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Review

Advances in paper-based battery research for biodegradable energy storage

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ABSTRACT

The increased demand for energy due to industrialisation and a steadily growing population has placed greater strain on the development of eco-friendly energy storage devices in recent years. Current methods with high efficiency are limited by high costs and waste. As a result, greater importance has been placed on the development of low-cost, lightweight, flexible, and biodegradable energy storage systems developed from paper and paper-like substrates. This study reviews recent advances in paper-based battery and supercapacitor research, with a focus on materials used to improve their electrochemical performance. Special mention is made of energy-storage configurations ranging from metal-air and metal-ion batteries to supercapacitors. Furthermore, methods of fabrication, functional materials, and efficiency are reviewed to offer prospects for future research into the field of paper-based Na-ion batteries. The review provides an updated discussion of recent research conducted in the field of paper-based energy systems published over the last five years and highlights the challenges for their commercial integration prospects.

1. Introduction and sustainability vision

The rapid growth of the global population and increased industrialization is creating a huge demand for energy, particularly in developing regions. Energy is the basic need to fulfill human social and economic development while increasing the status of human health and welfare [1]. The absolute repercussion of the increase in energy demand is the continued use of non-renewable fuel sources, namely, coal, oil, and natural gas. These have proven over the years to be the leading source of electricity generation worldwide responsible for human development [2, 3]. While they have shown to contribute to human development, they have major drawbacks through the emissions of enormous amounts of harmful air pollutants, as well as carbon dioxide (CO_2), the most consequential man-made climate changing greenhouse gas [4]. Mitigation of the dire effects of climate change brought about by using toxic fossil fuels, natural gas, and nuclear energy, among others, can be accomplished through the progressive implementation of renewable energy sources [5]. Therefore, renewable energy installations need to be paired with energy storage devices to facilitate the storage and release of energy during off and on-peak periods [6]. Over the years, different types of batteries have been used for energy storage, namely lead-acid [7], alkaline [8], metal-air [9], flow [10], and lithium-ion batteries (LIBs) [11]. These batteries have great power and energy density, giving them relatively good performance characteristics. However, they are not perfect solutions and have several notable drawbacks, including safety concerns [12,13], ecological impact [14], poor low-temperature performance [15], limited lifetime [16], large weight [17], lack of flexibility [17,18], limited resources [19,20], and expensive cost of production [21, 22]. As a result, the demand for inexpensive, lightweight, flexible, eco-friendly, and biodegradable energy storage has surged.

Paper-based batteries have attracted a lot of research over the past few years as a possible solution to the need for eco-friendly, portable, and biodegradable energy storage devices [23,24]. These batteries use paper substrates to create flexible, lightweight energy storage that can also produce energy. The fabrication process involves the amalgamation of highly conductive materials such as metals, conductive polymers and allotropes of carbon; graphite [25], fullerenes [26–28], graphene [29–31], carbon nanotubes (CNTs) [32–35], nanocrystals [36–38], and nanowires [39–42] with an ordinary sheet of paper [43–46]. They are electrical devices created with electrodes containing nanoscale structures (for increased surface area) of long-chain carbohydrate molecules to enhance the rate of electric supply [47,48]. They have comparable

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functions to traditional batteries, such as producing energy for portable electronics and toys, while also being eco-friendly and able to function both as high energy batteries and as capacitors. In addition, they are non-corrosive and therefore don't require any housing or encapsulation [49].

Paper offers interesting properties as a solution for the manufacturing of low-cost rechargeable batteries:

- It is widely available and is inexpensive.
- It is combustible, meaning that paper devices can be 'recycled' very economically through incineration.
- It is lightweight, flexible, and thin.
- It offers a huge amount of surface area for the reagent to be retained.
- It is environmentally friendly, biocompatible, and biodegradable [50].

Furthermore, paper has metamorphosed into various roles within the battery system, such as a current collector when coated with conductive material, substrate for electrode deposition, and electrolyte storage [51].

However, fabrication of paper batteries remains a challenge. To address this, ordinary paper could be employed as a backbone for the creation of paper-like nanocomposite films. That may be simply ramped-up for industrial use [52]. The nanomaterials are applied on paper by various printing processes with varying results. The printing procedure on paper may be achieved in an automated technique, like coating (spray coating or doctor blade) [53,54], screen printing [55], inkjet printing [56], filtration and more costly approaches including photolithography. Moreover, the possibility of a power source being directly merged on paper for system integration with biosensors, electrical circuits and displays has resulted in the development of interesting paper electronics [57]. The structural properties of paper, such as rough and porous surfaces, facilitate ion and electron transportation through the entire structure for attaining high power performance, distinctively within the electrode [51].

Up to now, different types of paper-based batteries and energy storage devices are produced for several applications, for example, paper-based fluidic batteries for on-chip fluorescence assay analysis on microfluidic paper-based analytical devices (μPADs) [58], urine-activated paper battery for biosystems [59], photoelectrochemical paper devices combined into supercapacitors [60], microbial paper-based fuel cells for disposable diagnostic devices [61], and high energy density paper-based lithium-ion batteries [62]. Supercapacitors and lithium-ion batteries are great energy sources for high-power paper electronics. For paper-based μPADs and small power devices that need power for only a small period (a few minutes), biofuel cells and paper-based electrochemical batteries are a great fit. For sportswear and military uniforms, paper-based nanogenerators are good candidates [50].

2. Paper-based batteries and biodegradable energy storage devices

Paper-based batteries are applied on the operating principles of conventional batteries such as metal-air and lithium-ion batteries (LIBs), as well as on different energy storage devices such as supercapacitors [63] (See Table 1). With cell components such electrolytes and separators integrated on the paper substrate to create a fully functional paper-based batteries. The sections below explain the incorporation of paper into the different types of battery and other energy storage devices in detail while stating the potential applications for this type of technology.

Table 1 shows the comparison between metal-air batteries (MABs), lithium-ion batteries, and supercapacitors. In operation, both metal-air and supercapacitors rely on redox reactions, while lithium-ion batteries use the Li^+ ion reaction. This means that metal air and supercapacitors undergo an electrochemical reaction where the metal at the anode gets oxidized to produce metal ions that travel through the electrolyte to the cathode and react with metal (O_2 in metal-air batteries), producing a redox reaction responsible for energy generation. In LIBs, the Li^+ ions are produced from the cathode and travel through the electrolyte to the graphite anode where they are stored. Existing battery technology can generate power in the μW to mW range for application as rechargeable energy supply in low-power biosensors and high-power electronics, alike. Specifically, the passivation layer of the solid electrolyte interfaces and poor cycling stability of MABs, the high cost and degradation of separators used in LIBs and the low-energy density and self-discharge ability of supercapacitors limit their widespread application. The low-cost, thin structure, light weight, flexibility, biodegradable nature and simple customization of paper-based energy systems continue to offer attractive possibilities for their transition to market. Incorporation of paper substrates into various battery components provides a competitive advantage. Research into paper-based and paper-like electrode and support materials is the primary focus of recent research for all energy systems discussed in this review while paper incorporation as reservoirs, ion-exchange membranes and separators in MABs has also been discussed (Fig. 1).

In this review, we provide a comprehensive summary of the integration of paper-based substrates into various energy storage devices. Different fabrication processes, battery components as well as application of the fabricated paper-based energy storage devices and their benefits related to their biodegradable nature are discussed. Specifically, recent advances and current trends into biodegradable paper-based energy systems is discussed. At the end of this review, the challenges involved in the research of paper-based batteries and energy storage are discussed, including the prospects of better realizing practical applications of integrated and electronic devices in the future.

2.1. Metal-air batteries

Metal-air batteries (MABs) are part of an innovative class of half-closed power sources working in an open-air environment [70]. A metal-air battery consists of a typical battery layout with two electrodes: an air-cathode and a metallic anode. It also has a current collector, separator, and electrolyte useful for operation. The oxygen reduction reaction (ORR) and the oxygen evolution reactions (OER) occur simultaneously during charge/discharge in the air cathode [71,72]. The main advantages of MABs are their remarkable low functioning costs compared to other batteries due to the vast availability of the cathode reactant, which is ambient oxygen, followed by the total eradication of the oxidation storage system. On the anode side, the considerable search for low-cost metals has focused on the incorporation of lithium (Li) [73], potassium (K) [70], magnesium (Mg) [74], zinc (Zn) [75–77], aluminium (Al) and iron (Fe) [78–80].

In addition to their cost advantage, MABs consistently exhibit large amounts of theoretical energy density, for example, 1218 Wh kg^{-1} for Zn-air, 5928 Wh kg^{-1} for Al-air, and 5779 Wh kg^{-1} for Li-air. Compare this with the relatively lower 265 Wh kg^{-1} for a standard Li-ion battery [81].

The development of MABs is both of fundamental and practical importance, and they are eco-friendly and non-toxic [82]. MABs are attractive not only as compact power sources for portable electronics and electric vehicles but also as compelling energy transfer stations or energy storage devices to manage energy flow among renewable energy generators, such as wind turbines and photovoltaic panels, electric grids and end-users [64]. Replacing conventional MAB components with paper substrates offers unique opportunities for improved disposal and incorporation into flexible electronics.

2.1.1. Zinc-air batteries

A Zn-air battery (ZAB) is fundamentally created from current collectors applicable to either anode or cathode, consisting of an ordinary configuration of Zn metal anode, an alkaline electrolyte such as potassium hydroxide (KOH), an electrically insulating separator to regulate ion transport, and an air cathode. At the anode, a Zn oxidation reaction

Table 1
Paper-based batteries and energy storage devices

	Metal-air batteries	Lithium-ion batteries	Supercapacitors
Operation parameters	Redox reaction	Li^+ ion reaction	Redox reaction
Application of paper	Reservoir, ion exchange membrane, electrode and/or supporter	electrode and/or supporter	electrode and/or supporter
Power generation	μW–mW	mW	mW
Electrode	Metallic catalyst	Lithium-based metal oxide materials	Metal oxides or other carbon-based materials
Potential application	Low-power biosensors	High-power electronics	High-power electronics
Remarks	Rechargeable	Rechargeable	Rechargeable
Advantages	High theoretical power density compared to LIBs [64,65]	Low self-discharge rate [66]	Balancing energy storage with charge and discharge times [67]
Disadvantages	Poor cycling stability [68]	Relatively expensive [66]	Low energy density [67]
Limitations	Formation of passivation layer called solid electrolyte interface (SEI) [69]	Separator damage can result in fire [66]	Self-discharge rates and gradual voltage loss [67]

takes place, while an oxygen reduction reaction occurs at the cathode for electricity generation [83]. Lately, ZABs have gained significant recognition for their various practical advantages, which include higher energy density compared to iron-air batteries, bigger reserves than Li-air batteries, and metal properties that are more stable than those in Al-air batteries. These characteristics of ZABs have proven effective in developing marketable prototypes in electronic hearing aid devices. Therefore, the present imminent progression of inexpensive, facile homogenisation and extremely efficient ZABs is necessary [81]. However, the dependence on metal shells prevents ZABs from being a truly biodegradable energy source. This has led to the development of paper based ZABs, allowing for the creation of low-cost batteries that are biodegradable and readily disposable.

The power generation of current ZABs remains a considerable problem because of the ORRs and OERs at the cathode. They compromise numerous protons associated with electron transfers and they are well known as slow in essence, leading to small current density and large electrode polarization. Also, passivation, corrosion, and dendrite growth of Zn metal during recharge brings about poor cyclability to readily accessible Zn anodes [84].

The bulk of research conducted on paper-based ZABs in recent years has centred on the use of carbon-based paper substrates as solid cathode supports in conjunction with or loaded with iron precursors for the fabrication of paper-based metal air batteries [85–91]. Carbon and carbon fibre solid supports have been extensively investigated for their large, inexpensive surface areas and excellent electrical properties to significantly enhance both OER and ORR catalytic kinetics in air cathodes exhibiting excessive potential with weak oxygen reversibility due to sluggish ORR and OER throughout charge and discharge [89,92–94].

Wang *et al.* fabricated a paper cathode by loading Fe_{20}@N/HCSs on the air cathode electrode to operate as a bifunctional electrocatalyst for ORR and OER. This showed excellent electrocatalytic activity towards ORR and OER compared to a Pt/C + RuO_2 conventional bifunctional electrocatalyst. By attaining open circuit voltage of 1.57 V, the highest power density was 140.8 mW cm^{-2}, while the specific capacity was 726 mA h g^{-1}. This was compared to an open voltage of 1.44 V, a maximum power density of 85.6 mW cm^{-2}, and a specific capacity of 699.4 mA h g^{-1} respectively for Pt/C + RuO_2 [91].

Wang *et al.* created a nonplatinum air cathode catalyst that was Fe, Cu coordinated zeolite imidazole framework (ZIF) derived loaded on a carbon paper for enhancing ORR. The synthesis Cu@Fe–N–C electrocatalyst displayed higher peak power density of 92 mW cm^{-2} compared to that of Pt/C (74 mW cm^{-2}) due to the bimetallic active sites, large surface area and abundant pores. It also displayed exceptional durability and methanol resistance in both acidic and alkaline solutions [89].

Alternative ways have been explored to further rectify the problems associated with ORR and OERs. An *et al.* created a filter paper derived bifunctional binder-free three-dimensional mesoporous carbon with Co_3O_4 loaded on the surface of the air cathode electrode. The Co_3O_4 enhanced the catalytic activity, corrosion resistance and stability. The electrical conductivity of the electrocatalyst was enhanced by the addition of the carbon matrix. The three-dimensional (3D) matrix improved the reaction kinetics, and its large surface area increased the number of actives sites on the electrocatalyst, enhancing the proceedings of ORR and OER while maintaining the merits of controllable size and being a binder free air cathode electrode. The battery displayed improvement in performance during charge and discharge, with a discharge power density of 71 mW cm^{-2}. However, it was less than the expensive commercial Pt/C at 74 mW cm^{-2}. The battery also maintained stability for 24 h during charge and discharge. Furthermore, at 20 mA cm^{-2} the battery achieved a specific capacity of 828 mA h g^{-1} and energy density of 488 Wh kg^{-1} [95].

Many strategies to enhance the sensitivity of paper-based Zn air batteries have been investigated recently. One such approach was to combine microfluidic flow systems with conventional Zn-air batteries. The microfluidic Zn-air cells (μZACs) consist of a co-laminar flow which is propelled via a 3D electrode with hole channels for electrochemical energy conversion. The permeable electrodes are designed with regulatable Zn electrodeposits to increase activity by improving the surface area and stabilizing the Zn deposit by manipulating its morphological features. This device was created through electrodeposition of Zn on permeable carbon paper electrodes to allow KOH solution to spread through the anode, therefore increasing the contact surface.

As such, at 4 M KOH and 3 mL h^{-1}, the highest obtained power densities were 308 and 317 mW cm^{-2} respectively, when Pt/C was replaced with α-MnO_2 and Mn_3O_4 cathodic materials respectively. The plethora of Mn^{3+} atoms in α-MnO_2/C is associated with the electrocatalytic activity and cell performance attained by the MnO_2 cathode. Furthermore, the cell performance achieved with these materials was discovered to be comparable to or better than reported Zn-air batteries operating in a "passive flow" manner, implying that μZACs are suitable for renewable-energy [96].

Over the past few years, a limited amount of research has been focused on the Zn anode. However, new interest in Zn anodes has arisen due to the higher effective surface area of zinc particles, which is beneficial for improving battery performance [97]. Furthermore, the incorporation of additives such as binders helps to enhance conductivity and Zn metal distribution to form thin, compact Zn deposits [98].

Correspondingly, Chotipanich *et al.* fabricated a flexible ZB with addition of a carbon support and binders such as sodium silicate ($NaSiO_3$) and bismuth oxide (Bi_2O_3). The fabricated anodes were Zn/C–$NaSiO_3$ and Zn/C–Bi_3SO_3 in 9 M KOH electrolyte. The battery performance was tested and with the introduction of $NaSiO_3$ and Bi_2O_3 the battery performance was enhanced. $NaSiO_3$ helped to reduce the passivation on Zn active sites during discharge by increasing the solubility of ZnO in the electrolyte while enhancing discharge capacity. Bi_3O_3 improved the ionic conductivity for the anode and inhibited passivation of Zn particles by clearing the way for the transportation of hydroxide ions (OH^-) across the multilayer Zn particles. The addition of additives offers a promising future in extending the application of ZABs [99].

Recent material on ZABs has been focused on using non-platinum bifunctional catalyst for the cathode electrode. The catalyst is coated into the carbon paper, filter paper, and polypropylene membrane

substate via screen printing and spray coating. This has results in the rise of metal nitrogen co-doped carbon (M-N-C) electrocatalyst such as Fe_{20}@N/HCS and Cu@Fe–N–C. Other non-platinum based electrocatalysts include Co_3O_4, α-MnO_2 and Mn_3O_4. On the anode electrode, the binder infused zinc electrode have shown a huge rise namely Zn/C–$NaSiO_3$ and Zn/C–Bi_3SO_4. This shows that there is room for expansion in finding different materials for paper-based ZABs.

2.1.2. Aluminium-air batteries

There is great interest in aluminium-air batteries lately, driven by promising battery technology as the power source for the imminent future. This is mainly due to the low cost of aluminium as the earth crusts most plentiful metal with a relatively low price per kg [100], high performance, high theoretical voltages (2.7 V), high energy density (8100 Wh kg^{-1}) and capacity (2980 mA h g^{-1}), and its theoretical capacity is similar to lithium (3860 mA h g^{-1}) [101]. Despite their great performance, Al-air batteries are still plagued by a well-known issue known as Al self-corrosion, which is caused by high reactivity of Al with alkaline electrolyte. Other concerns include complicated water management and possible leakage risk. These concerns have limited the use of Al-air batteries, perhaps most notably in the case of portable electronic device application [102]. Various approaches have been investigated to tackle these problems, for example, combing aluminium with other metals to produce alloys and using non-aqueous electrolytes to prevent anode corrosion and passivation [103]. Nonetheless, these approaches increase fabrication costs, making them unsuitable as power sources for small devices (Fig. 2).

Recent research has been focused on integrating microfluid cells on paper-based substrates to create electrolyte storage for the mitigation of Al corrosion while maintaining water management within the cell and reducing the potential of leakage hazards. The paper-based microfluidic Al-air battery (abbreviated as μAl-air) may be utilized as a downsized power source for small-scale biodegradable electronics. Shen *et al.* applied the concept of microfluidic cells on a paper substrate by creating a highly efficient paper-based μAl-air constructed from a thin sheet of fibrous paper called the paper channel. The channel is a filter paper containing cotton linter fibres of pure alpha cellulose with small lignin and polysaccharides placed in the middle of an Al foil anode and graphite foil cathode. The paper channel is connected to an absorbent pad on the one end, which is submerged in 1.5 M KOH electrolyte. This assembly works without employing a high-cost air electrode or any exterior fluidic transport device to maintain water management within the cell and reduce leakage potential, as shown in Fig. 2 below [80].

The distinct microfluidic layout can assist in reducing crucial setbacks of typical Al-air batteries such as product-induced electrode passivation, battery self-discharge, and high-cost, intricate air electrodes. These are the main barriers for Al-air batteries to gain access to commercial market. The paper-based μAl-air battery pouches are fabricated by combing the anode, cathode, paper channel and absorbent pad through lamination using an office laminating machine. This fabrication method allows for low-cost, easy fabrication technique, while creating great electrochemical performance. The produced battery delivered a specific capacity of 2750 A h kg^{-1} (@ 20 mA cm^{-2}) and an energy density of 2900 Wh kg^{-1}, which are 8.3 and 12.6 magnitudes larger than their non-fluidic counterpart and remarkably surpasses numerous miniaturized energy sources.

A variety of paper substrates have been studied for battery fabrication due to their widespread availability. The sorption capabilities and rate of fluid flow are important criteria for selecting the perfect paper source to enable the electrolyte to travel through the device by capillary action without the need of external pumping and to act as electrolyte storage. Avoundijian *et al.* displayed an enhanced low-cost paper-based aluminium-air battery using KOH as an electrolyte. The battery was fabricated from Kim Wipes, which were chosen as the paper substrate due to the fast flow rate of the electrolyte in contrast with chromatography paper. The highest amperage and power was 17.4 mA and 30 mW respectively, with 1.5 M KOH being employed as the electrolyte and a 9 cm^2 battery configuration with a 5.1 cm^2 anode electrode area and a 3.75 cm^2 cathode electrode area [104]. The battery was able to produce enough energy (in a series layout) to power a pregnancy test, a flashlight, and light-emitting diodes (LEDs), while maintaining a cost-effective fabrication process and being environmentally friendly. As a way of avoiding the problems associated with water management and leakage risk, the aqueous alkaline electrolyte is solidified with a gelling agent to produce a gel electrolyte.

Wang *et al.* created a paper-based liquid-free Al-air battery based on a porous cellulose framework that was able to store the alkaline paper-based gel electrolyte (PBGE). This enabled fabrication of a flexible, ultrathin, and biodegradable device. Furthermore, this allowed for the substitution of the electrolyte and the Al anode following their depletion to enable the battery to be used many times when applied in a mechanical-rechargeable Al-air battery, while also eradicating any water management issue and leakage risk. The PBGE is intercalated between the anode and cathode with two cubic casts fabricated from poly(methyl

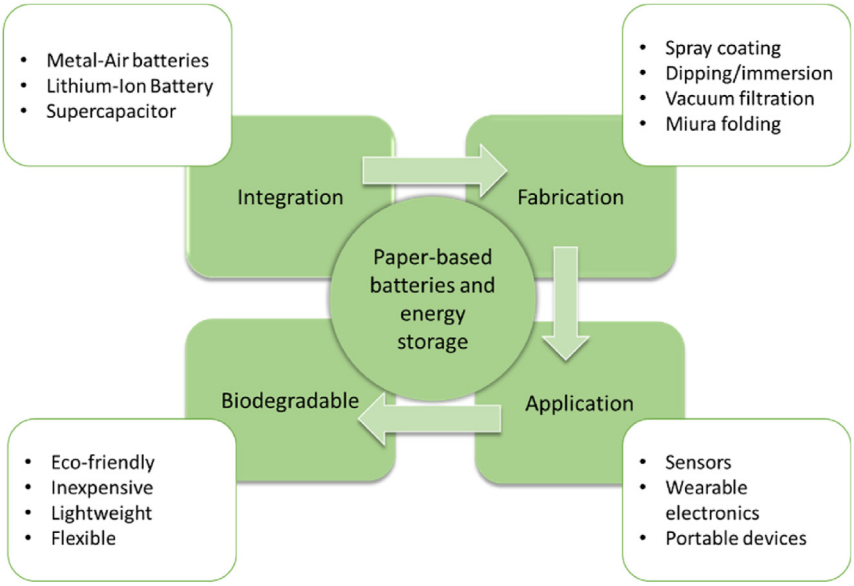

Fig. 1. Illustration of the review of paper-based batteries and energy storage devices: integration, fabrication, typical applications, and biodegradable.

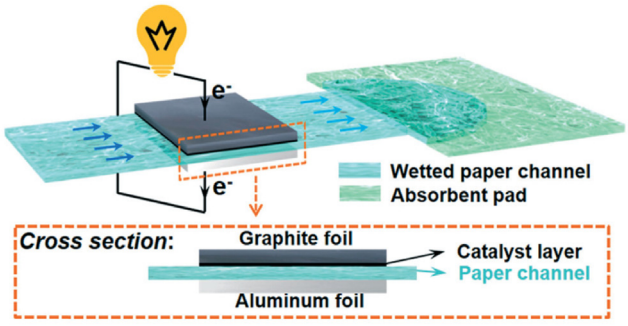

Fig. 2. Diagram of a paper-based μAl–air battery [80].

methacrylate) (PMMA) supplying the pressure and acting as the battery shell, as shown in Fig. 3 below. This battery configuration produces an open circuit voltage (OCV) of 1.5 V, a peak power density of 3.5 mW cm^{-2} and a maximum current density of 4.3 mA cm^{-2} [105].

This design holds much promise for the development of a fully integrated self-sustaining, flexible, inexpensive, and biodegradable Al-air battery.

Different routes have been taken over the few years to tackle the anode corrosion reaction and the self-discharge with troublesome oxygen evolution reaction in an alkaline electrolyte. One method is to dope the aluminium metal to decrease the self-discharge loss in efforts to improve anode behaviour. As such, Yu *et al.* fabricated an Sn-doped Al anode for Al-air batteries. The anode is 3D printed using laser sintering on a current collector for improved high mass loading. The laser sintering provided actual elimination of organic solvents in the slurry, improved the conductivity, and enhanced the electrical contacts of Al nanoparticles and electrochemical performance of Al-air batteries. The cell produced a discharge capacity of 239 mA h g^{-1} and an operational voltage of 0.95 V. This combination of additive procedure and laser printing technology allows the chance of producing batteries with high mass loading for improved battery performance [106].

The bulk of the research for Al-air batteries is focused on mitigating Al corrosion caused by the electrolyte spillage. This has resulted in most of the paper-based Al batteries being focused on using the paper substrate as an electrolyte storage medium. Different types of paper substrates have been used, ranging for filter paper with cotton linter fibres of pure alpha cellulose with small lignin and polysaccharides to Kim Wipes immersed in KOH electrolytes and paper-based gel electrolyte (PBGE). The use of different types of paper substrates allows room for expansion into different aspects of the batteries, which may result in improved Al-air batteries.

2.2. Lithium-ion batteries

Lithium-ion batteries have attracted significant attention due to their high operation voltage, high energy density, low self-discharging rate, and comparatively adequate stability and durability. However, they are limited by safety concerns, poor low-temperature performance, high bulk and weight, limited lifetime, and expensive production. They also create environmental waste and are difficult to dispose of. As a result, researchers have begun looking into how to integrate paper substrates into lithium-ion batteries to create thin, flexible, and lightweight batteries to meet the demand for flexible and wearable electronics [107].

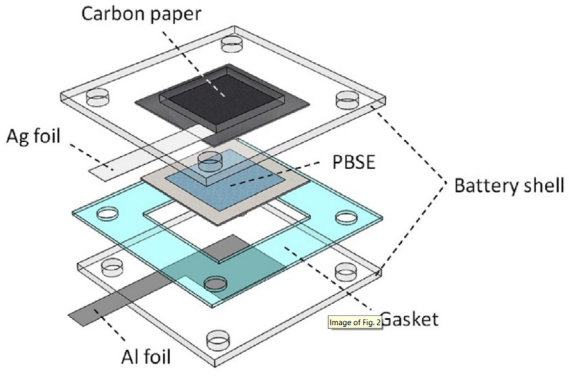

Fig. 3. Diagram of liquid-free Al-air mechanical-rechargeable battery prototype [105].

Paper-based lithium-ion batteries have unique properties, namely (i) large surface roughness and porous structure for enhancing power generation, and (ii) mechanical flexibility that is well suited for use in flexible electronic devices [50]. The application of paper and paper alternatives can be expanded even further by integrating them with single-walled carbon nanotubes (SWCNTs) to create highly conductive paper for lithium-ion batteries. The main advantage of the integration of lithium-ion batteries onto a paper substrate is that paper allows the control of electrons and ion transfer throughout the whole construction of the battery, particularly inside the electrode, and helps to attain high-power performance [51]. Wang *et al.* demonstrated how conformal dipping of SWCNTs on the surface of polycellulose paper immediately transformed the paper into a highly conductive channel for electron transport. This was accomplished by the development of a Li-ion battery of single-walled carbon nanotubes/polycellulose paper (SWCNT/PP) [108]. Fig. 4 below shows the fabrication process of SWCNT/PP.

The full cell created from the SWCNT/PP displayed a speedy capacity of discharge of 153.3 mA h g^{-1} with 90.6% of coulombic efficiency at 0.1 °C [109].

The need for flexible electronic devices has driven researchers into finding different ways to create flexible electrodes for LIBs. As such, Wang *et al.* used a simple and inexpensive vacuum filtration method to create a paper-based anode electrode for LIBs. The electrode contains linear N-doped carbon nanotubes (N-CNT, one-dimensional) and spherical C-coated MoS_2 (MoS_2@C, zero-dimensional), entrapped into a porous recycled paper (RP, two-dimensional) that is attained from wastepaper. The fabricated N-CNT/MoS_2@C/RP film (N-M-RP film) is used as an anode material for paper-based LIBs. The N-M-R film delivers a discharge capacity of 989.6 mA h g^{-1} after 50 cycles (high ICE of 70.5%) at a current density of 100 mA h g^{-1} [110].

Another way to create a flexible paper-based electrode is through solution-based paper making process followed by in situ carbonisation. Cao *et al.* demonstrated the fabrication of ultra-thin MoS_2 nanosheets (active material) exfoliated from bulk counterparts using TEMPO-oxidized cellulose nanofibrils (CNF) as assisting agents. The framework of the MoS_2 was accomplished with CNTs acting as conductive fillers followed by additional in situ carbonisation. The synthesised carbonised MoS_2/CNF/CNTs films displayed improved electrical conductivity and specific surface area due to the in-situ carbonisation. The MoS_2/CNF/CNTs films delivered a specific discharge capacity of 930 mA h g^{-1} and cycling stability of the reversible specific capacity gradually decayed from initial 330 mA h g^{-1} to 255 mA h g^{-1} after 100 cycles at 200 mA g^{-1}. The carbonized MoS_2/CNF/CNTs hybrid film has proven that it can directly act as an ideal paper-electrode for LIBs without using traditional metallic current collector makes it appropriate for the next-generation flexible/wearable electronics in future [111].

Recently, Li-ion paper-based battery research has devoted much effort to paper folding techniques. The techniques allow for continuous folding of the paper substrate to create a compact battery which offers a high energy density of real energy. The batteries created from this technique are not small but have increased energy in every footprint of their surface area. Li-ion batteries are significantly enhanced by simple and complex folding techniques, such as Muira folding [108].

Cheng *et al.* [147] tested the simple folding technique and intricate Miura-ori pattern on a paper-based Li-ion battery by investigating various versions of folded batteries. Planar, unfolded, one-fold, two-fold, and three-fold versions are shown in Fig. 5 below.

The folding effects of the electrode were electrochemically tested. The voltages of the folded, unfolded, and planar cells were comparable with

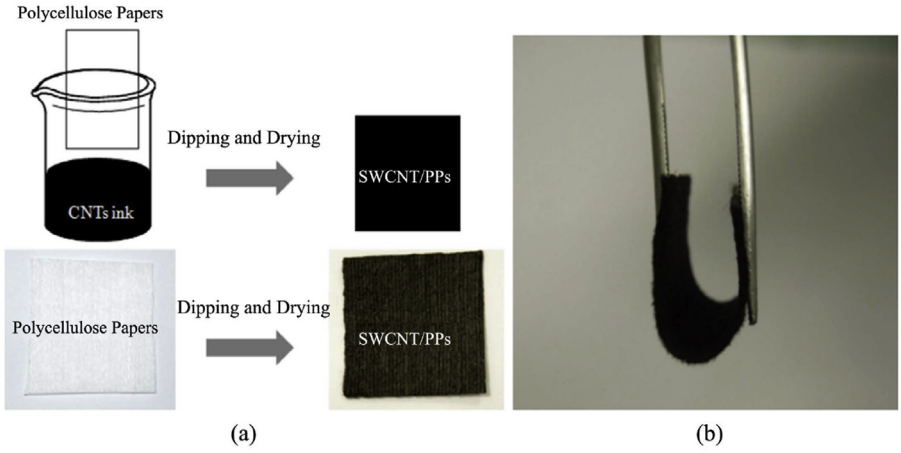

Fig. 4. (a) The creation process of conductive SWCNT/PP and (b) a photo illustrating the flexibility of the fabricated material [108].

similar gravimetric capacities. Folding increased the area capacity of the cell, with the capacity increasing with each extra fold. This effect resulted in the batteries with one-fold, two folds, and three folds having 1.9×, 4.7×, and 10.6× the areal capacity of the planar one. The number of folds resulted in enhanced contact area in the middle of the active materials and the CNTs loading, resulting in the folded batteries having higher coulombic efficiency than the unfolded cell. This confirms that multiple folding can be used on Li-ion batteries to enhance electrochemical performances [108] (Fig. 6).

Furthermore, Miura folding was employed as a highly effective way to create a more compactly folded paper. The Miura folding allowed the cell to have 25 layers with a geometric area of 1.62 cm^2. Once folded, the cell was sealed in an aluminized polyethylene (PE) bag to avoid air seepage.

Miura folding substantially enhanced the areal capacity in comparison to planar cells. In the 20^{th} cycle, the Miura folding products is about 14× the areal capacity for folded cells. This may allow additional enhancement in the energy density of Li-ion batteries. For future applications, paper-folding can help create Li-ion batteries with substantially higher areal energy densities while being flexible, lightweight, low-cost, and biodegradable [108]. The benefits of folded paper energy systems allow for highly compact batteries to be created. Layered systems with high surface contact between adjacent layers is possible resulting in improved ion transport and energy density. The folded battery research creates unique possibilities for further expansion into layered batteries in future applications. Stacked and layered batteries with hydrophobic separators created from alternating functionalized paper and paper-like electrodes and dividers are possible.

3. Supercapacitors

Lately, supercapacitors (SCs) have gained substantial recognition as a major group of energy storage devices. They serve as a crucial technology aider in categories varying from transportation to consumer electronics. In contrast to batteries, supercapacitors are superior in terms of high-power density and extended cycle life, rapid charge and discharge rate, and lesser environmental implications [112,113]. However, their energy densities are generally lower than Li-ion batteries. The notion of paper-based supercapacitors has obtained a tremendous amount of traction because of the need to manufacture thin, lightweight, flexible, and inexpensive devices [114]. Paper-based supercapacitors are constructed on composites of conductive active materials with cellulose fibres. Which are made up of continuous cellobiose components with high aspect ratio capabilities of forming strong inter and intramolecular hydrogen bonds emanating from the fibres with hydroxyl-functionalized surfaces [115]. The above-mentioned advantages make cellulose fibres an ideal structural component in affiliation with natural conductive active materials such as carbon nanotubes [116] and graphene [117–120].

Zhang *et al.* presented the integration of supercapacitor components into nanocomposite paper aligned with multi-walled carbon nanotubes (MWCNTs) embedded on micro-fibrillated cellulose (MFC) to fabricate electrode sheets and produce a strong, flexible, solid-state paper-based supercapacitors [121]. This paper-based supercapacitor displayed adequate capacitive behaviour in addition to calculated specific capacitance at 154.5 mF cm^{-2} at 20 mV s^{-1} from cyclic voltammetry [121].

Alternatively, CNT-based paper supercapacitors have again emerged with distinct manufacturing processes [122–125]. Hu *et al.* made a hybrid supercapacitor electrode on cellulose paper with an electrode configuration of cellulose paper/CNT/MnO_2/CNT. The electrode was prepared by separate immersion and electrodeposition processes. The electrochemical performance of the electrode was analysed with a standard three-electrode system in Na_2SO_4 solution. The electrode demonstrated impeccable potential in supercapacitor application with an achieved capacitance of 327 F g^{-1} at a scan rate of 10 mV s^{-1} [126].

Furthermore, carbon nanofiber-based supercapacitors have an extensive outlook in powering wearable electronics. As such, Ni *et al.* fabricated a 3D needle-like polyaniline@hollow carbon nanofibers composite for flexible supercapacitors, by preparing a poly(acrylonitrile-*co*-*β*-methylhydrogen itaconate) copolymer for utilisation in the synthesis of flexible hollow carbon nanofibers (HCNFs) via a electrospinning method. Followed by polymerisation with needle-like polyaniline (PANI) to produce 3D flexible HCNFs/PANI composites, which displayed a high capacity 1196.7 F g^{-1} at 1 A g^{-1} and good cycling stability (90.1% retention at 5 A g^{-1} after 3000 cycles). When applied to a symmetrical supercapacitor the HCNFs/PANI composites produced an exceptional electrochemical performance with high energy/power density (60.28 Wh kg^{-1} at 1000 W kg^{-1}), with great cycling stability (90% capacitance retention after 5 A g^{-1} 3000 cycles). This established that HCNFs/PANI composites have a great potential in various applications of flexible energy storage devices [127].

Recently, Nagar *et al.* developed an all-paper-based supercapacitor by employing graphene to create conductive substrates using Xerox paper substrates with a redox-active electrolyte. The redox-active electrolyte (iodine redox couple) is used for enhancing the paper device's performance. The hybrid device displayed enhanced volumetric capacitance of 130 mF cm^{-3} compared to the 29.6 mF cm^{-3} of the printed graphene paper on which the device was based [128].

Yun *et al.* used a non-conventional mulberry paper to create a paper-based supercapacitor. The paper substrate had carbon black as the active material and Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) as a highly efficient, transparent, conductive polymer with high ductility [129]. The mulberry paper was used as it had longer fibres than commonly used paper (2–2.5 times longer) [130]. The mulberry paper-based supercapacitor cell exhibited high energy density of 29.8–39.8 Wh kg^{-1} and power density of 2.8–13.9 kW kg^{-1} with 90.7%

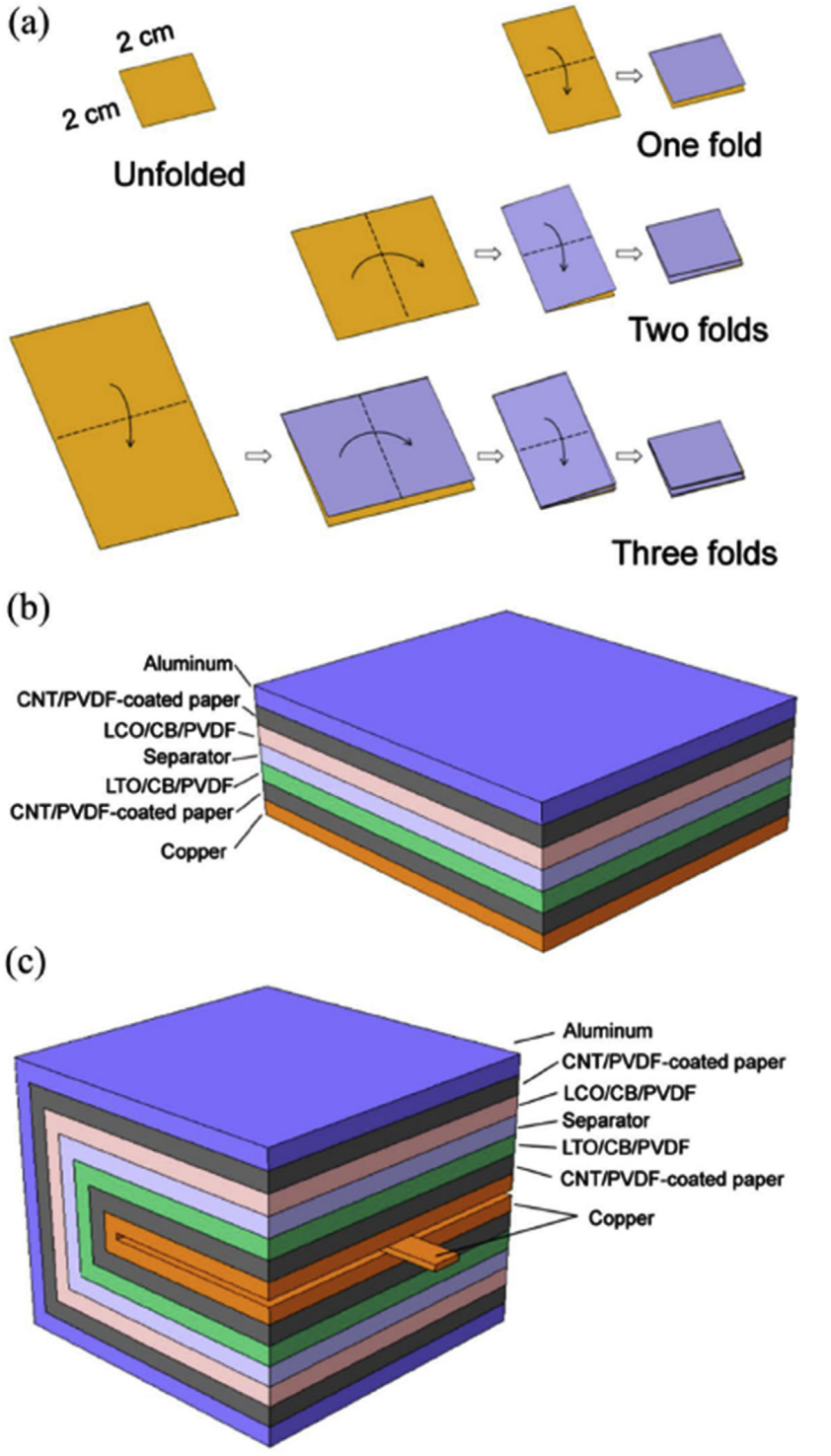

Fig. 5. (a) Folding process of one-fold, two-fold and three-fold configurations, (b) planar, unfolded complete cell, (c) complete cell with a single fold.

retention of its initial capacity over 15000 charge-discharge cycles [131]. The encapsulation of many paper supercapacitors come from papery composites of cellulose such as Li-ion batteries. As a result, the incorporation of supercapacitors into other paper electronics demands additional research, even though they can produce high current/power.

4. Other cell components

Apart from electrode materials, the performance of batteries and other energy generation and storage systems is facilitated by cell components responsible for ionic and electron transport and layer separation. The sorption capabilities, liquid transport through capillary wicking action and ease of functionalization of paper-based substrates is harnessed.

4.1. Electrolytes

An electrolyte is a medium inserted between the anode and cathode that enables ion transfer to take place. It has a crucial function in managing the power density of the battery. Furthermore, for a battery to

produce high capacity, stable, and flexible energy storage, the electrolyte must have properties such as the following: [132] high ionic conductivity ($\geq$0.1 mS cm^{-1} at room temperature and beyond), high ion transference number and minor electronic transference number, high flexibility, high electrochemical steadiness window, good mechanical and thermal stability, robustness (efficiently retaining its performance for a long enough time inside the working requirements of the device), and low cost. It must also be eco-friendly [133].

The resulting material should encapsulate the eco-friendliness of water electrolytes and the higher voltage capabilities of the organic salts. However, these stipulations are not easily achievable using conventional materials [134].

There are several types of electrolytes commonly used in paper-battery research with different properties pertaining to the type of application they are applied in. Vastly used electrolytes are namely, aqueous, gel and polymer electrolytes. Aqueous electrolytes, consisting of water-soluble salts, are water-based electrolytes which are non-combustible and non-sensitive to moisture during the fabrication process, resulting inexpensive and easy to handle electrolytes [135–137]. However, the struggle to maintain their performance due to the very limited voltage window, that results in electrolysation of water when too much voltage is applied has been a major setback for their use in commercial batteries [138–140]. Aqueous electrolyte storage is most used in paper-based battery research due its low cost and maintenance free application resulting from the low decomposition over organic electrolytes.

In the last few years, studies have highlighted the advancements of solid polymer electrolytes (SPEs) as their framework is a solid or gel-like ion-conduction membrane comprised of a salt distributed in a polymer matrix, producing an ionically conducting solution. As a result, SPEs have many beneficial traits, including elevated operating temperatures, no flowing and corrosion after damage, and simplicity of application to electrochemical devices [141].

Gel electrolytes are substitutes for liquid-free solid polymer electrolytes and highly conductive regular liquid electrolytes [142–144]. The gel electrolyte is basically created by restraining certain amount of liquid electrolyte into a polymer backbone. The polymer functions as medium of retaining the physical strength and shape of the electrolyte [145–148]. The solidification of an electrolytes into a gel electrolyte is achieved through the addition of a gelling agent into the alkaline solution used as an electrolyte. The gelling agents mostly used are poly(vinyl alcohol) (PVA) [149–152], sodium polyacrylate (SPA) [153,154], polyacrylic acid (PAA) [151,155,156], and poly(ethylene oxide) (PEO) [105,157–159]. This change from liquid to the solid or gel makes the electrolytes versatile with application in paper-based batteries.

The use of gel electrolyte was demonstrated by Wang *et al.* with the creation of a paper-based gel electrolyte incorporated into a liquid-free Al-air battery [105]. The paper-based gel electrolyte (PBGE) was made by combining a gelling agent, sodium polyacrylate (SPA), chosen due to the high water-absorbing ability and its inexpensiveness, with a strong alkaline, sodium hydroxide. This mixture was impregnated onto the paper through high pressure suppression to produce a semi-transparent PBGE product as shown below in Fig. 7.

The electrolyte is functional in both mechanical-rechargeable or single-use Al-air batteries, delivering an (OCV) of 1.5 V and a peak power density around 3.5 mW cm^{-2}. Furthermore, the battery discharge specific capacity is as high as 900 mA h g^{-1}. Additional improvements can be made to the electrolyte properties by increasing casting time, enhancing gel loading, and adjusting polymer concentration to increase the battery power output to 6.4 mW cm^{-2} [105].

Numerous researchers have explored the advancement of aqueous Al-ion batteries in the past few decades. In the beginning, only diluted Al salt solutions were used as electrolytes, for example, $AlCl_3$ [160,161], $Al_2(SO_4)_3$ [162–164], $Al(NO_3)_3$ [165,166], and $Al(CF_3SO_3)_3$ [167]. These electrolytes exhibited complexities when applied to full battery systems with Al foil as the negative electrode, notably an H_2 evolution side reaction. Differently, non-Al materials were utilized for both positive and negative electrodes to prevent this problem. However, they resulted in drastic loss in specific capacity [166,168,169].

To address the reduction in specific capacity and reduce electrolyte cost, a water-in-salt electrolyte can be employed, as demonstrated by Wang *et al.* They developed a paper-based aqueous Al-ion battery that could store a water-in-salt electrolyte of highly concentrated $AlCl_3$ and with the capacity to reduce H_2 evolution potentials to −2.3 V vs standard hydrogen electrode (SHE) [162]. The cellulose paper was utilized as a substrate for water-in-salt electrolyte storage via capillary action [50]. The excess water was eliminated via the baking process to leave a highly concentrated $AlCl_3$ inside the paper. The fabricated battery delivered a voltage output of 1.6–1.8 V and a specific capacity of ~140 mA h g^{-1} at 1 A g^{-1}. This battery technology will soon offer a good solution for operating large-quantity radio frequency identification (RFID) tags, smart packages, and wearable biosensors.

4.2. Separators

In previous decades, separators have been overshadowed by advancements in electrode materials and electrolytes as they are not active materials in batteries. However, they play a crucial role in enabling ionic conduction and isolating electrical contact between electrodes [170]. They also impact cell performance, namely cycle life, capacity, and safety [171].

Recently, integration of paper as separators for paper-based batteries has risen tremendously, driven by favourable properties of paper such as large pore size, high porosity, low-cost, naturally abundance, biocompatibility, large surface area, electrolyte storage, and control of electrons and ion transfer [172–174]. The current commercially available and popular polyolefin microporous separators offer superior chemical stability and great mechanical strength. However, high thermal decline and inadequate electrolyte wettability may impact its separation function and ion transportation throughout the entire life cycle of the battery [171].

Different strategies have been used to rectify these problems. Yang *et al.* fabricated a biodegradable zinc battery out of hydrogel reinforced cellulose paper (HCP). The hydrogel paper functioned both as an electrolyte and as a separator. The thick separator created by the hydrogel in the HCP efficiently inhibited the mixing of anode and cathode inks while preventing short-circuiting. Furthermore, the mechanical strength of cellulose fibres was greatly improved, and the composites ion conductivity was maintained. When HCP is immersed in an alkaline KOH/LiOH (lithium hydroxide) solution it exhibits excellent electrolyte performance with a conductivity of 33 mS cm^{-1} as measured by impedance, along with a columbic efficiency above 95%. Printed batteries comprised of HCP electrolytes display good cycle performance an HCP printed zinc-metal (Ni and Mn) batteries displayed impressive volumetric energy density of 26 mWh cm^{-3} [175].

Another approach to fix the problems associated with separators has been the use of lyocell fibrillated fibres as paper-based separators for lithium-ion batteries. Lyocell fibrillated fibres (LF) are cellulosic fibres created through eco-friendly processes by dissolving cellulose in tertiary amine oxide *n*-methylmorpholine-*n*-oxide (NMMO). The resultant fibres exhibit exceptional heat resistance. Furthermore, properties of LF such as increased pore sized, porosity, and greater electrolyte affinity provide outstanding cycle performance with various current densities at 30 °C [176].

The lyocell fibrillated fibres could be further enhanced with the addition of a gelling agent, PVA, to produce a polyvinyl alcohol/lyocell dual-layer (PDL) paper-based separator for primary zinc-air batteries. This was proven by Wang *et al.*, who developed a PDL paper-based separator through a dual-layer forming papermaking technique, comprising a PVA fibre layer and a lycocell fibrillated fibre layer, which result in superior discharge and high temperature storage performance. The PLD dual layer porous structure allows for submicron-sized holes and efficient electrolyte preservation. This revealed that PLD paper-based separators, together with an inexpensive papermaking technique, can substitute the current commercial composite separator used for primary zinc-air batteries [177].

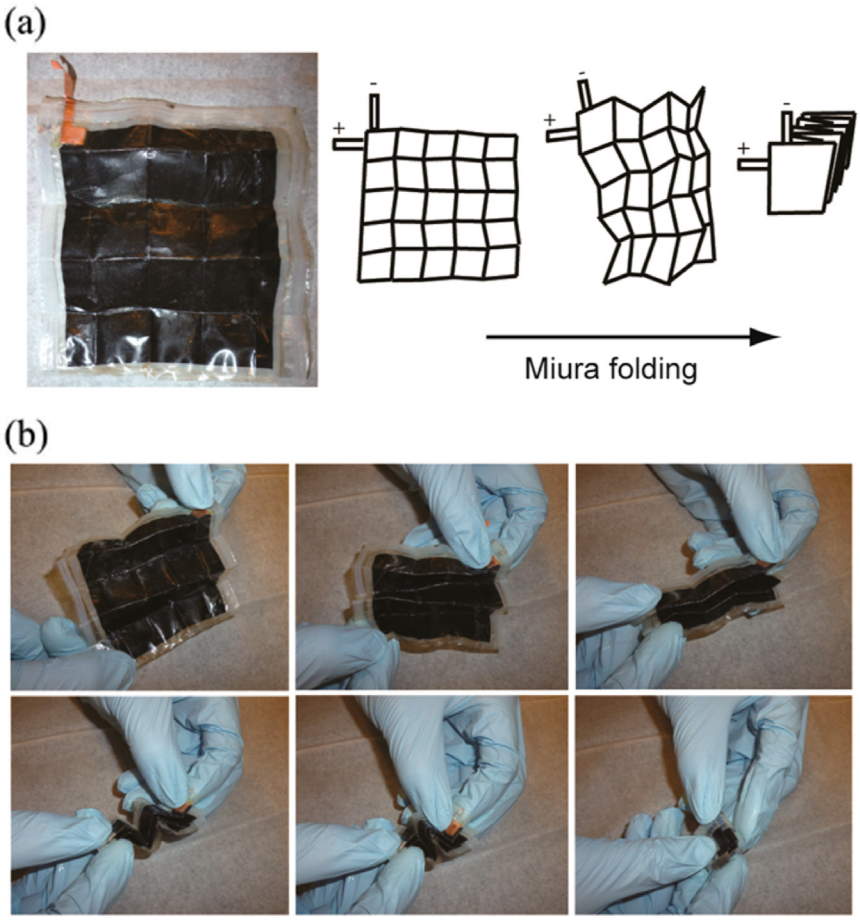

Fig. 6. (a) Miura folding process for a 5 × 5 pattern with a 6 cm × 7 cm sealed battery in an unfolded stated (sealing material Perylene-C), (b) Miura folding technique to produce a compact battery in a folded state [108].

5. Prospects for sodium-ion biodegradable batteries

Over the past few years, LIBs have been used for different applications from electric vehicles to portable devices [178,179]. Nevertheless, they still have a few drawbacks, such as high cost, cycle life, inferior low temperature performance, and safety [180].

Furthermore, the growing demand of LIBs puts immense pressure on the supply of lithium, pushing up the cost of this already precious metal. The goal is to find alternative battery materials that are readily abundant with scalable and cheap production processes [133].

One promising alternative is sodium (Na) metal, which is found right underneath Li in the periodic table. It is the 6th most common element in

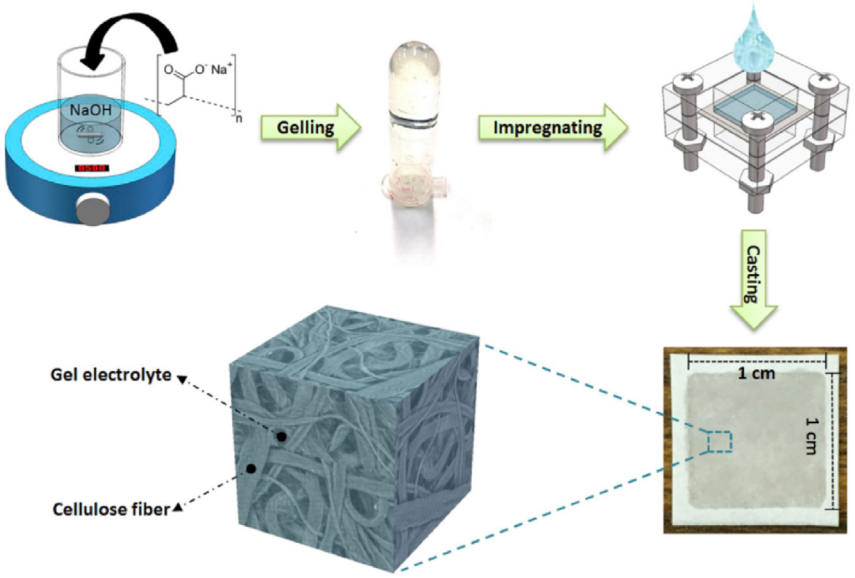

Fig. 7. Fabrication process of PBGE [105].

the earth's crust, at approximately 2.6 percent, and has a uniform geographic distribution, most notable as dissolved salts in sea water. This natural abundance makes it a relatively cheap. Furthermore, sodium-ion batteries (NIBs) do not rely on poisonous or uncommon materials such as cobalt, which is used as cathode material in LIBs [181].

Sodium has comparable chemical properties to lithium, meaning NIBs and LIBs will share a similar cell chemistry. Although sodium falls short of lithium with regards to open circuit potential (E^{o}) and charge-storage capacity (for metal electrodes), the enormous availability and inexpensive cost make it an attractive option for satisfying large-scale energy storage demands. Table 2 shows the advantages and disadvantages of NIBs and LIBs.

With the promising characteristics of sodium, NIBs could fill a gap in the available energy storage options. NIBs can be developed that are small, lightweight, thin, flexible, portable, eco-friendly, biodegradable, and relatively cheap.

Current research in paper-based Na-ion battery devices has been limited to paper-like substrates where active materials are doped onto carbon-based fibre supports to act as scaffolding. Zhang *et al.* developed a bifunctional polypyrrole (PPy) conductive polymer, encapsulated with cobalt phosphide (CoP) and nanowires (NWs) cultured on carbon paper (CP). The results for the fabricated 1D core-shell CoP@PPy NWs/CP anode displayed great electrochemical performance while enhancing charge transfer and reducing volume expansion. The anode material achieved a high areal capacity of 0.521 mA h cm^{-2} at 0.15 mA cm^{-2} after 100 cycles and 0.443 mA h cm^{-1} at 1.5 mA cm^{-2} after 1000 cycles [182].

Furthermore, Kong *et al.* developed MoS_2 nanosheets enhanced with Fe_3O_4 quantum dots cultured on graphite paper as free-standing anodes, with MoS_2 acting as a flexible substrate accepting volume variation of Fe_2O_3 quantum dots during the cycling process. The Fe_3O_4 functioned as spacers to balance the composite structure, creating active surfaces of MoS_2 nanosheets available for electrolyte penetration during charge/discharge processes. This resulted in excellent cycling and rate performances by the Fe_3O_4@MoS_2-GP anode, achieving 468 mA h g^{-1} and 231 mA h g^{-1} current densities at 100 mA g^{-1} and 3200 mA g^{-1}, respectively [183,184].

Attaining dependable mechanical flexibility and exceptional electrochemical performances still poses huge difficulty resulting in fewer numbers of flexible Na-ion storage cathodes. Fundamentally, a different approach must be taken in producing flexible Na-ion batteries. Particularly, the use of carbon cloth for the fabrication of $Na_3(VOPO_4)_2F$ cathode material was demonstrated by Deng *et al.* The cathode displayed exceptional rate capability and stable cyclic performance up to 1000 cycles as a result of surface-assemble of crystalline nanotubes on carbon fibres. The novel cathode was applied to a fully assembled Na-ion battery with $Na_2Ti_2O_5$ as the anode. The battery delivered an increased gravimetric energy/power density of 220.2 Wh kg^{-1}/5674.7 W kg^{-1} and can power LED indicators at bending angles of 0°–180° [185].

The findings demonstrate that conductive, porous, paper-like anodes/cathodes show great potential as electrode materials in Na-ion battery fabrication. To date, however, no work has been conducted on the direct transfer of Na-ion battery components into biodegradable paper-based substrates. In pursuit to create paper-based energy storage in this area, some challenges that may arise need to be considered. For Na-ion batteries a crucial issue would be whether the paper-based electrode is mechanically/structurally robust enough to accommodate large volume change during Na-ion insertion and desertion, since the ions are very large compared to Li-ions.

Table 2
Advantages and disadvantages of LIBs and NIBs

	Lithium-ion	Sodium-ion
Advantages	• High working platform • High energy density • Long cycle life • Fast charge and discharge • Good safety performance, no pollution, and no memory effect line control • Small self-discharge • Large temperature range	• Strong stability • High safety • Long service life • Wide range of applications • Easy access to raw materials • Waste recycling process is simple and pollution free • Fast charge and discharge • Sodium ion batteries have no over discharge characteristics, allowing sodium ion batteries to discharge to 0 V
Disadvantages	• High cost • Cannot discharge large current, need over-charge and over-discharge protection	• The comprehensive electrochemical performance is worse than that of lithium-ion batteries • The storage mechanism of hard carbon is not clear

6. Applications for emerging electronics

For diverse applications, new forms of biodegradable paper batteries are being developed, because batteries account for a significant portion of electronic waste pollution. Paper batteries can be twisted, folded, crumpled, moulded, cut, and sculpted, making them suitable for a wide variety of uses [186]. They are also relatively light in weight and are biodegradable. The combination of carbon nanotubes and cellulose paper allows for long-term use, consistent power, and energy bursts in the paper battery. The developed electronic devices can be applied in fields such as wearable electronics, medical sciences, vehicles, aviation, and other fields [187–189].

Wearable electronics have allured a significant amount of interest of the past few years, focusing on gathering various set of information such as temperatures, [190] strain (mainly bending strain) [190,191], pressure [192–195], light [196,197], biopotential [190,198], pH [190], gas [199–203], humidity and respiration [204,205], biochemical compositions [206,207], fitness tracking, medical diagnostics, and human-machine interface [208–213]. Even though flexible and bendable batteries to power them are fast growing, supplying adequate energy to power wearables is difficult [108]. However, researchers have been developing different ways to fix these challenges and produce wearable electronics with sufficient energy to power them. Zhao *et al.* developed a wearable smart watch for constant observation of sweat glucose levels by using flexible photovoltaic cells to produce energy and flexible paper-based Zn–MnO_2 batteries to store the energy. The potential to create flexible power sources on smart watches makes them a desirable device relating to user comfort, size, and safety [214]. Furthermore, paper-based batteries provide a long-lasting working time because of enhanced energy storage capacity and with rate of degradation upon long term use being similar to ordinary batteries at about 1000 cycles. This signifies an encouraging future for the next generation of wearable electronics devices.

7. Conclusion

The need to develop low-cost, lightweight, flexible, and biodegradable energy storage systems has led to several promising developments using paper and paper-like substrates. Active materials, separators, electrodes, and electrolytes might be used to improve flexibility, which could then be combined to create flexible electronic devices with promising electrochemical capabilities when compared to typical batteries. The use of cellulose paper in the production of flexible batteries started with research into the material's suitability as a substrate for energy storage devices, as evidenced by its good conductivity, porosity, and chemical stability.

By way of technology advances, the application of energy storage devices expands into new areas. Exploration of paper-based devices for the creation of light, flexible, and biodegradable electronics is dependent on the device's intended use. Lithium batteries, supercapacitors, and metal air batteries are among the battery types available. Electrochemical batteries or biofuel cells may be required for low-power, paper-based biosensors, as in medical devices, while lithium-ion batteries or supercapacitors are used for high-power electronics. However, both

performance and integration within a paper-based system present challenge. Almost none of the known technologies for producing a completely working paper-based system are commercially viable.

Further, future developments are needed in different areas of paper-based energy storage devices to ensure the growth paper-based devices. Novel materials and new fabrication techniques need to be discovered for making new paper-based electrodes with efficient electrochemical properties and stability for application in paper-based energy storage devices that operate under harsh condition. The new paper-based electrode must include self-healing to be able to fix the damage within the electrode and expand their lifespan, which can proof to be beneficial for certain energy storage devices. Electrolytes can crucially affect the energy density, power density, specific capacitance/capacity, and the stability of energy storage devices. The interfacial interaction relationship between the paper-based electrodes and electrolytes needs to be better understood. To create highly conductive and electrochemical efficient energy storage devices.

However, several problems are still evident in the performance and integration into a paper-based system. Furthermore, almost none of the known methods are industrially available to produce a fully functional paper-based system. In addition, most of the known methods to fabricate those power devices on paper substrates are either time-consuming or costly. Therefore, efficient fabrication and integration techniques are in urgent need of development to meet the practical use of these power devices for paper-based applications.

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Declaration of competing interest

The authors declare no conflict of interest.

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