Multifunctional Batteries: Flexible, Transient, and Transparent

Linda A. Wehner,† Neeru Mittal,† Tian Liu,† and Markus Niederberger*  

ABSTRACT: The primary task of a battery is to store energy and to power electronic devices. This has hardly changed over the years despite all the progress made in improving their electrochemical performance. In comparison to batteries, electronic devices are continuously equipped with new functions, and they also change their physical appearance, becoming flexible, rollable, stretchable, or maybe transparent or even transient or degradable. Mechanical flexibility makes them attractive for wearable electronics or for electronic paper; transparency is desired for transparent screens or smart windows, and degradability or transient properties have the potential to reduce electronic waste. For fully integrated and self-sufficient systems, these devices have to be powered by batteries with similar physical characteristics. To make the currently used rigid and heavy batteries flexible, transparent, and degradable, the whole battery architecture including active materials, current collectors, electrolyte/separator, and packaging has to be redesigned. This requires a fundamental paradigm change in battery research, moving away from exclusively addressing the electrochemical aspects toward an interdisciplinary approach involving chemists, materials scientists, and engineers. This Outlook provides an overview of the different activities in the field of flexible, transient, and transparent batteries with a focus on the challenges that have to be faced toward the development of such multifunctional energy storage devices.

1. INTRODUCTION

Progress in portable electronic devices has been so rapid that the performance and range of features of a device just 10 years old seem awfully old-fashioned. What has not changed so much is their physical appearance. Even modern devices remain mostly solid, rigid, and fragile. However, this has begun to change, and there are prototypes and new models on the market that incorporate flexibility/bendability, transparency, or degradability/transience into the design. Clearly, such innovations also affect the battery as the main energy source. For fully integrated and self-sufficient systems, the battery as part of the device must have the same physical properties.

The rapid evolution of multipurpose (transparent, flexible, and degradable) electronic devices has the potential to revolutionize our life. Biodegradable medical implants, which serve as platforms with sensing and stimulation functions to support biological processes such as wound healing, tissue regeneration, and brain activity, are just one example of how technological innovations in electronics fit together with practical application scenarios that will profoundly influence our future lifestyles.

In contrast to flexible, transparent, and transient electronics, corresponding breakthroughs in batteries are still limited, which is certainly not due to the lack of scientific and technological interest, but due to the high complexity of the battery structure. The implementation of new physical, mechanical, and chemical properties into a multifunctional battery requires that all its components (active materials, current collectors, electrolyte/separator, and packaging) have to be flexible, transparent, and/or degradable, while maintaining the electrochemical function. From a materials science point of view, this is a great challenge because the materials currently used in lithium-ion batteries (LIBs) do not offer any of these properties. In fact, typical batteries are rigid bodies enclosed in a metal container. Even in the case of the much lighter pouch cells, the battery is protected in a rigid plastic case. Such a rigorous design is necessary to meet all the requirements for safe and long-lasting operation. Accordingly, the fabrication and assembly of a multifunctional battery require a broad expertise in materials and their processing.

In this Outlook, we want to highlight some of the latest developments in the field of flexible/stretchable, transparent,
Looking beyond electrochemical storage capability, our focus is on the challenges coming with equipping all different battery components with the additional properties. The structure follows that of the battery discussing each component from active materials, current collectors, electrolytes/separators, and packaging.

2. FLEXIBLE AND STRETCHABLE BATTERIES

The interest in flexible electronics, such as wearable devices, on-skin sensors, flexible displays, or environmental sensors, has triggered immense research activities in the field of energy storage systems that can be bent, folded, crumpled, and stretched while maintaining their electrochemical properties.\(^{12}\) However, the high standards of today’s battery technology and user expectations have set the bar very high for good electrochemical performance, affordable prices, and high safety.\(^{13,14}\)

There are two general approaches to introduce flexibility in batteries: intrinsically stiff materials are replaced by soft and bendable compounds, or stiff materials are processed into structures that are flexible.\(^{15-17}\) In both cases, for stable performance the contacts between the battery constituents have to be guaranteed under repeated deformation. Furthermore, leakage and evaporation of electrolyte have to be prevented by appropriate packaging. While these aspects are mostly given in rigid batteries with solid cases, they can represent a serious safety issue in pouch cells and soft batteries.

In the following section, we present and discuss the most relevant approaches for flexible battery components and introduce some concepts for a stretchable battery design.

**Active Materials and Electrode Structure.** Intrinsic electrochemical stability and interfacial adhesion define the robustness of a flexible battery. Using established chemistries, the remaining challenge is reduced to strengthening the adhesive forces between the battery components. Conductive additives and binders help with sandwiching the electrode layer between the self-standing, relatively strong components of the current collector and separator. Therefore, research efforts
focus on combining the performance of existing chemical systems like lithium-ion, lithium sulfur (Li/S), or zinc-ion (Zn) batteries with structures that exhibit new mechanical properties. Similar to traditional batteries, the electrode materials are usually present in the form of powders that have to be processed in such a way that a strong adhesion to the current collector is achievable. Certainly, the requirements to avoid any delamination are much stricter for a flexible or stretchable than for a rigid battery. 5,19

LIBs clearly represent the state-of-the-art in portable energy storage due to their high energy and power density as well as their long-term stability. 20 However, the current standard electrode architectures in LIBs do not allow physical bending without considerable performance loss. 21 Through careful structural engineering, flexible LIBs have been realized. Oh et al. utilized carbon-nanotube-decorated α-iron oxide particles and lithium iron phosphate as the anode and cathode, respectively. 22 Fibrous mats of these materials (Figure 1a) as electrodes as well as a gel polymer electrolyte allowed for stress-minimized bending of the electrochemical pouch cell. Their study focused on the synthesis of the electrochemically active materials and collecting the data in the bent state. For applications in flexible devices, however, one-time bending is far from practical relevance. Bendability over 5000 cycles has been introduced into well-known LIB chemistry system based on lithium cobalt oxide (LiCoO₂) and graphite through patterning, resulting in a high contact area at the interfaces, which promotes adhesion and helps the stress distribution during bending. 18

Zn-ion batteries represent another promising technology with relatively high capacity. Active materials for this type of battery are low-cost, abundant, safe, ecofriendly, and sustainable. Like LIBs, zinc-based flexible batteries consist mostly of well-established material combinations. 23,24 Zamar-ayeva et al. reported a Zn metal anode along with a manganese oxide (MnO₂) cathode, where performance retention after bending was facilitated through highly flexible binders such as poly(vinyl alcohol) (PVA) and poly(acrylic acid) (PAA). 25

A third technology intensively studied for flexible energy storage is Li/S batteries. Although the long-term stability has not yet been satisfactorily solved, the high theoretical capacity makes this system attractive. 26,27 The anode has to facilitate a uniform lithium distribution, while the cathode has to be composed of a conducting fine-pore network to accommodate the insulating sulfur. An example was presented by Chang et al. employing metal-coated carbon fibers and a N-doped carbon to collect the sulfur (Figure 1b). 28

Other battery chemistries based on metallic lithium ₂₉ or silicon ₃₀,₃₁ are still in their infancy. They struggle with quick capacity fading, even in the classical rigid design, and are therefore not yet ready for implementation in flexible devices.

Current Collector and Conductive Additive. Although copper and aluminum ₃₂ exhibit certain flexibility as thin foils, the main challenge utilizing them in a flexible battery is the poor adhesion of the electrode materials to the metal surfaces. ₃₃,₃₄ This problem can be reduced by maximizing the contact area between the two materials, for example, by using components with large surface areas or interlocked structures. ₁₈ Researchers have also tested metal nanowires ₃₅,₃₆ and electrochemically deposited metal films. ₃₇ However, the most promising alternatives are different types of carbon-based structures due to their low weight, high mechanical stability, and tunable adhesive forces. ₃₈-₄₁ Li et al. utilized carbon nanotubes and graphite as the electron collector and conductive additive (Figure 1c).²²

To ensure physical contact of the components, traditional cell manufacturing relies on binders. Their adhesive forces add to the integrity of the layer as well as to the interlayer stability. Besides the common polyvinylidene fluoride (PVDF), another approach makes use of hydrogels as binders, with which Wu et al. achieved high stability in a silicon nanoparticle anode. ₃₃ Directed deposition guided through surface architecture can further enhance the interconnection between the current collector and the active material layer. ₄₀,₄₄

Electrolyte and Separator. When selecting the electrolyte, its stable voltage window and its state of matter at the working temperature decide whether a separator or a matrix is necessary, and this also determines the permeability requirements of the packaging. Whereas the combination of liquid electrolytes with most common battery separators, gel, and polymer electrolytes are inherently flexible, ₄₅,₄₆ most solid-state electrolytes are fragile and inflexible and require a mixed matrix approach to achieve flexibility. ₄₇,₄₈

In balancing volatility and flexibility, gel polymer electrolytes based on PVDF, PAA, polyionics liquids, or poly(ethylene oxide) (PEO) seem to be particularly promising. ₄₉-₅₄ These materials are mechanically flexible, safe, and easier to pack compared to liquid electrolytes. Mechanical stability is particularly important, when metal anodes (like lithium or zinc) are involved, and dendrite formation is a major concern for their functionality. ₅₃,₅₅ The main challenge of polymer electrolytes is their low ionic conductivity at room temperature. Gel electrolytes are reported, among others, for Zn₆-₅₈ and Li/S batteries, ₅₉,₆₀ and they have demonstrated good compressibility, ₆₁ which makes them good interlayer materials for stress accommodation. ₆₂

Packaging. Most of the active components require robust packaging that is impermeable to water vapor and oxygen, which excludes the use of polymers as the sole packing material. ₆₃ Traditional packaging, mainly metal cases, provides stability, impermeability, and electric terminals. For flexible batteries, the standard materials are multilayered laminates, ₆₄,₆₅ composed of a thin metal layer, mostly aluminum, ensuring low water, solvent, and oxygen permeability with an exterior protective and an interior heat-sealable polymer film. ₆₅-₆₇ These materials meet the safety requirements in LIBs only at the price of additional stiffness. For less-sensitive battery chemistries like aqueous Zn-ion batteries, flexible but sufficiently safe packaging is provided by polymers like polyethylene naphthalate (PEN)/polyvinyl chloride (PVC). ₆₅

Performance Comparison in Full Cells. For a flexible full cell battery, all components must provide both the appropriate electrochemical performance and the intrinsic mechanical stability upon bending. Therefore, the fabrication of a flexible full cell is not the mere assembly of the components, but the combination of the different layers with robust interfaces, so that they do not lose contact under mechanical stress. Cha et al. proposed a flexible battery based on a patterning process for interlocking the different layers (Figure 1d).¹₈ Hydrogel interlayers, which accommodate occurring stresses and/or soft packaging, which avoid additional stresses, are common strategies as well (Figure 1e).⁶₈

Generally, choosing a robust battery chemistry is as important as a stress accommodating structure for a flexible battery. ₆₉,₇₀ To date, this fact seems to lead to a trade-off between capacity and bending capability (Figure 2).
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Unfortunately, high-capacity materials like those used in Li/S batteries are already exposed to high internal stress due to their high lithium uptake and large volume change. This effect is amplified, e.g., in the sulfur cathode, which possesses weaker interactions resulting in capacity loss upon bending.74

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In general, the comparison of performance values has some limitations and should be considered together with information on device structure, materials use, and experimental details. Whereas cycling performance parameters are already on the way to be standardized,72,73 no guidelines for performance testing under bending are available. Cycling data broadly range from performance reports after one flexion up to thousands of bending cycles.18,25,28,55,71,74–86 Furthermore, the results of mechanical testing depend not only on the bending radius but also on the angle, speed, and tension stemming from packing and bending stresses of the various layers.16 The development of evaluation standards seems to be urgently needed to better classify and compare future progress in research on flexible batteries.

Current Challenges. Due to the elaborate manufacturing process and the associated high costs, as well as the fact that the technology is not yet ready for the market, most flexible batteries are still at the stage of company R&D72 and in the startup phase. Companies like Blue Spark Technologies or Molex LLC produce zinc polymer batteries for personal electronics. Big players in the field like Samsung SDI, Apple Inc., or Panasonic Corporation released or patented flexible batteries but are, however, not yet using them in their devices. Foldable phones currently on the market still contain bulky batteries and are, therefore, only bendable along defined axes, and they still come with technical and durability issues.87 It is expected that future cells with improved power efficiency and longer battery life will increase the use and demand for flexible devices in the coming years.

By balancing material properties and introducing hierarchical architectures, flexible full cell assemblies could reach the performance and stability needed for practical device integration. A robust flexible battery technology with stable electrochemistry would open up fascinating possibilities for including other functionalities. First attempts have already been made and many more areas will benefit from research on flexible batteries: functional energy storage textiles,88,89 pressure-sensing hybrid batteries,90–94 self-charging batteries,95–97 electrochromic microbatteries as screen components,98 transparent cells for integration into smart windows,99 or in solar cells,100 as well as batteries with transient functionalities.99 For all these new functionalities, flexibility can enhance their applicability and bring them one step closer to interesting customer-ready products.

3. DEGRADABLE BATTERIES

Transient or degradable electronics is an emerging type of technology wherein devices are built to operate for a well-defined period and, when no longer needed, can degrade at controlled rates.100 Such electronic platforms can offer a multitude of applications including implantable biomedical devices that resorb in the body after realizing their diagnostic functions, ecofriendly devices that reduce the problems associated with growing electronic-waste, and data-secure hardware systems which self-destruct to prevent unauthorized access to sensitive information.7,10 To realize self-sufficient degradable electronics, the development of transient batteries as an autonomous on-board power source is important. To date, there are very few examples of transient batteries in the literature. The main challenges are the deficit of suitable soluble materials, fabrication schemes, and battery designs that must fulfill completely different requirements than for traditional batteries. To ensure rapid progress in this field, it is necessary to expand the materials toolbox beyond classical battery materials and to optimize the battery structure with respect to transiency. Inspiration can be drawn from research activities in the field of bioelectronics, nature-inspired/derived biomaterials, and green electronics.
In the following section, a critical discussion of the materials used as components in transient batteries and their full cell assembly will be presented.

**Active Materials and Electrode Structure.** Transient primary batteries have been mainly developed based on the use of Mg metal (Figure 3a), or sodium ion chemistry. Mg as an anode material offers excellent electrochemical properties and good biocompatibility. However, its rapid corrosion in aqueous environments presents a serious challenge. To overcome this issue, several strategies such as alloying of Mg with bio compatible metals (Al, Zn, Zr), surface coating with biomaterials (β-tricalcium phosphate (β-TCP)), and design optimization (use of electro plated Mg) have been proposed. Mg and its alloys are coupled with benign cathode materials (e.g., Fe, Mo, W, molybdenum oxide, polypyrrole, Au) to develop fully degradable batteries. Jia et al. utilized a biore sorbable Mg alloy anode and Au nanoparticles as the cathode to demonstrate a biodegradable Mg–air battery, whose enzymatic degradation was observed in buffered protease solution at 37 °C (Figure 3b). Degradable primary sodium-ion batteries were developed using aqueous electrolytes and naturally occurring or edible electrode materials such as melanin, activated carbon, and manganese oxide. These electrode materials can reversibly bind with the sodium ions that are present inside the human body, thus offering in vitro applications.

For secondary/rechargeable systems, the most representative type comprises LIBs (Figure 3c). Fu et al. demonstrated the first rechargeable transient LIB based on dissolvable electrodes including vanadium oxide (V$_2$O$_5$) as the cathode and lithium (Li) metal as the anode. In this work, V$_2$O$_5$ was selected as the cathode due to its comparatively high theoretical capacity and its ability to dissolve in alkali solution, formed by the reaction of Li metal with water. Subsequent research efforts focused on improving the electrochemical performance while still achieving fast transience behavior. Nevertheless, their application in biomedical and ecofriendly devices is still severely limited due to the generation of an alkaline environment that can have adverse biological and ecological effects. As an alternative, one could target greener energy-storage materials derived from abundant resources like redox-active biopolymers, which can be tuned via nano- or microstructur ing to undergo programmed degradation.

**Current Collector and Conductive Additive.** Biodegradable metals such as Mg, Mg-based alloys, Fe, Mo, and W serve as current collectors for transient primary batteries. Most of these metals are known to play significant physiological functions and demonstrate appropriate degradation rates in vivo. They are used either as self-supporting foils compatible with most processes, temperatures, and solvents or as highly flexible polymer-supported thin films. The difference in their dissolution behavior arises from surface morphology, grain structure, and presence of pit holes that control the kinetics of dissolution. Secondary transient batteries have been demonstrated with thin layers of inert metals such as Cu, Al, and Ni deposited onto degradable polymeric substrates as current collectors. As a promising substitute to metallic conductors, researchers have also reported the use of carbon-based current collectors (e.g., carbon black) that are blended with degradable polymers to achieve transiency.

**Electrolyte and Separator.** The electrolytes used in transient batteries so far can be classified into nonaqueous electrolytes, aqueous electrolytes, and polymer electrolytes. The nonaqueous electrolytes were used in transient secondary LIBs in combination with a separator made of water-soluble polymers such as PVA, PEO, and polyvinylpyrrolidone (PVP). The organic electrolytes provide a considerable advantage in terms of their large voltage window, which enables high power density, but their toxicity is a major concern. Environmentally benign aqueous electrolytes were mostly used to activate transient primary batteries designed for...
implantable medical devices.\textsuperscript{123} For example, using physiological fluid (PBS) as the electrolyte, polycaprolactone (PCL)-coated biodegradable Mg–Fe batteries delivered an average power of 30 mW for 100 h, sufficient to power a commercial pacemaker for up to 4 days.\textsuperscript{102} Volatilization of solvent leading to changing electrical properties and a small potential window of aqueous electrolytes hinder practical applications.\textsuperscript{123} As an alternative, polymer electrolytes have been proposed that can also function as a separator and as glue to improve mechanical integrity.\textsuperscript{101} Reported examples tested for transient batteries are the solid polymer electrolyte composed of sodium chloride and PCL\textsuperscript{124} and gel electrolytes based on a biocompatible ionic liquid (choline nitrate) embedded in silk\textsuperscript{103} (Figure 4a,b). Unfortunately, the performance of the gel electrolyte was found to decline at high discharge rates due to its low ion migration rate.

**Packaging.** The concept of transience relies on packaging strategies balancing device operation lifetime and dissolution rate. Effective encapsulation is required to ensure stable operation for the desired period before degradation starts. To date, biodegradable polymers such as sodium alginate, silk, polyanhydrides, PVA, and PCL were applied as encapsulation layers for transient batteries.\textsuperscript{101–103,114,120} Their properties such as crystallinity, thickness, and composition were tuned to obtain a more predictable or programmed battery lifetime.\textsuperscript{102} For instance, an additional crystallized silk film on top of an encapsulated primary Mg battery extended its stable operation in PBS solution from 64 to 109 min (Figure 4c).\textsuperscript{103} Nonetheless, the high water permeation rates of biodegradable polymers limit the development of batteries with longer lifetimes.\textsuperscript{126} One viable solution can be the coating of biodegradable packaging materials with thin films of metal oxides and/or nitrides (\(\text{SiO}_2\), Si\(_3\)N\(_4\)), or with hydrophobic compounds (beeswax, paraffin). The low water permeability of these materials can facilitate longer battery lifetimes together with programmed degradation.

**Performance Comparison in Full Cells.** To improve the practical application of transient batteries, continuous efforts should be invested in fabricating degradable full cell batteries. Rogers and co-workers first reported a biodegradable, polyanhydride-encapsulated primary Mg–Mo battery that exhibited a stable voltage of 0.4–0.7 V for 24 h and demonstrated transience in PBS solution. The battery, however, suffered from low power density and comparably short lifetime.\textsuperscript{101} Huang et al. developed a high-performance primary Mg-MoO\(_3\) battery with extended lifetime, delivering a stable voltage of 0.6 V for 250 h. The battery powered a standard LED for 16 h in PBS solution and was shown to be fully degradable both \textit{in vivo} and \textit{in vitro} (Figure 4d–f).\textsuperscript{11} Fu et al. fabricated the first transient secondary LIB that operated in organic electrolytes yet rapidly dissolved in an aqueous environment due to triggered cascade reactions.\textsuperscript{114} The battery provided a high working voltage of 2.8 V but could only be charged and discharged for four cycles. These examples show that, by careful selection of different battery components and fabrication schemes, innovative prototypes of transient batteries can be achieved. Clearly there is a trade-off between a battery’s electrochemical performance and transient behavior, and therefore, it is necessary to find an optimum balance.

**Current Challenges.** The field of transient batteries is still in its infancy, but it is expected that in the future transient batteries would serve as advanced power supplies for green, disposable, and transient electronics. To boost the development, new electrode materials and electrolytes must be investigated, and their degradation behavior should be studied in great depth. New modes of trigger such as light, heat, temperature, pH, etc. can be utilized to precisely control the onset of the degradation process. At the end of life, the resulting degradation products can ideally be recaptured and recycled to enhance economic and environmental viability of transient electronics. Further efforts should be directed toward developing strategies for seamless integration of such batteries in functional transient systems to achieve completely self-powered devices.\textsuperscript{128,129}
4. TRANSPARENT BATTERIES

Although it still seems a bit like science fiction, research on transparent electronics has increased significantly. The most fascinating property of such devices is that their transparency makes it possible to superimpose a virtual electronic image on a real background. Displays become invisible, which opens up the possibility of integrating them into car windshields, eyeglasses, or windows to display information without obstructing the view. In smartphones or tablets, transparency enables unique designs. If the energy source is to be built-in, it must be transparent not to impair the optical appearance.

Up to now, the literature on transparent batteries is relatively scarce, although a few instructive examples of transparent energy storage devices have been reported.130,131 Similar to flexible batteries, which have to overcome the rigidity of typical battery components, and to degradable batteries, which have to replace nondegradable components with transient materials, transparent batteries face the challenge that neither traditional electrodes nor usual separators are transparent.

In this section, we focus on presenting some recent progress about different components and their combination into transparent batteries.

Active Materials and Electrode Structure. There are three major approaches to reach transparency for active materials. Firstly, wide-band-gap transparent conducting oxides that show high transmission in the visible range can be applied as active materials.132,133 Al-doped ZnO thin films, deposited by a midfrequency sputtering technique, as the anode for LIBs offered a high transmittance of 84% and a high specific capacity of 301 mA h g\(^{-1}\).134 The electrochemical tests, however, were performed on nontransparent stainless-steel substrates. The second strategy is to reduce the dimensions of active materials and decrease the electrode thickness down to a scale below their optical absorption length.135 In Figure 5a–e, Roeder et al. applied a sol–gel dip-coating technique to prepare a transparent full cell battery consisting of a 600 nm thick Li4Ti5O12 anode and a 150 nm thick LiMn2O4 cathode.136 Li4Ti5O12 presented a visible light transmittance of 30–75%, transitioning from dark-blue to colorless depending on the charge/discharge state. The LiMn2O4 electrode showed a green color in the uncharged state and turned to orange when being charged. The cycling behavior of the full cell was not reported. A transparent LIB composed of single-walled carbon nanotubes and V2O5 nanowires as the anode and cathode, respectively, was produced by layer-by-layer assembly (Figure 5f). The transmittance of these thin film electrodes was adjusted by the number of sprayed layers. The 255–300 nm thick anode showed a transmittance of 87%, while the cathode presented 93% transmittance at a thickness of 150–300 nm.137 Another approach toward transparent electrodes involves the...
preparation of gridlike electrodes with feature dimensions below the visual acuity of human eyes. The grid-structure approach was used for fabricating thin-film electrodes in a transparent, all-solid inorganic LIB. LiCoO2/lithium phosphorus oxy-nitride (LiPON)/Si structures were fabricated on glass substrates using photolithography and etching processes to achieve a transmittance of 60% with 65.3% of open area (Figure 5g). Yang et al. used a microfluidics-assisted method to process LiTi3O5 and LiMn2O4 into grid-structured electrodes for a transparent LIB. A transmittance of 62% in the visible and near-infrared range for the electrode was obtained (Figure 5h–k). The single electrode had 65% areal vacancy and exhibited a transmittance of 62%. Since the transparency of the electrodes decreases with increasing thickness and covered area, but the stored energy increases linearly with the mass loading of the active material, a trade-off between electrode loading and transparency has to be accepted when working with gridlike structures.

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Current Collector and Conductive Additive. Current collectors for transparent batteries are expected to meet the following requirements: transparent, lightweight and thin, and electrochemically stable in the electrolyte and over the operating voltage window of the electrodes. In addition, to achieve high rate capability and energy density, their resistance should be as low as possible to provide enough conductivity, while high currents are applied. Transparent conductive films are well established and integral components of electronic devices such as touch screens, displays, and solar cells. Alternative materials include metallic nanowires (24.5 Ω sq−1 sheet resistance, 71% transmittance), carbon nanotubes (57 Ω sq−1, 90%), graphene (350 Ω sq−1, 90%), and conductive polymers like poly(3,4-ethylenedioxythio-phene):poly(styrenesulfonate) (PEDOT:PSS, 260 Ω sq−1, 90%). Examples of opaque batteries include lithium/sulfur batteries fabricated on a graphene current collector, which were cycled between 1 and 3 V and Cu nanowire arrays in combination with Fe3O4 as the LIB electrode working between 0.02 and 2.5 V. To further improve the transparency in these examples, the film thickness of the current collectors has to be decreased.

Electrolyte and Separator. In traditional batteries, the liquid electrolyte is completely transparent, while the separators (e.g., glass fiber, polypropylene/polyethylene) are opaque. Thus, transparent solid electrolytes have been considered as the most promising for transparent batteries. For practical use, transparent solid electrolytes, including inorganic and polymer electrolytes, should possess high transparency, good thermal/chemical stability, a wide electrochemical voltage window, low electronic conductivity, and high ionic conductivity (higher than 10−4 S cm−1). As an inorganic and transparent solid electrolyte, Al2O3-doped LiLa2Zr2O12 delivered a bulk conductivity of 9.9 × 10−4 S cm−1 at 25 °C, but the transmittance was only 30%. Polymer electrolytes mainly employ monomers with suitable donor atoms like O and N to coordinate cations, and to form polymer salt complexes. Polymer electrolytes with low crystallinity for high transparency, low viscosity for high ionic conductivity are preferred, such as PVDF and its copolymers with hexafluoropropylene (PVDF-HFP), PEO, polyacrylonitrile (PAN), and poly(methyl methacrylate) (PMMA). For instance, a gel electrolyte composed of a PVDF-HFP membrane infiltrated with 1 M LiClO4 in ethylene carbonate/diethyl carbonate (EC/DEC) achieved an ionic conductivity of 2 × 10−3 S cm−1 with a transmittance of about 99%.

Packaging. There are no systematic studies of the various packaging methods for transparent full cell configurations. For all-solid-state batteries, usually tight adhesion between the electrode and electrolyte layers is achieved, which makes packaging obsolete. If the electrodes are fabricated on rigid substrates like glass, the batteries can be sealed simply by applying methyl ethyl ketone or double-coated adhesive polyester tape to the edges of the electrodes. To obtain better protection in air, a transparent battery was encapsulated into a closed thermoplastic PVC bag. Future work should focus on exploring transparent polymer materials with high transparency, low water vapor permeability, and thermal sealability, such as PEN, polyethylene terephthalate (PET), polycarbonate (PC), and polyphenylene sulfide (PPS).

Performance Comparison in Full Cells. A fully transparent battery requires all the integrated components to be transparent. However, in most of the transparent battery reports, only the performance of individual components is reported. Here, the main challenge lies in ensuring long cyclability along with high transparency. Yang et al. fabricated a transparent and flexible battery with a transmittance of 57% using LiMn2O4 and LiTi3O5 as the cathode and anode materials. However, at a current density of 100 μA cm−2, it could only cycle for 15 times with a remaining capacity of 80 mAh g−1. Pat et al. reported a transparent all-solid-state battery by stacking silver paste/Li4Ti5O12 anode/Li4PO4 electrolyte/LiFePO4 cathode/ITO/glass. A capacity of 600 μAh g−1 was achieved with a high transparency of about 80%, but no data on cyclability were reported. Apart from the lack of long-term performance, energy densities of transparent batteries are insufficient for applications in smart windows and displays. To address this issue, an interesting strategy could be to explore high-theoretical-capacity battery systems, like the Li/S battery (1675 mA h g−1), Li–air battery (3840 mA h g−1), Zn–air battery (820 mA h g−1), and Al–air battery (2980 mA h g−1).

Current Challenges. Transparent batteries are realized through layer-by-layer assembly, microfluidics-assisted methods, sol–gel dip coating, and magnetron sputtering. In order to make transparent batteries even more attractive for emerging electronics, they should additionally be equipped with flexibility and electrochromic properties. In spite of all the progress, significant challenges still exist. Specifically, full cell configuration, cycle life, and their integration into devices (transparent smartphones/tablets, e.g., a commercially avail-
able smartphone needs an energy density of 246 Wh kg⁻¹,¹⁵⁸ smart windows, or displays are still missing. The capacity of the batteries has to be increased by exploring high-capacity systems as active materials for anodes and cathodes. To achieve high mass loadings of active materials in transparent electrodes, new electrode architectures have to be designed, and to maximize the voltage operation window, the electrode–electrolyte combination has to be carefully selected. Overall, to meet the requirements for practical use, energy density and lifespan of the batteries have to be improved without sacrificing the transparency.

5. CONCLUSIONS AND OUTLOOK

Driven by the rapidly changing market for consumer electronics, the topics of battery research have expanded significantly. In addition to the continuous improvement of electrochemical performance, which of course remains an important branch of research, the search for new electrochemical systems beyond lithium-ion and new battery designs has become the focus of interest. Batteries, which in addition to energy storage also offer new properties such as transparency, flexibility/stretchability, or degradability, open up fascinating possibilities for innovations in wearable, optoelectronic, implantable, or ingestible electronic devices.

Robotics,¹⁶⁰ unmanned aerial vehicles (UAVs or drones),¹⁶¹ and prosthetic devices¹⁶² are some other popular areas that can greatly benefit from the advancements in multifunctional batteries. Power sources are usually the biggest constraints on the potential capabilities of these devices. Developing batteries with multiple functionalities could offer new degrees of freedom in designing these devices, which is not possible with the traditional bulky and rigid batteries. In fact, these batteries could seamlessly integrate onto the soft, elastic, and curvilinear surface of such electronics, thus becoming a structural element of the device rather than just a power source.

Regardless of whether the battery is flexible, transparent, or degradable, these properties are completely opposite to those of a conventional battery, and therefore, the entire battery design, including all components, their processing, and their arrangement and assembly, has to be rethought from scratch. Although there have been impressive advances in this area, they are mainly at the level of individual components, and examples of full cells that combine these additional features with acceptable electrochemical performance are very rare. Considering that the electrochemical performance of traditional batteries has been optimized over several decades, it is obvious that concerted and dedicated research efforts will be required to produce multifunctional batteries with the same level of energy storage capabilities. For a long time to come, there will probably still be a fine line between balancing the compromise between electrochemical performance and desired additional properties. Nevertheless, the prospect of having a truly multifunctional battery is fascinating and worth any research effort.

AUTHOR INFORMATION

Corresponding Author

Markus Niederberger — Laboratory for Multifunctional Materials, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland; orcid.org/0000-0001-6058-1183; Email: markus.niederberger@mat.ethz.ch

Authors

Linda A. Wehner — Laboratory for Multifunctional Materials, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland; orcid.org/0000-0001-6654-8157

Neeru Mittal — Laboratory for Multifunctional Materials, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland; orcid.org/0000-0003-3499-2944

Tian Liu — Laboratory for Multifunctional Materials, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland

Complete contact information is available at: https://pubs.acs.org/10.1021/acscentsci.0c01318

Author Contributions

L.A.W., N.M., and T.L. contributed equally.

Notes

The authors declare no competing financial interest.

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