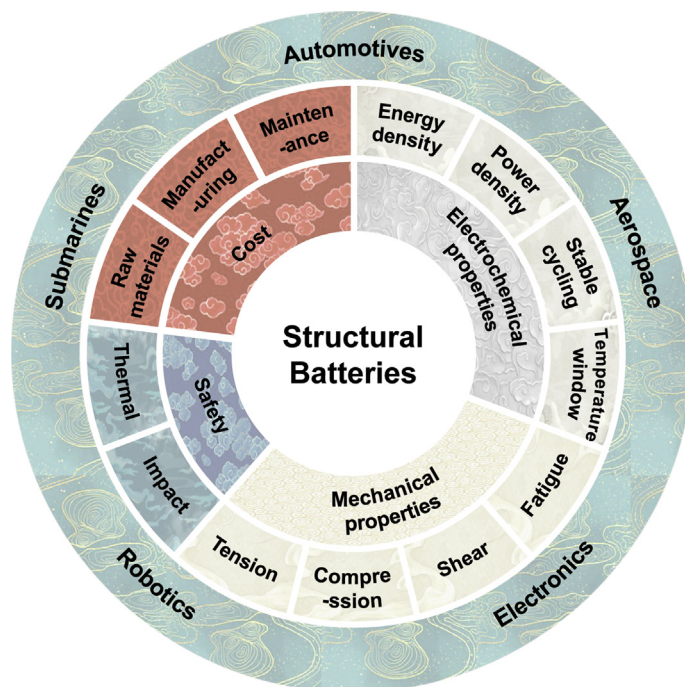


FIGURE 8

Key parameters and challenges for structural batteries toward practical applications.

numerical methods to study the coupling, and tactics to suppress its detrimental effects [187–189].

#### *Mechanical/electrochemical performance in real systems*

To achieve satisfactory multifunctional efficiencies and gain savings in mass, structural energy storage devices should always have good mechanical and electrochemical properties simultaneously. As discussed in Section *Fundamentals of structural energy storage devices*, the criteria for those two properties should be set based on the target application. Mechanically, a key issue is that structural batteries usually need to have high strengths in multiple directions. But even with CFs, conventional batteries with a rectangular shape may only satisfy requirements on tensile properties rather than shear and bending ones, so other strategies such as binders and solid-state electrolytes should be adopted [25]. Electrochemically, besides energy density, structural batteries usually need to have a cycle life of hundreds and even thousands of cycles, and reasonable power densities. Currently, CF electrodes can barely cycle for tens of cycles, thus further modifications are needed to improve their cycling performance [190]. Similarly, an electrolyte or a binder proposed should be electrochemically stable to meet these basic requirements.

Currently, most structural battery studies are still in the early stage of concept demonstrations, and other passive components in real systems are rarely involved such as battery management systems and cooling systems. They may considerably reduce the mass gains from structural batteries especially if the structural battery designs are complicated. Hence those parasitic elements in real systems must be taken into consideration when commercialization is considered.

### Long-term performance stability during serving

Since structural energy storage devices usually work in harsher conditions than conventional batteries, the stability of their performance under mechanical loads and during long-term serving are critical for practical applications. For example, continuous external loads and consequent deformations may lead to misalignment and cracks in battery components. Moreover, delamination between electrode particles and other components such as current collectors, binders, and polymer electrolytes will be more prone to happen in structural batteries since they suffer more loads than conventional ones, which can significantly deteriorate their electrochemical and mechanical performance. Practical designs of structural batteries must consider these challenges.

On the other side, electrochemical cycling can also degrade mechanical performance. The volume change of electrodes during charge and discharge also negatively affects the interfaces between electrode particles and other components [191,192], and results in worse mechanical performance, especially flexural properties. Similarly, the evolution of solid electrolyte interphase (SEI) may also affect such interfaces, which is rarely studied. Besides interfaces, repeated lithiation and delithiation will impair and even crack CFs, which may lead to mechanical failures of the whole device. Whereas, such fatigue tests are rarely included in past studies, and should be emphasized in the future.

### Safety

Safety is one of the top requirements for all batteries and it is even more critical for structural batteries since their working environment is mechanically much harsher. Therefore, safety requirements on structural batteries should be more strict than conventional batteries. Additionally, if extra protections need to be added, the protections' mass and volume must be taken into account when evaluating the multifunctional efficiency of a proposed structural battery. Hence, although not widely discussed in previous reports, safety tests should be indispensable when the field approaches practical applications, as discussed in these two studies [193,194]. To achieve a high multifunctional efficiency and enhanced safety simultaneously, researchers may consider adopting nonflammable liquid electrolytes [195–197], solid-state electrolytes [146,163,198], thermally stable separators and binders [25,78,88,199], smart designs of current collectors [200,201], and strong and lightweight external protections [202].

### Costs and maintenance

Ultimately, structural batteries should have reasonable costs, including raw materials, manufacturing, and maintenance. For example, it remains a question of how to integrate CF current collectors with active electrode materials in a scalable way. In addition, one has to take maintenance and replacement into account in practical applications. It may not be intractable if structural batteries are used as the cases of portable electronic devices, but it could be more complex to maintain or repair the structural batteries serving as airplane wings or vehicle chassis. Especially, when multiple cells are integrated into a single structural component, all cells and even the structural parts may need to be replaced even if only one cell malfunctions electrochemically or mechanically [82]. Accordingly, new battery manage-

ment systems and maintenance knowledge need to be developed due to the high coupling of electrochemistry and mechanics [194,203]. The consequence of these issues needs to be analyzed carefully in real applications. Certain applications can afford a high cost, such as aircraft and military applications, but the costs of structural batteries need to be reduced to a reasonable level for large-scale applications such as electric vehicles.

### Conclusion

Structural energy storage devices have been demonstrated experimentally and numerically to improve the mass efficiency of systems such as electric vehicles and aircraft and extend their operational duration. To promote practical applications of this concept, studies were intensively conducted to enhance the multifunctionality of batteries, including embedding SOA batteries into strong external reinforcements, developing robust electrodes and separators, and adopting binders and electrolytes to advance the load transfer efficiency among battery components. Extensive simulation works were also performed to better understand the coupling effects of electrochemistry, kinetics, and mechanics at both material and device levels, and provide valuable guidance to structural battery designs. These developments and major results are summarized in this review, together with their inspirations for future researchers and the current issues of each topic such as the mechanical and electrochemical degradation of CFs during cycling. Also, with various reported assessment methods reviewed, this review emphasizes the importance of evaluating the multifunctional efficiency of a proposed structural battery and suggests the key evaluating criteria for structural batteries by analyzing their potential applications. Finally, the remaining challenges in this field such as safety, costs, and long-term performance stability in harsh practical environments are briefly discussed with future research directions proposed. All information indicates that structural batteries are promising solutions to enhance the performance of electrified transportation, and more transformative research and progress in material and device levels are needed to accelerate their implementation in the real world.

### Data availability

No data was used for the research described in the article.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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