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Paper-based devices for energy applications

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ABSTRACT

Paper-based analytical devices are lightweight, inexpensively produced, effective, and easily disposable; allowing for their suitable implementation in resource-limited areas. They allow effective handling of quantitative analysis in a diverse range of areas, from standard healthcare and environmental monitoring to water quality monitoring. Nonetheless, such devices often require an energy source for their complex assays or readings, preventing their effective use. Most commonly, conventional batteries are integrated into the device to serve as an energy source. However, considering its non-environmentally friendly approach to energy generation and its difficulty of being effectively disposed, a search for a new power source has begun. In light of the newly found potential of cellulose-based entities in the energy field, attention has been drawn towards a supposedly unlikely material: paper. Considering the potentials of such technology, this manuscript aims to describe the benefits of current and future technologies of paper-based devices in the energy sector. Here, we discuss the role of paper as a main platform or part of energy storage and conversion devices such as fuel cells, lithium-ion batteries, and alkaline batteries thoroughly.

Contents

1. Introduction

The issue of sustainable energy production from the environment to the supply power for the electric devices of various sizes is a crucial topic and receives much attention from the scientific community [1–3]. There has been a growing interest over the last decade in obtaining solutions for the world's demand from these sustainable energy sources [4–6]. Electricity is an intrinsic aspect of our current economy, being one of the more common services in everyday life. Electrical power dominates most of the important aspects of our economy. However, since most conventional methods of energy generation are dependent upon the exploitation of fossil fuels, a finite and non-environmentally friendly energy source, the world has begun witnessing an astounding increase

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of carbon dioxide in the atmosphere. Recently, the amount of atmospheric carbon dioxide has been measured to have reached 400 parts-per-million which results in an increase in the greenhouse effects. Therefore, there is an urgent need for a clean, renewable, sustainable, cost effective, and efficient source of energy [7,8].

Renewable sources are defined as resources that are reduced, but can feasibly be made to be sustainable. Paper is an example of such sources. When enough time is given, the trees grow into harvested trees and paper can be produced sustainably. Unlike plastic substrates, papers are made of wood cellulose rather than nonrenewable petroleum dependent sources. Thus, paper-based devices will not intensify the situation caused by the white pollutants, the long-lived plastics thrown in the environment [9–13]. "Green" flexible energy and electronic devices made of paper substrate are receiving a significant interest from research community because they are eco-friendly, as well as being cost effective, lightweight, available widely, and contain high flexibility and mechanical properties [8,14–18]. Excellent optical transmittance and mechanical strength of nanostructured papers allow the integration of flexible electronics and optoelectronics devices into the renewable materials [19].

Additionally, the expanding demand for a renewable energy supply is directly correlated to the need for environmental friendly energy devices. In other words, it is desirable to develop disposable energy devices that can be degraded easily. The miniaturized paper based fuel cells, for instance, can produce high energy density while being highly degradable [20,21]. In the area of energy storage devices, the paper based supercapacitors can play a significant role due to their low cost, high power density, quick charge–discharging rate, long-term stability, and nature friendly design [6,22].

The paper based devices are also recyclable, meaning that these devices can be used several times in a sustainable way. Recycling paper can significantly reduce the amount of greenhouse gas emission per ton of paper which can result in a decrease in the rate of global warming [23–26]. However, there is no doubt that the greenhouse gases are emitted during the paper recycling process. In this regard, some studies showed different ways to minimize the adverse impacts of recycling paper and make this process as environment-friendly as possible [27–29].

Generally, conventional diagnostic devices are expensive and bulky, thus the access to low-income remote regions is limited. In addition, with their complex construction and need for modern laboratories, distribution is hindered as targeted clientele are unable to provide personnel with the requisite technical training. With the invention of an inexpensive, flexible, and easily manufacturable device, developing economies can begin to have a starting point for cheap and accurate medical assays [30]. If such new products with some modifications, especially in relation to the cost of materials and fabrication procedure, went through, a global impact would be achieved. For example, using paper as a substrate greatly benefits the production of microfluidic paper-based analytical devices (μPADs) as it offers a number of useful attributes [31]. Initially constructed with glass or silicon as substrates, microfluidic devices were shown to be costly while preventing accurate analysis of fluidic samples. Difficulties associated with glass or silicon as substrates are based on the fact that neither possesses the characteristics necessary, such as permeability to gases, to work with live mammalian cells efficiently. In the last decade, the fundamental materials of microelectronics have expanded in such a manner that rigid, expensive substrates were replaced by low-cost flexible materials [32,33]. Recently, paper has become an increasingly popular choice for substrates in microfluidics for rapid diagnostics tests [34]. In μPADs, hydrophobic barriers are created on hydrophilic paper by different methods in order to make microfluidic channels that guide the fluids into discreet test areas. μPADs features a number of interesting attributes such as its capacity to wick aqueous fluids, which allows the passive transportation of liquids throughout the device without the use of external pumps and it can easily transport micro liter volumes samples to several detection zones. Additionally, its strong adhesion to several different materials, the resulting flexibility, and sturdiness of the fabricated devices are capable of being bent to varying angles without tearing (for most conventional fabrication methods). Moreover, the topology of the microfluidic channels can be extended into the third dimension by folding or stacking sheets of paper [35,36]. A current trend in this field is the deviation from centralized laboratories due to an increased demand for accurate and rapidly distributed medical information. This leads to a regression in the already deficient capabilities of resource limited regions to efficiently provide results and treatments to patients [37]. An integration of biological assays with multianalyte low-cost diagnostic platforms presents an ingenious way of performing health care across the globe. Fluorescent, electrochemical, and electrochemiluminescent assays are all quantitative assays that demand power to function [38,39]. In extremely resource-limited areas, the use of standard batteries pose difficulties in terms of their disposal, release of toxic pollutants, uncertainty regarding remaining charge, and requirement of a manual transfer to different devices [40]. Popular alternative power supplies for such assays include the integration of galvanic cells within the devices.

Although the paper based devices are receiving much attention at this time, there is no comprehensive report that covers different aspects of these devices in the area of Energy. In this review paper, we describe various applications of paper-based devices. This includes the recent developments in this area, fabrication methods, advantages and shortcomings of each technology followed by comparing the performance of different devices in each group.

2. Overview of fabrication methods

One of the main attractions of using paper as a substrate is its capability of wicking fluids through capillary action. In order to accomplish wicking, paper needs to be patterned or have micro channels built onto its surface, allowing for the transportation of fluids by the construction of hydrophilic and hydrophobic contrasts. The ability to pattern paper allows for a larger fluidic control not previously found in most elastomer microfluidic devices. Distinct fluidic operations which include: mixing [41], splitting [42], programmed delays [43,44], filtration, and even specific sample separation are now possible and have prompted further development in μPADs [31], which are now capable of advanced multiplexing and performing assays with greater sensitivities [45]. Most patterning methods can be subdivided into the following: a more physical approach comprised of sealing the paper's pores, the insertion of a hydrophobizing reagent onto the paper's fibers, and the chemical manipulation of the fiber surfaces with the aid of agents reactive to cellulose. There are many predominant methods such as plasma etching, ink jet printing, photolithography, wax printing and laser treatment used to pattern paper [34,46–53]. In order to allow the reader to have a generalized overview of the different fabrication methods associated with paper-based microfluidics, the following paragraphs will summarize the more commonly used fabrication procedures.

A patterning technique that is easily and rapidly performed is wax printing, which involves printing the desired wax patterns onto the paper's surface and then melting such patterns into the substrate to form the hydrophobic barriers [51]. Compared to other patterning techniques, wax printing is inexpensive and well

suited for mass production. When the wax melts, it spreads both laterally and vertically into the substrate. The vertical spreading is responsible for the formation of the hydrophobic barrier across the thickness of the paper, while the lateral spreading results in a decrease in the patterns resolution, as well as developing patterns that are wider than the original etching, and therefore are not as defined as when photolithography techniques are employed. Such a situation is primarily due to the layering of the paper's fibers, which are more commonly horizontal rather than vertical. As a result of the fibers' horizontal formation, the lateral spreading is much faster than vertical, resulting in a pattern wider on the front of the page than on the back.

Another patterning approach, called plasma etching, has the benefit of allowing the patterned device to maintain a large amount of flexibility. When utilizing plasma etching as a patterning approach, a sheet must first become hydrophobic, then treated with a conjunction of plasma and mask. As the untreated areas remain hydrophobic and prevent capillary penetration of sample liquids, a series of functional elements, such as switches, can be installed into the patterns. The resultant patterns are, however, shown to be slightly larger than the original design due to the tendency of plasma treatment to over etch the substrate under the mask, a controllable predisposition.

3. Paper-based devices for energy applications

The main application of paper-based microfluidic devices is to develop a low-cost and effective analytical platform for assays to be easily used in developing regions. One difficulty, however, is the development of the device's complete independence from any type of "external aid" for its use, such as external energy sources. Such development is difficult to achieve, especially during the infant stages of the technology as more specialized readers would require a larger amount of energy, forcing developers to implement external sources. Eventually, the apparent potential of cellulose based entities, such as paper, as an energy foundation for the readings will make the fabrication of such a device more feasible. The following sections will discuss different categories applied to paper-based energy devices, with a larger emphasis on batteries and energy storage devices, in which batteries refer to the use of fuel cells, as well as lithium-ion and aqueous/alkaline methods to generate power while energy storage is attributed to more generalized methods of energy storage.

Around 2.5 billion people live in extreme poverty, deprived of adequate healthcare, schooling, well established hospitals, and sufficient food [54]. This problem exists in our current global population of 7 billion, expected to increase by at least 2.3 billion by 2050 [55]. Those who live in resource limited environments neither have the technical commodities nor the human resources that others in more urbanized areas are accustomed to preventing their effective development [56]. For a country to have effective growth, it must have at the very least the following characteristics: (i) established healthcare institutions, (ii) international integration, (iii) adequate schooling systems, and (iv) efficient means for energy generation. As discussed earlier, microanalytical devices show a promising future in developing nations; nonetheless, their expansion is limited by the deficit of resources, especially for more intricate assays which require a power source. Paper-based energy storage devices span a wide variety of applications, such as photovoltaic, grid storage batteries, portable electronics, and unattended sensor networks [57].

Electrochemical energy can be stored in two main methods: using batteries or using electric double-layer capacitors as the medium. In batteries, the charge storage is a consequence of the electron transfer that generates a redox reaction in the electroactive materials [58]. In double-layer capacitors, specifically in supercapacitors, the charge storage is not produced by electron transfers, but rather by an electrostatic storage. Ideally, a suitable energy storage device would combine the benefits of both storage methods, bearing the high storage capabilities of batteries while also possessing the rapid power-discharge characteristics of capacitors [59].

Due to the capability of batteries to generate energy, they are more commonly associated with energy storage devices [60]. While the term "battery" is most commonly used, its basic electrochemical use is in reality its cells. Cells are directly integrated into a battery, with configurations that vary with different combinations of parallel and/or series connections, depending on the desired voltage and capacity [61]. The battery function, hereby named as the ability to generate power, is an important component of more specialized medical devices, especially in relation to more complex readings in microfluidics [62]. Fig. 1 shows the various applications of paper in energy sectors.

3.1. Paper as a highly conductive material

As already demonstrated, paper can provide a reliable microfluidic platform. Paper can also be an invaluable tool in energy storage as it can be made very conductive, with a resistance as low as $1\,\Omega\,sq^{-1}$. Such a condition can be stimulated by following a simple solution process composed of the conformal coating of single-walled carbon nanotubes (CNT) and silver nanowire films [68]. CNTs have high stability, electrical conductivity, mechanical flexibility, and surface area [22,69].

Such a method can lead to a low cost solution for high-performance energy-storage devices. As demonstrated by Cui et al., the process requires a surfactant and CNT ink, with sodium dodecylbenzenesulfonate (SDBS) chosen as the medium. The ink was applied by using the Meyer rod coating method, transforming the sheet quickly into a highly conductive paper with a sheet resistance of around $10\,\Omega\,sq^{-1}$ (Fig. 2). As paper easily absorbs solvents and has strong adhesion with CNTs, the fabrication process for a highly conductive sheet of paper is easier than that of plastic or glass. This use of substrate also benefits from the fact that ink rheology is easily applied to and absorbed by paper. This differs from plastic, in which the ink surface energy has to perfectly match with that of the elastomer, and its viscosity must be high enough to prevent defects caused by surface tension. The price for a plastic-based device further escalates when various additives are incorporated into the ink to prevent such consequences from arising when using ink rheology, eventually leading to the decrease of the final film's conductivity. Additionally, paper does not require surfactant washing processes, unlike other materials, in order to achieve high film conductivity. This simplifies the general fabrication process.

This method demonstrates how paper has another potential use in the energy field. Its use as a substrate dramatically improves adhesion, simplifies the overall coating process, and significantly lowers the production and final product cost.

Printing highly conductive material on paper is another method to make a highly conductive paper based material. In addition, this method has vast applications, such as film transistors [70–72], magnetic devices [73,74], light-emitting devices [75], solar cells [76,77], sensors and detectors [78,79]. However, due to the fact that the resistance of printed CNTs on a substrate is too high, these devices cannot be used widely unless the electrical resistance is improved using different methods. One of the common methods to handle this shortcoming is to add Ag nanowires or Ag nanoparticles due to their high conductivity [80–82]. The clogging of the nozzle is another issue that should be taken into account. From this respect, the Ag nanoparticles are preferred to the Ag nanowires, and can reduce the sheet

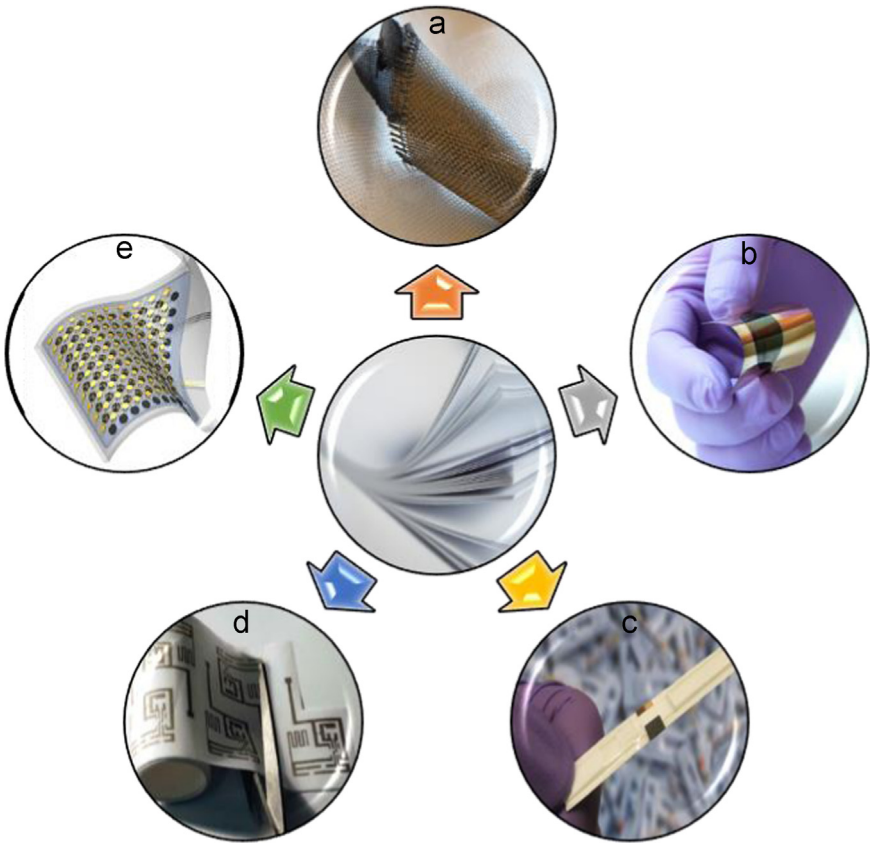

Fig. 1. Various applications of paper in the energy sector, including: (a) highly conductive materials [63], (b) flexible energy storage devices [64], (c) electronic circuits [65], (d) fuel cells [66], and (e) Li-ion batteries [67].

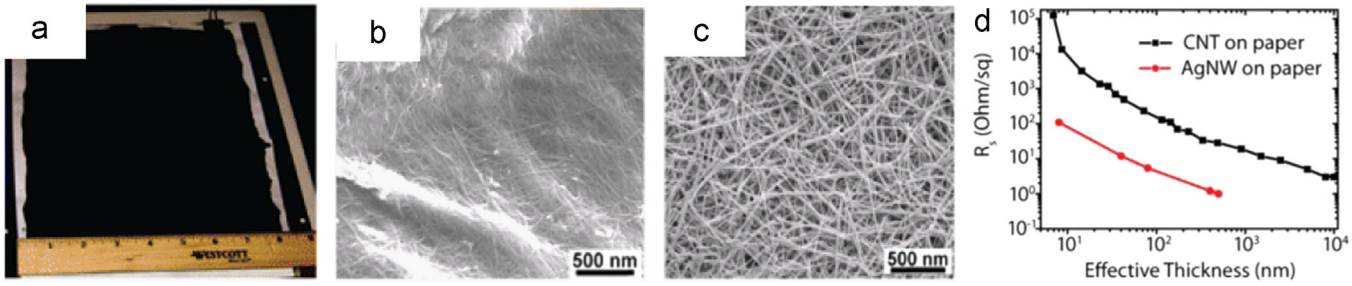

Fig. 2. Depiction of the method for conformal coating of CNT or Ag NW ink on a sheet of paper. (a) Highly conductive Xerox paper after the coating with an average resistance of around 10 Ω sq^{-1}. SEM images of (b and c) the CNT coating along the paper's fibers and Ag NW coating on the Xerox paper, respectively; (d) sheet resistances of the conductive paper after the coating process with either CNT or Ag NW ink with varying thicknesses [68].

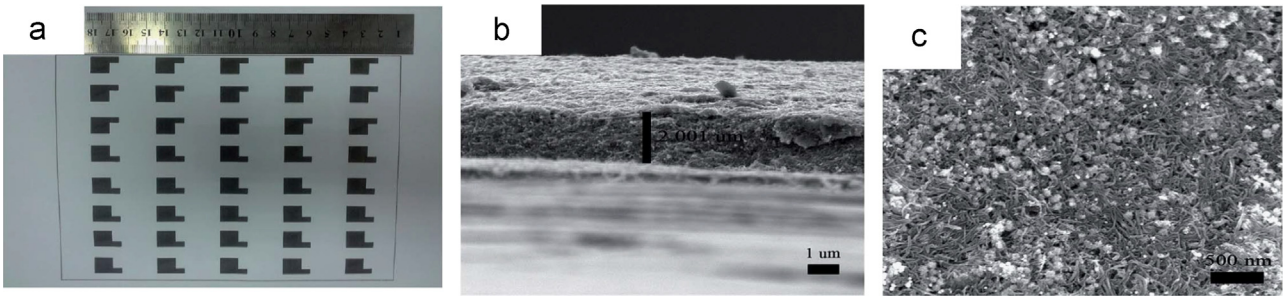

Fig. 3. The fabrication process of inkjet-printing using a personal computer and printer. (a) Printed conductive patterns; (b and c) the side view and morphology of MnO_2–Ag–MWCNT on paper substrates, respectively [22].

resistance considerably [83]. Another solution to enhance the high resistance of the papers with printed CNT is by using transition metal oxides [84–86]. MnO_2 is commonly applied for supercapacitors due to the fact that its theoretical specific capacitance is extremely high (1370 F g^{-1}). Additionally, it is widely available, low-priced, and eco-friendly [87,88]. Wang et al. reported that by adding the combination of Ag nanoparticles and MnO_2 to the CNT, i.e. MnO_2–Ag–MWCNT, the electrochemical performances can be enhanced significantly. Fig. 3

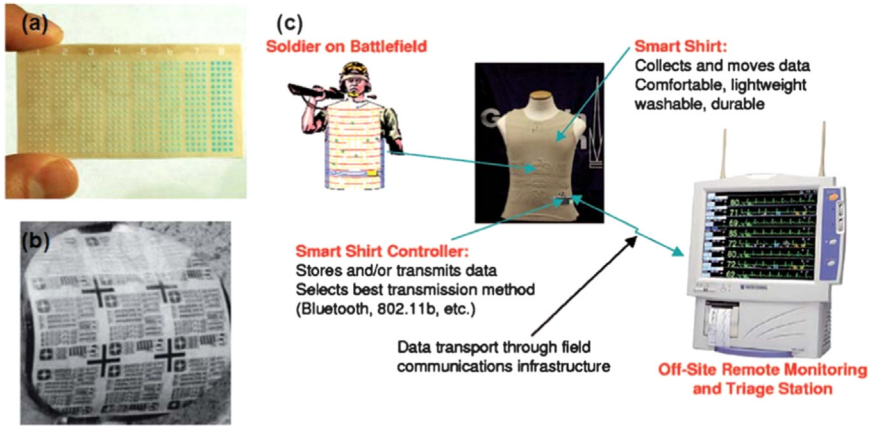

Fig. 4. (a) Three-dimensional paper-based microfluidic device; (b) pentacene TFTs and integrated circuits on paper; (c) developed smart shirt [89].

illustrates the fabrication of the conductive patterns on a paper using a personal computer and a home printer. One of the important variables is the ink concentration. Using ink with high concentration results in clogging and low values increases the printing time. The inkjet-printed conductive patterns are shown in Fig. 3(a). The thickness of the conductive pattern and its morphology are displayed in Fig. 3(b) and (c), respectively [22].

The results showed that incorporating the CNTs with Ag and MnO_2 results in achieving excellent electrochemical performances. For example, the energy density and power density were found to be 1.28 mW h cm^{-3} and 96 mW cm^{-3}, respectively. In addition, after 3000 cycles, the retention ratio was measured as 96.9% of its capacitance [22].

3.2. Flexible energy storage devices

As modern technology develops, the need for an efficient, thin, and flexible energy storage device begins to grow as well [58]. A study performed by the Massachusetts Institute of Technology has shown that it is possible to manufacture a highly flexible energy storage device by incorporating several different components with distinct electrochemical and interfacial characteristics into a single product, composed of nanoporous cellulose paper embedded with aligned carbon nanotube electrodes and electrolytes [59]. The manufacturing process consisted of two important materials, cellulose and CNT, which would provide the necessary innate flexibility and the porosity of the resultant device's system. To overcome the inherent insolubility of cellulose in most conventional solvents, the use of an ionic liquid (RTIL) -butyl, 3-methylimidazolium chloride ([bmIm][Cl]) at room temperature was used. Another benefit that comes with the use of RTIL is its ionic nature, which allows it to be used as an electrolyte, thus permitting the assembly of the electrode, separator, and electrolyte by the means of a simple scalable procedure. The resultant supercapacitor demonstrated a good electrochemical performance, across diverse range of temperatures, electrolytes, and mechanical deformations.

One of the most used techniques to transform paper into a conductor, is comprised of covering the substrate's surface with a chosen metal (Fig. 4). It has to be noted, however, that the metal has to be thick enough (~50 nm) to obtain a decent conductance. In an experiment performed by Cui et al. [89], a highly conductive paper was fabricated by a solution-based printing technique. Amongst the materials used were SWNTs, graphene, and nanoink. The group used solution-based deposition in order to take advantage of paper's efficiency as a printing substrate.

Paper is shown to be a more suitable material for printed electronics mostly due to its porous nature, which results in a smaller application angle of nanoink, which is much smaller than those on plastic-based materials. Nanoink has a higher affinity for paper, and it also does not require the additives needed in order to adjust the ink properties. The SWNT can be jet-printed onto the paper in specific patterns, which is an advantage, as most materials would require conductive pads for such an action. The ability to directly print patterns with jet printers leads to a quick and inexpensive process.

In order to use the developed paper in supercapacitors or in Li-ion batteries, continuous nano-films are needed. The group used a scalable Meyer rod coating method, having the SWNT ink applied onto the paper surface, and a Meyer rod rolled over the ink, transforming the paper into a highly conductive material, with low sheet resistance (about 1–10 Ω sq^{-1}). Such an instantaneous transformation is due to paper's porosity, which allows the rapid absorption of solvents as well as its strong capillary forces, which increases the contact area between the SWNTs and paper fibers. Due to the size difference between the SWNTs and fibers and their strong binding force, the SWNTs increases the conductivity of each individual fiber. The resulting conformal coating led to a highly flexible conductive paper that was capable of being bent to a radius of 2 mm without a change in the sheet's resistance.

Interestingly enough, a technique has been developed by Hu et al. [90] which involves the fabrication of an integrated structure, where the anode, cathode, and separator are all installed into a single sheet of paper. The fabrication method used the continuous Meyer rod coating method and an ink-jet printer for the formation of channels. Since the use of SWNT may sometimes lead to short-circuits, the paper substrate was treated with polyvinylidine fluoride (PVDF), which is an overcoat used on both sides of the porous membrane to fabricate the separator membrane for the Li-ion batteries. The SWNT coating uses paper's rough surface as a means to increase access of the electrolyte to the electrode material.

The fabrication procedure required a Meyer rod coating over the PVDF ink, having the paper dried at 65 °C in an oven for 20 min. The same procedure is then repeated on the other side of the substrate. Even with the coating of PVDF, the paper still allows the transportation of electrons within itself; as such the resulting material can still be used as an electrolyte membrane and separator. Jet printing is then used to print the supercapacitors arrays on the paper sheet. In comparison, using PET substrates with the ink jet printer resulted in lines and defects. The result was a fully integrated, printed supercapacitor device, with the potential for use as a power source for other paper electronic devices [91–93].

Supercapacitors are one of the main energy storage devices with a wide range of applications such as transportation and

general electronics. Compared to batteries, supercapacitors have a longer cycling life, faster charge and discharge rates, and a smaller environmental impact [94,95]. Zhang et al. presented a fabrication technique to manufacture solid-state, flexible, environmentally friendly paper-based supercapacitors [95]. This work was inspired by the noticeable space constraints found in vehicles and portable electronics which would ideally require flexible, light-weight supercapacitors. Normally, solid-polymer electrolytes (SPE) are used for the solid electrolytes. To fabricate the solid supercapacitors, adequate contact with the conducting electrodes is to be ensured for the SPEs. Because of this, researchers usually coat active electrode materials (such as carbon nanotubes) onto the

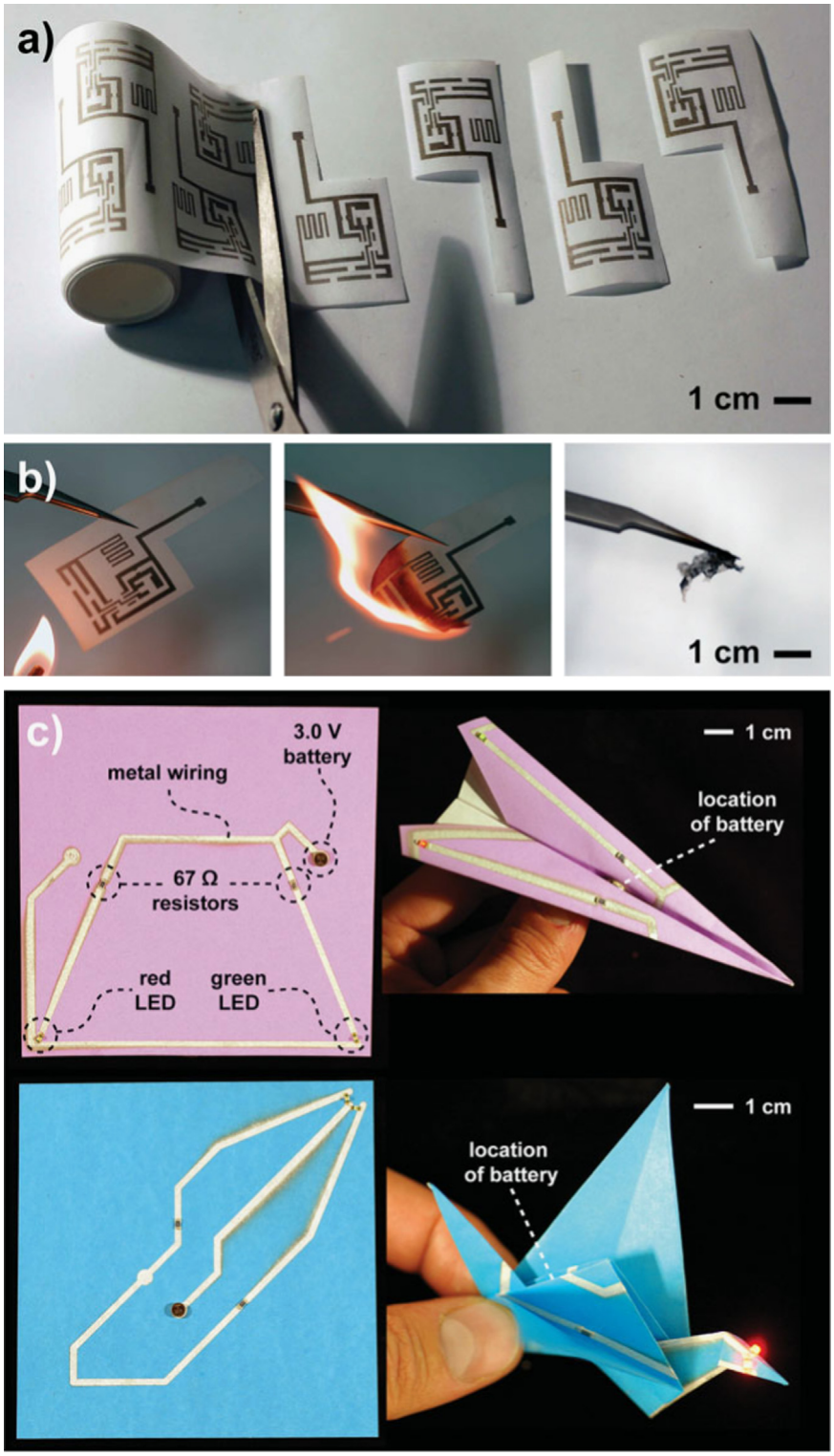

Fig. 5. (a) Cutting and burning fiber-based electronic circuits; (b) three seconds burning of the paper circuit; (c) topology of the electronic circuits demonstrating its capability of becoming a flexible electronic circuit. (Top) A folded paper airplane circuit shown unfolded (left) and folded (right) with battery-powered red/green LED wingtips. The circuit bears a very small weight, less than a gram. (Bottom) A folded paper crane with red LED eyes, shown unfolded and folded [65]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

SPEs. However, the active electrode materials easily peels off, hardly ever form strong bonds with the SPEs, leading to the deterioration of the supercapacitor. To overcome these problems, the researchers used microfribillated cellulose (MFC) as the skeleton material and multi-walled carbon nanotubes (MWCNT) as the electrodes. The MWCNTs were chosen due to their high specific surface area, high conductivity, high electrochemical stability, and low-cost. Due to MWCNT's tendency to cluster in water, the MWCNT was oxidized with –COOH groups which allowed it to easily mix with MFC slurry, resulting in uniform sheets of paper. Additionally, the porous MFC structure provided sufficient passage for ion movement, forming a strong porous sheet to hold the MWCNT and electrolyte in place, while still allowing the ions to diffuse in the electrode sheets. The MWCNT/MFC supercapacitor demonstrated a great advantage in mechanical strength. Its tensile strength and modulus were measured to be 1 MPa and 123 MPa, respectively. As such, its tensile strength and modulus are much lower than that of a pure MFC sheet. The resultant supercapacitors reached up to 154.5 mF cm^{-2} at 20 mV s^{-1} from cyclic voltammetry. Its capacitance is almost unchanged after the supercapacitors are bent to varying different curvatures.

Additional research performed by Yao et al. [96] also strived to develop a low-cost, flexible supercapacitors, introduced by the fabrication of graphite/polyaniline hybrid electrodes on paper for flexible solid super capacitors. Based on the detailed fabrication process developed in the paper, the following procesure will closely reproduce the fabrication results.

PVA powder was added in a sulfuric acid aqueous solution to prepare a polyvinyl alcohol (PVA)/sulfuric acid (H_2SO_4) electrolyte. Then, the mixture was heated and kept at a specific temperature until the solution became clear. Finally, graphite/polyaniline paper electrodes were immersed into the PVA/H_2SO_4 electrolyte and then assembled into a supercapacitor by sandwiching a cellulose membrane as a separator. The hybrid electrode showed a high aerial capacitance of 355.6 mF cm^{-2} and the solid-state supercapacitor was able to achieve a high energy density of 0.32 mW h cm^{-3} at a power density of 0.054 W cm^{-3}, while retaining 83% of its initial capacitance after 10,000 cycles.

An alternative to using normal printing paper as a substrate for the supercapacitors is to use absorbent cotton paper as demonstrated by Hu et al. [97], who developed a solid-state supercapacitor of high energy density by employing porous and absorbent cotton paper coated with single-wall carbon nanotubes using PVA/phosphoric acid as the electrolyte. A solution-based method was used to coat SWNT onto the substrate, by evenly dispersing it into water. The performance of the fabricated supercapacitor was tested by using cyclic voltammetry and constant current charging and discharging using a two-electrode setup. The specific capacitance of the fabricated supercapacitor was 115.83 F g^{-1}, and the specific energy was 48.86 W h kg^{-1}. Compared to previous supercapacitors, with an average of 4–137 F g^{-1} and a maximum specific capacitance of 180 F g^{-1}, the developed supercapacitor is in the same range as the best supercapacitors found in literature. However, most supercapacitors do not have the benefit of being both solid and flexible simultaneously. While the supercapacitor was shown to have a higher performance in terms of capacitance and specific energy when compared to EPCS cell type and is comparable to the ESMA cell type, the equivalent serial resistance (ESR) was shown to be higher than both commercial supercapacitors. This is due to the SWNT-coated paper electrode having a much larger resistance when compared to that of the metal electrodes in commercial supercapacitors.

Ordered mesoporous carbon (OMC) is a promising material for electric double layer capacitators (EDLC) due to the fact that it is cheap, with a high specific surface area. In addition, the ordered pore channel is easily accessible [98–100]. Nevertheless, pristine OMC electrodes have some shortcomings such as poor electrical conductivity, as well as undesirable specific capacity and cycling stability. Some studies have been done to improve the capacitance of OMC using different methods in order to overcome its shortcomings [101–108]. The best electrical conductivity (up to 762 S cm^{-1}), mechanical robustness, and capacitance (213 F g^{-1}) so far was obtained by a novel combination of silver nanowires, graphene paper, and OMC (Ag–GF–OMC), to handle poor electrical conductivity and rigidity of OMC.

3.3. Electronic circuits

While there has been an increase in the development of flexible semiconductor-based integrated circuit technologies, the scientific community has yet to see such effort being placed in regards to inexpensive, flexible printed electronic circuits and hardware. Phillips et al. has reported the creation of a low-cost flexible electronic circuit on paper, a technology which presents a large potential as flexible batteries and biomedical devices [65]. Such printable circuit boards typically use plastic as a substrate, manufactured using a silk-screen printing process, a method most commonly associated with the production of rigid paper circuit boards (PCB). Even though this method is the most conventional procedure, and is quite useful for the inexpensive production of a large amount of circuits, it does present two main disadvantages, limited flexibility of the circuits and expensive costs incurred in low volume production. Paper, on the other hand, presents an easily acquirable substrate alternative, whilst also being an ideal substrate for a flexible device. For the device to maintain its flexibility, use of flexible metals such as tin or zinc is required. Such metals are also rapidly evaporated at low temperatures, therefore, requiring low amounts of energy during the fabrication process (Fig. 5). To pattern the metals onto the paper surface, the group used evaporation and sputter deposition. The chosen adhesive for the electronic components can vary between surface-mountable devices and other electronic components.

Phillips et al. showed that papers with high porosity and/or surface roughness demonstrated a better penetration with metal wires. Another benefit of PCBs is its ability to allow the fabrication of an electrically conductive pathway on both sides of the substrate by: (i) patterning electrically conductive through-holes in the substrate, and (ii) patterning foldable electrically conductive tabs that connect to the paper's other side. Such characteristics make paper-based PCBs great alternatives for powering medical sensing devices in resource limited areas. The paper based circuits could also be folded from angles ranging from -180° to $+180^{\circ}$ and still maintain their function. Nonetheless, the conductance of the wires did decrease in proportion to the angle of folding. For example, -90° curvature presented about 5% decrease, while a $+90^{\circ}$ curvature led to an 11% decrease in conductance.

Paper, like other degradable materials can be used as a platform for conductive patterns [109]. A certain technique which uses inkjet printing, allows for the rapid and low cost fabrication of very conductive patterns onto flexible substrates, while also supporting large area sensors and high frequency applications. Developed by a collaboration between Georgia Institute of Technology, Tokyo University, and Microsoft, this research was capable of developing patterns whose conductivity emerged after only a few seconds. Such a technique uses the progress made in a fabrication method called chemical sintering [110], which removes the need for thermal sintering, which in turn facilitates the use of conductive inkjet printing in laboratory settings.

The true turning point was the development of ink, which supports chemical sintering. Dissolving silver nanoparticles smaller than 0.1 μm in a solvent composed of polymer latex and halide

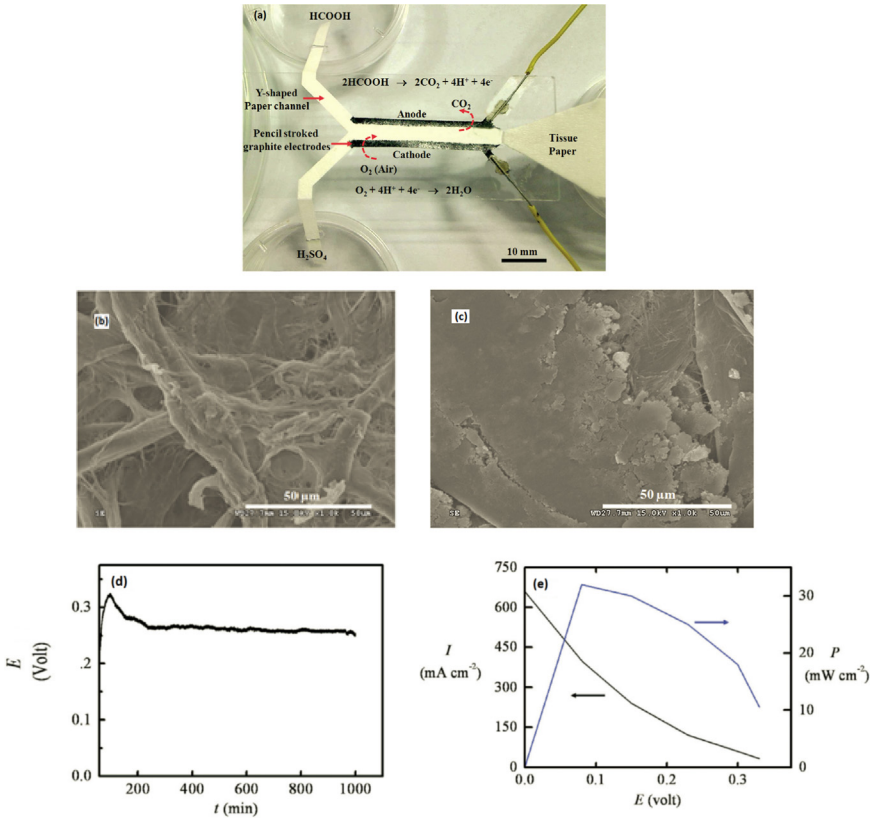

Fig. 6. Paper based self-pumping and self-breathing fuel cell using pencil stroked graphite electrodes (a) a snapshot of the paper based channel. SEM images of the paper; (b) before and (c) after stroking; (d) the open circuit potential and (e) polarization and power curves of a single paper fuel cell [114].

emulsion leads to a conductive substance after the solution is dried. It is believed that this is a direct result of the formation of a 3D structure by the polymer latex and silver nanoparticles and the accelerated formation of interconnections among the silver nanoparticles by the halide. Introducing water to the substance stabilizes these formations. It has been found that silver nanoparticle ink from Mitsubishi Paper Mill, number NBSIJ-MU01, possesses an appropriate dispersing medium for inkjet printers. Such ink can be bought for about ($5 USD) per meter of a 1 mm wide trace.

3.4. Fuel cells

Fuel cells are a promising means for energy generation as their derived power is highly efficient and has low environmental impact. Its efficiency is high due to the fact that fuel cells do not rely upon the intermediate steps of heat and mechanical work production that most conventional power generation methods are associated with [111]. Furthermore, as fuel cells do not depend upon combustion as a source for heat or work, they consequently do not release pollutants into the atmosphere during their energy production.

As such, considering the potential of fuel cells, it is being implemented as a power source for microfluidic devices [112,113]. Arun et al. [114] developed a paper-based fuel cell such that instead of using an external pumping system for micro-fuel cell, a self-pumping method can be applied to increase fuel cell efficiency. For this purpose, they used pencil stroked graphite electrodes to push the fluids in separate streams (Fig. 6(a)). The SEM images of the paper taken before and after repeated pencil stroke are provided in Figs. 6(b) and (c). This figure shows that when the pencil strokes are repeated, the paper fibers can absorb graphite for continuous flat-shaped electrodes. As given in Fig. 6(d), for this system with Y-shaped paper channel, a constant open circuit potential of 0.27 V can be generated for up to 1000 min.

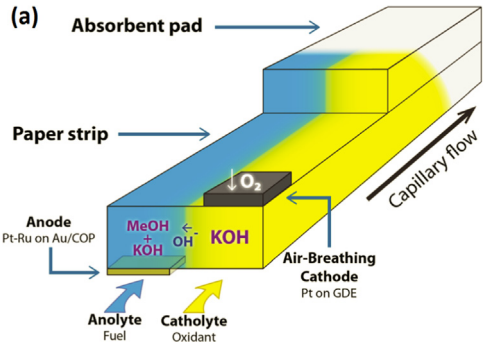

Fig. 7. Schematic of a paper-based microfluidic fuel cell design [66].

The current-potential and power-potential curves of the paper fuel cell are shown in Fig. 6(e). Based on this figure, the maximum power density and current density of 32 mW cm^{-2} and 660 mA cm^{-2} can be obtained via this system.

Additionally, the lateral-flow based microfluidic fuel cells as paper-based on-board energy sources is introduced by Esquivel et al. [66]. For this work, porous membranes are applied to the fuel cell. In this condition, similar to paper-based fuel cell [114,115], the need of external pumps is eliminated since reactants flow by capillary forces. A schematic of the reaction zone for this working microfluidic fuel cell is provided in Fig. 7.

The results demonstrate that the open circuit voltage for different methanol concentrations is 520 ± 60 mV. The maximum power density of 3.2 mW cm^{-2} can be obtained at a current density of 15.5 mA cm^{-2} in methanol concentration of 4.0 M. Moreover, with changing the KOH concentration, a similar

behavior can be observed. This time, the maximum power density can be raised to 4.4 mW cm^{-2}.

Another example of such a use is the manufacturing of a microfluidic H_2–O_2 fuel cell led by Mitrovski et al. [116] composed of electrodes immersed in liquid electrolytes with the fuels being supplied through a gas-permeable membrane. Following the design of conventional microfluidic devices, the constructed fuel cell is passive, as it neither requires pumps to supply fuel nor to circulate the electrolytes throughout the electrodes. The materials used were two 1.23 mm^2 platinum electrodes, separated by a 1 cm gap, and integrated within a gas-exchanged-membrane sealed whithin PDMS microfluidic channels (Fig. 8). The array of electrodes was composed of 0.1 μm thin films imbedded in quartz, with the rest of the device being comprised of PDMS.

Galvanic cells have been used in such a way that μPADs are capable of powering themselves with the simple addition of a sample [21], fundamentally adjusting a sample to both conduct an assay and power the device for such a task. Thom et al. [21] designed a μPAD with layered paper and tape, the method used to pattern the materials was one similar to the procedure performed by Noh et al. [43], bearing a difference in the amount of time used to bake the paper (105 s as opposed to the suggested 120) after printing out the wax patterns. The fabrication led to the construction of a device composed of several variants of fuel cells with batteries of up to twenty-four cells. Fig. 9(a) illustrates the expanded view of the device, which is 14 mm wide × 8 mm long. The salt bridge is located in the second layer and the electrolytes in the fourth. Aluminum and silver pieces are located in the fifth layer, covered by copper tape. The device's performance was tested by the connection of a multimeter to the pieces of tape, followed by the insertion of 5 μL of distilled water into Layer 1. The expanded view of the two cell battery is shown in Fig. 9(b), which is 16 mm wide × 7.8 mm long. The salt bridges are located in the fourth layer and the electrolytes in the sixth. Aluminum and silver were placed in the seventh layer, covered by the copper tape, which served as an electrode and as a connection between both cells. The design was tested in a similar way to the single cell battery [21].

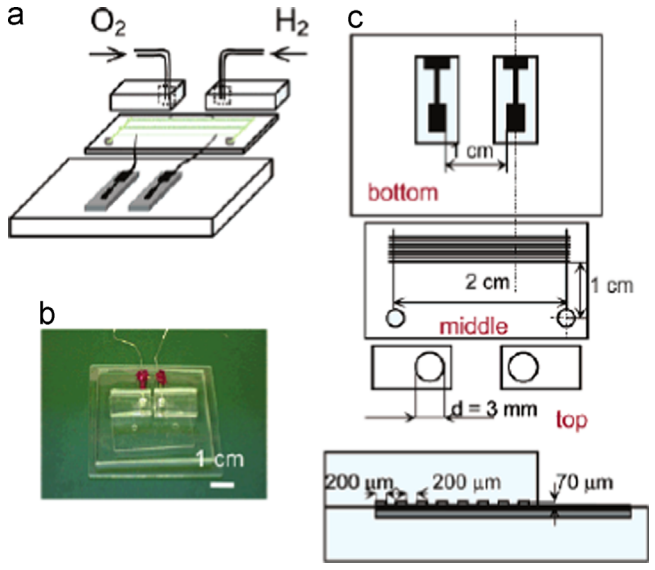

Fig. 8. Depiction of the top, middle, and bottom layers of the microfluidic device, the cell's dimensions, and its order of assembly (a) the fuel cell's general design; (b) photograph of the device (c) top of the cell's PDMS layers and a cross sectional view along the dashed line in between the bottom and middle layers [116].

The use of galvanic cells as a method to store energy for microfluidic devices was one of the starting points for the study of μPADs in the energy field. As demonstrated, the use of the so called fluidic batteries is an efficient way to store energy, while also exemplifying how the correct use of electrolytes is able to extend the device's lifetime, further enhancing its use as energy storage medium.

In this design, the galvanic cells are installed directly into the microfluidic channels, thus establishing a direct link between the device's power source and analytical function, allowing the sample to aid the cell to both power and conduct the assay. The use of galvanic cells also allows for the reduction of the batteries' fabrication price and the reduction of the hazardous wastes associated with the disposal of the microanalytical device. Galvanic cells can have their potentials and currents tuned accordingly and the device's electrolytes can be adjusted in order to use the minimum quantity necessary for a particular application. Such a device would be particularly well suited for extremely resource limited areas as the batteries are able to generate power by simply adding samples for a designated assay, turning on simultaneously. The number and configuration of the galvanic cells in relation to one another could also be easily and rapidly manipulated, making the device pretty malleable in terms of its use.

Such fluidic batteries require four basic components in order to be used: electrolytes (which have to be integrated into the hydrophilic regions of the device prior to its construction), electrodes (integrated

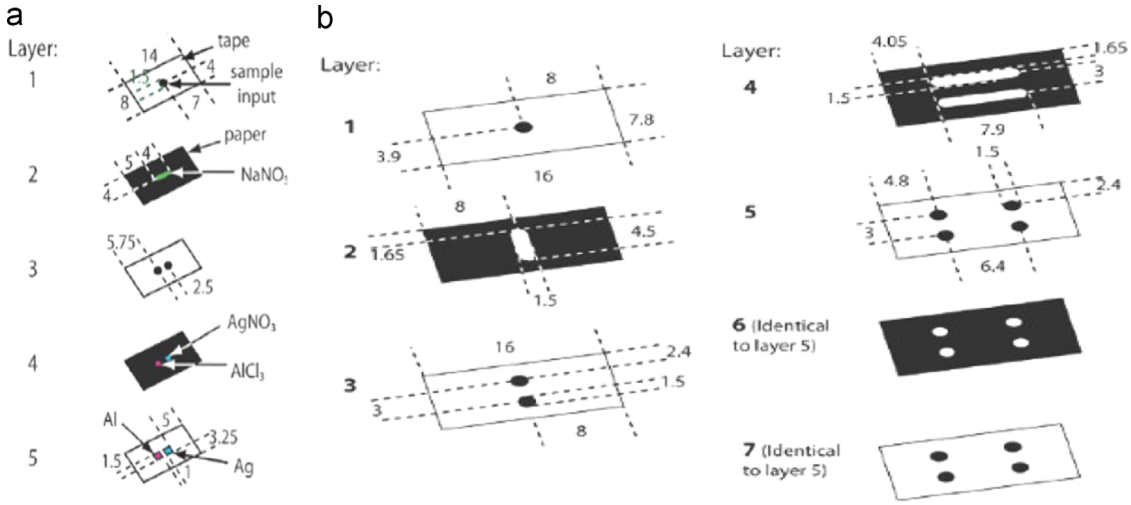

Fig. 9. The design of fluidic batteries with layered paper and tape. (a) Single cell battery; (b) two cell battery [21].

into the air gaps of the patterned tape during the device's construction), salt bridges (its quantity varies according to the number of galvanic cells), and conductive connections (in this case copper tape) linking the galvanic cell to the device. The chosen electrolytes were silver nitrate and aluminum chloride, which were deposited in a 3:1 ratio of Ag–Al per cell. The chosen electrodes were composed of either aluminum or silver, with either a square shape (1.5 mm × 1.5 mm × 250 μm thick), or with other geometrical designs according to the cell's configuration. The salt bridges containing sodium nitrate were pre-deposited from a saturated aqueous solution and then dried in the appropriate hydrophilic paper layer prior to the device's fabrication. In a test scenario, the device did not present a loss of performance even after being stored for at least five weeks in open air.

Paper-based microanalytical devices have emerged as a promising mechanism for the conduction of diagnostic assays in resource limited environments. However, such devices sometimes require a power source for their functionality and the use of fluidic batteries have been shown to be a common and pretty efficient method for power generation in μPADs, as they are not expensive, nor do they pose a hazard during their disposal. Thom et al. have shown that the architectural design of μPADs can be manipulated in order to directly incorporate multiple galvanic cells. The cells can be arranged in either parallel or series in such a manner that it is possible to obtain the desired potential and current, while the possibility of optimizing the batteries to function for limited amounts of time (10 to 15 min), prevents the unneeded loss of energy.

Paper-based microanalytical devices usually perform assays which require some sort of powering such as fluorescent, electrochemical [117], and electrochemiluminescent readouts [118]. In such cases, the use of some sort of batteries are needed, either as a part of the procedure itself, or just to run the reader which measures the output of the designated assays [119]. The same authors have previously described a paper-based microfluidic battery which comprised of galvanic cells [120] as a power source. The integration of the battery in the device was performed by the use of a co-fabrication process, where the batteries and microfluidic channels are constructed simultaneously using similar fabrication methods. The resulting battery was tested with a device performing an on-chip fluorescence assay. According to the device's structure, the selected sample would split up as it reached the microfluidic channels, where each channel was responsible for performing certain tasks: the first turned on the battery, whereas the second was responsible to lead the sample to the assay reagents, the region where the assay would be performed. The results were satisfactory, as the device successfully powered a UV LED within the μPAD.

Additionally, there is another type of microfluidic fuel cell, known as the air-breathing microfluidic fuel cell, that utilizes gas diffusion as an air-breathing cathode [121]. Jayashree et al. proposed the air-breathing microfluidic fuel cell for the first time [122]. As displayed in Fig. 10, the blank electrolyte stream (oxidant) is used in order to prevent the direct contact of the fuel stream to the cathode [20]. Because the concentration and diffusion coefficient of oxygen in air is considerably higher than those of the aqueous media, the power density of the microfluidic fuel cell can be improved significantly [122–124]. In addition, due to the fact that air-breathing microfluidic fuel cells do not have membranes, a wide range of oxidants and fuels can be applied [124,125]. Shaegh et al. [20] could obtain the maximum power densities of 26.5 mW cm^{-2} and 19.4 mW cm^{-2} for the flow-through and flow-over anodes, respectively after running the experiment with different concentrations of formic acid and flow rates. However, this device has one important issue that needs to be optimized in order to decrease its adverse effects. In fact, the increase of the flow rate, reduces the residence time of the fuel in the channel. As a result, the power density increases, but the fuel utilization decreases. In order to handle this problem and increase the fuel utilization, several methods have been used such as hydrodynamic focusing or chaotic mixing generation through the channel [126–129]. Moreover, the air-microfluidic systems can be applied as a micro electro mechanical systems (MEMS) sensor to measure the atmospheric particulate matter (PM) with high sensitivity of 2 μg m^{-3} or higher. The application of this device is to measure the concentration of tobacco smoke and diesel exhaust which are two important PM sources. Because the size of this device is small, it can be connected to a cellphone and one can find the PM concentration in different places. As a result, the health risk would be improved and there would be a trend for the energy systems, working with fossil fuels, to expose less PM in the air [130].

Apart from the experimental work, some numerical simulations have been done in this area [131,132]. Xuan et al. could properly predict the effects of the involved parameters on the electrochemical activities of air-based microfluidic fuel cells [131].

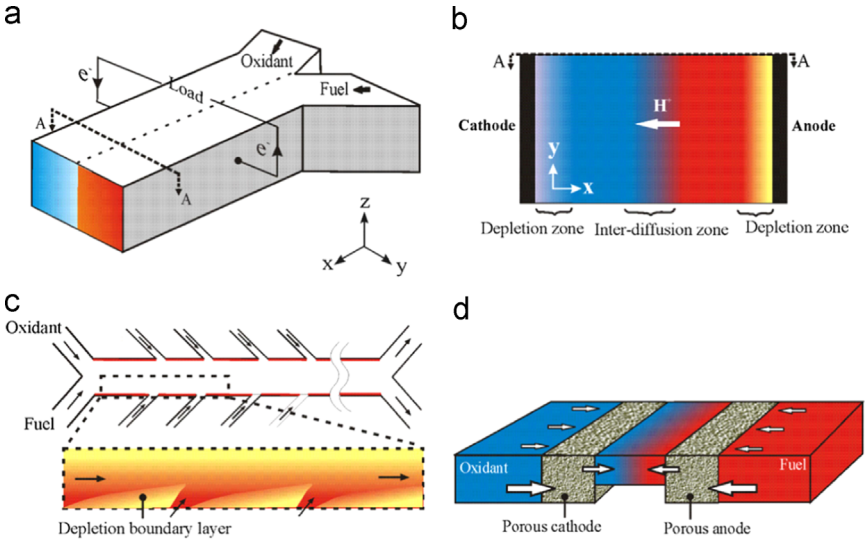

Fig. 10. Membraneless air-breathing microfluidic fuel cell, (a) a schematic of the device in a Y-shape channel; (b) cross section of the channel; (c) the process of adding fresh reactant into multiple inlets; (d) cross flow through the electrode [20].

They found that the electrode kinetics can play a significant role in the relationship between fuel usage and the current density.

3.5. Li-ion batteries

Leijonmarck et al. developed a method to create flexible batteries integrated into paper, demonstrating an innovative procedure that helps to suppress the increasing need for mechanically flexible batteries [133]. The paper-based Li-ion batteries were manufactured using an aqueous paper-making procedure. The nano-fibrillated cellulose was developed from a TEMPO-oxidized, dissolving pulp from Domsjö Fabriker AB.

The battery cells (2–3 cm^2) were fabricated as pouch cells under an argon atmosphere in a glove box. The batteries were soaked in an electrolyte (as chosen by the authors, 1 M LiPF6 in ethylene carbonate (EC):diethyl carbonate (DEC) 1:1 by weight with an addition of 2 wt% vinylene carbonate (VC)) for 5 min under a vacuum and then inserted between the current collectors. The negative side of the papers was placed to face the copper and the positive side in front of the aluminum. The developed method allowed complete control over the active material loadings of the electrodes due to the controlled additions from the respective water dispersion.

Two-dimensional graphene demonstrates fantastic electrical and mechanical properties due to its high specific surface area (2630 $m^2 g^{-1}$), high electrical conductivity, and good flexibility, leading it to be an invaluable tool in energy research [134]. However, due to the strong Van der Waals attraction between individual graphene layers, a restacking of the graphene occurs, harming its potential in the energy sector. To overcome this, Hu et al. group reported an approach to manufacture flexible graphene–TiO_2 hybrid papers with a high mass loading of oxides [134]. To do so, TiO_2 was loaded into the wet graphene papers after the development by filtration of the well-dispersion graphene solution.

The unique properties of the hybrid graphene composite paper combined with the uniformly distributed TiO_2 nanospheres in the 3D open space demonstrates several advantages when used as paper electrodes for LIBs. The high conductivity of the graphene network generates an efficient charge transfer, and the sound structure of the device leads to a reliable and high rate of performance. This approach can also be applied to the fabrication of other metal oxide-graphene papers for the flexible energy field.

As mentioned previously, an important aspect of high-rate performances in energy storage devices is the effective manipulation of electrons and ions. One area that has been receiving an increased amount of attention is energy storage using aerogels, which demonstrate the potential to be an excellent platform for fast ion transportation. Nonetheless, such materials have not been used with Li-ion batteries. As such, in this part we will explore the work of Cui et al. [135] who developed conductive CNT-cellulose aerogels coated with SI by a plasma-enhanced CVD (PECVD) method. One of the advantages is that such nanocomposites were identified as flexible anodes for Li-ion batteries. Due to the low-cost materials and scalable fabrication process, this procedure could also be advantageous for large-scale energy storage.

The first step is the fabrication of NFC:CNT dispersion. The NFC:CNT slurry was dewatered in order to reach a desired level of porosity, by the use of a vacuum filtration (on a 0.1δ μm mixed cellulose ester (MCE) membrane filter using a PG90 glass filter setup for 2 h at room temperature). The generated hydrogel is then transferred to an aluminum mold and then dipped into liquid nitrogen for 2 min. The sample is then placed in a freeze-dryer at −50 C and 50 μbar for 24 h to provide proper drying. The resulting aerogel is then heat-treated for 30 min at 140 C using a vacuum oven (to improve web strength).

The paper sheets were manufactured by a semi-automatic sheet former, followed by the application of CNT onto the paper. Aqueous CNT ink of 1 mg mL^{-1} in 1 w/w% sodium dodecyl benzene sulphonate (SDBS) in Dl water was used as the coating material. CNT ink was then applied onto the paper by using Meyer rod coating method followed by drying in a vacuum at 100 °C for 10 min. The resulting nanostructure demonstrated a better performance than Si thin films on regular paper, bearing a better cycle stability and capacity retention.

3.6. Alkaline batteries

Alkaline batteries primarily generate energy by a reaction between zink and manganese dioxide (Zn/MnO_2). Alkaline zink–MnO_2 batteries employ macro porous nonwoven separators made from cellulose based fibers, and unfortunately, any attempt to improve separators in this area have been mostly unsuccessful in the past [136].

Gallo et al. [137] reported the comparisons between different cellulosic fiber properties and their influence on the battery separators performance. Several cellulosic fibers were considered, amongst these were: TENCEL, Viscode and Pulp. These were blended with synthetic PVA fibers and water soluble PVA binders. The basic properties of a suitable separator include dimensional stability (good mechanical and chemical resistance), a balanced electron insulation and ion conductivity. In terms of maintaining a decent shelf life, the separator has to have a high dimensional stability and low deterioration in alkali. It has testable characteristics using the following tests: “Area Shrinkage in KOH”(linked to dimensional stability) and “Weight Reduction Rate in KOH” (linked to deterioration resistance). As defined by Gallo et al., area shrinkage rate is measured by the ratio of a square sheet 120 mm × 120 mm (A1), and the area of wet sample (A2) Sheet immersed in 40% KOH solution at 70 C for 8 h. The weight reduction rate is measured by the ratio of a piece of separator (W1); dried sample at 80 °C for 1 h; and W2 (the dried sample immersed in 40% KOH solution at 70 °C for 8 h, washed with water, and dried at 80 °C for 1 h). For samples made with cellulosic fibers in varying proportions and 0% TENCEL, the shrinkage of the paper is above 8%. However, adding TENCEL into the blend dramatically reduces the paper shrinkage with 50% the paper shrinkage lowered to less than 4%. This is due to the long fibers of TENCEL which clasp tightly during the paper fabrication and eventually “lock” together with the paper structure. The weight reduction rate of the paper varied depending on the cellulosic fiber used, with the largest result occurring in a eucalyptus pulp/viscose mixture, and the smallest in a cotton linter pulp mixture. The TENCEL brings superior properties to the alkaline battery separators.

3.7. Nanotechnology

Nanotechnology plays significant roles in various sectors of economy, healthcare, transportation, agriculture, and energy. Additionally, it has shown high potential to boost the environment directly and indirectly. In other words, we can directly detect, avoid, and eliminate pollutants as well as indirectly create cleaner materials and methods in order to achieve environmentally friendly products [138].

Although previous sections include many studies in the area of nanotechnology in different energy sectors, more research efforts are reviewed here due to the inevitable role of the nanotechnology in paper-based devices for energy applications.

In a recent study, a highly conductive material was applied to improve the performance of pseudocapacitors, a type of electrochemical capacitor that stores electrical energy Faradaically. These

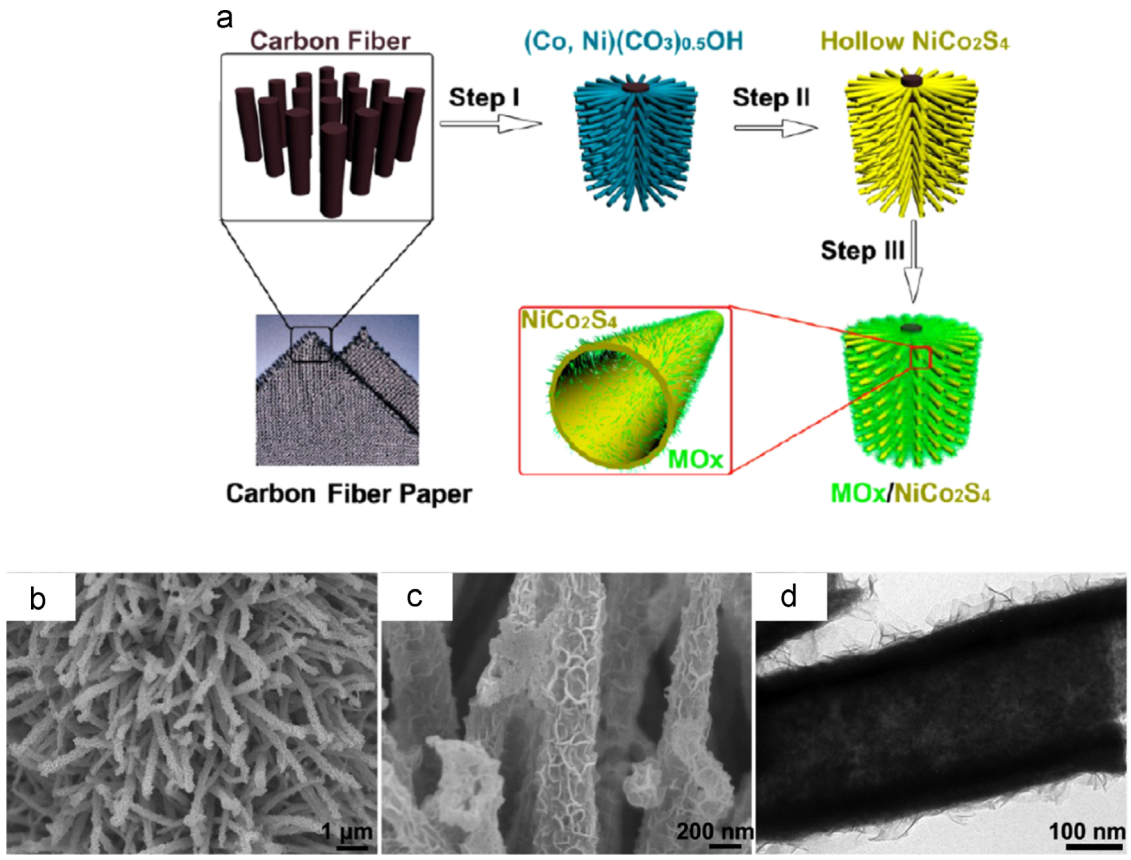

Fig. 11. (a) A schematic of the process of adding the highly conductive transition metal sulfide ($NiCo_2S_4$); (b–d) SEM and TEM of $Co_xNi_{1-x}(OH)_2/NiCo_2S_4$ composites grown on the flexible carbon fiber paper (CFP) [139].

kind of capacitors provide higher specific capacitance due to the fact that the Faradaic reaction happens at or near the electrode surface. In addition, they have low cost, low toxicity, and high flexibility. Nevertheless, they are not capable of supporting the fast electron transport due to their low conductivity. In this report, the pseudocapacitor is made of flexible carbon fiber paper (CFP) and a transition metal sulfide ($NiCo_2S_4$), with high conductivity, and was grown on the CFP. Then, this product is coated with electroactive materials ($Co_xNi_{1-x}(OH)_2$, MnO_2, or FeOOH) which is schematically displayed in Fig. 11(a). The results show that the highest specific capacitance of 2.86 F cm^{-2} with excellent cycling stability of a 4% loss after 2000 cycles can be achieved using $Co_xNi_{1-x}(OH)_2/NiCo_2S_4$ nanotube array electrodes. The scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of $Co_xNi_{1-x}(OH)_2/NiCo_2S_4$ nanotube are shown in Fig. 11(b–d) [139]. There are some other studies about the important role of $NiCo_2S_4$ on pseudocapacitor [140–142].

Moreover, the low conductivity of the paper-based supercapacitors can be improved using: transition metal oxides, e.g., Co $(OH)_2$; Co_3O_4; $CoMoO_4$; $MnMoO_4$; MnO_2; and $Ni(OH)_2$ [143–147], metal nitride, e.g., VN and TiN [148,149], and conducting polymers, e.g., polyaniline (PANI) and Polypyrrole (PPy) [6,17,144]. Li et al., for instance, acheived highly conductive free-standing membrance using the polymers of bacterial cellulose (BC) and polypyrrole (PPy) (BC/PPy) in order to be applied as supercapacitor electrodes [6]. The maximum capacitance obtained using these polymers was 2.43 F cm^{-2} at a discharge current of 2 mA cm^{-2} and a mass loading of 11.23 mg cm^{-2}.

Nanogenerators (NGs) are another group of devices that can convert the mechanical energy into electricity with different mechanisms compared to the conventional generators. The performance of piezoelectric NGs, for instance, is dependent on the piezoelectric effect of nano structure [3,150,151]. They are very sensitive to external strain, and their output power can be found based on the piezoelectric coefficient and external strain [152,153]. Liao et al. [3] fabricated a flexible hybrid NG using ZnO NWs on carbon fibers at the center and Au-coated ZnO NWs on paper. They could achieve a flexible, wearable, robust, and foldable source with desired shapes and an output current that can be improved significantly by increasing the number of fibers. The application of these NGs can be in wearable systems, self-powered nano/micro devices, and implantable devices.

4. Summary table

The table summarizes different work in the area of paper-based devices. It is categorized based on different applications of the devices in their energy sector followed by their fabrication method, specific capacity, power density, advantages, and limitations. Table 1.

5. Concluding remarks

The world is in need of greater investments in energy sources as the average energy consumption is predicted to increase significantly. Developing regions, in turn, are also predicted to increase by 5% annually, making their access to suitable energy sources even more problematic. Microfluidics are shown to have potential in the energy sector as demonstrated by recent developments that have occurred in microfluidic biofuel cells [156–160] and in light-driven microfluidics

Table 1

	Authors, reference	Fabrication method	Specific capacity [F g^{-1}]	Power density [mW cm^{-2}]	Advantages	Limitations
Paper as a highly conductive material	Hu et al. [90]	Printing carbon nanotube films with Meyer rode coating or ink-jet printing onto paper	33	–	• Extremely simple structure • Scalable • The device can be used to power other paper electronic devices • Light weight • Method allows for high speed printing • Method prevents the short-circuiting of the device with the addition of SWNT	• A possible concern is the chemical stability of conductive paper in electrolytes. Currently the paper was shown to be stable in the electrolyte for a few months, somewhat limiting its shelf life • The specific capacitance of the assembled paper supercapacitor decreases with the current density
	Hu et al. [68]	Conformal coating of single-walled carbon nanotubes (CNT) and silver nanowire films on paper	200	–	• Simple and scalable coating procedure • The device can be used as a lightweight current collector in lithium-ion batteries • Light weight	
	Karthika et al. [154]	Using polyester cellulose paper coated with carbon nanotubes	270	–	• Mechanical flexibility which allows for changes in shape depending on the device requirements • Good film-forming capability • Lightweight • High energy density • This device can be rolled, twisted to any curvature, and returned to its original shape, but the same capacitance can be retained • High degree of electrolyte absorption and increased access to electrode material for the electrolyte due to rough surface of cellulose fibers • Extremely large specific surface area for the electrode/electrolyte interface due to its porous structure • No significant difference in the impedance when the device is twisted • Low interfacial resistance	
	Sawangphruk et al. [155]	Coating Ag- polyaniline (PANI)-graphene on flexible carbon fiber paper	520 and 830	–	• Obtaining ultrahigh specific capacity is feasible • Nanocomposite of AgNP–PANI–grapheme can be used in supercapacitators • Flexible	
	Wang et al. [22]	Inkjet printing of highly conductive patterns of MnO_2–Ag–MWCNTs	50.1	96	• Good stability • Low sheet resistance (300 Ω sq^{-1}) • Wide operating potential window • High retention ratio • Sheet resistance does not change significantly after 20% curvature	• High resistance of printed CNTs on a flexible surface • Clogging possibility in the nozzle
Paper-based fuel cells	**Authors, reference**	**Fabrication method**	**Specific capacity [F g^{-1}]**	**Power density [mW cm^{-2}]**	**Advantages**	**Limitations**
	Esquivel et al. [66]	Using lateral flow test strips to develop the self-pumping paper-based microfluidic fuel cell	–	1–5	• No need for external pumps due to use of porous membranes • No need for ionic exchange membrane • Light-weight • Straightforward and cost-	• Depending on how the device is fabricated, placing several sheets of paper in certain areas of the device diminishes the light of the LED, making it harder to see the results of the assay

Table 1 (*continued*)

	Authors, reference	Fabrication method	Specific capacity [F g^{-1}]	Power density [mW cm^{-2}]	Advantages	Limitations
					effective integration due to utilization of the lateral flow test strips • Minimum environmental impact • Portable • All of the produced power is available for use	• When operating continuously for 3 h at its maximum power, the cell demonstrates a depletion of about 75% of its output. The cell also demonstrates significant diffusive mixing of gases within the electrolyte
	Arun et al. [114]	Using Hb-pencil stroked graphite as a porous electrode on paper to make a self-pumping microfluidic fuel cell	–	32	• No need for external pumps due to application of pencil stroked graphite electrodes • No need for membrane • Cost-effective • Easy electrode preparation • Easy to design • Overcome oxygen delivery issues • The ability to generate power for more than two days without any loss • Overall dimension becomes smaller by eliminating the external pumping system • Scalable design • Very low formic acid consumption	
	Thom et al. [21]	Integrating galvanic cells into the microfluidic channels	–	–	• Capable of generating its own power • Provides sufficient power to illuminate a UV LED • Useful for powering electrochemical detection, separation schemes, and optical readout for assays • Cost of battery is minimized • Less hazardous waste associated with disposing of used paper-based microfluidic device • Minimum quantity of metal electrodes and electrolytes are used	
	Shaegh et al. [20]	Air-breathing laminar flow-based fuel cell	–	26.5	• High concentration and diffusion coefficient of oxygen in air improve the power density • High efficiency	• Fuel utilization reduces by increasing the power density • It is difficult to remove the bubbles from the active site
Flexible dnergy storages	**Authors, reference**	**Fabrication method**	**Specific capacity [F g^{-1}]**	**Power density [kW kg^{-1}]**	**Advantages**	**Limitations**
	Pushparaj et al. [59]	Integrating electrode, separator, and electrolyte into single contiguous nan-composite units	22–36	1.5	• Easy to assemble integrated nanocomposite energy-storage system • Works on wide range of temperatures and environmental conditions • High degree of cyclability since the charging and discharging involve no chemical phase and composition changes • Ability to function over large ranges of mechanical deformation • Cost-effective energy storage device • Occupies minimum space • Adapts to accurate shape	• The use of batteries may present disposal hindrances • It still requires more studies to optimize the equivalent serial resistance of the supercapacitor to achieve a more adequate power density • No study was made on how the mechanical capabilities of the device would be affected after bending the device to an able larger than 90° • Flexibility of the device significantly decreases by adding Ag nanowires which is not proper for flexible devices

Table 1 (*continued*)

Authors, reference	Fabrication method	Specific capacity [F g^{-1}]	Power density [mW cm^{-2}]	Advantages	Limitations
Zhang et al. [95]	Microfibrillated cellulose (MFC) as the skeleton materials and MWCNT as the electrodes of the supercapacitor- MWCNT oxidation with –COOH groups allows mixing with the MFC slurry	154.5 mF cm^{-2}	–	• Environmentally friendly • The paper-based supercapacitors have excellent mechanical properties compared to the conventional liquid or gel based soft supercapacitors • Higher specific capacitance can be obtained by making thicker sheets with more electrode materials • Excellent mechanical strength for the electrode sheet can be achieved • Long cycling life • High power density • Fast charge and discharge rate • Lightweight • Easy packaging	
Hu et al. [97]	Applying SWNT-coated cotton paper as electrodes and PVA/ phosphoric acid as electrolyte	115.83	–	• Performs as well as supercapacitors on the market • Solid-state configuration • High energy density • Appropriate for applications such as hybrid electric vehicle and portable electronics	
Yao et al. [96]	• Using Polyvinyl alcohol and sulfuric acid as an electrolyte solution • Graphite electrodes immersed into the electrolyte and then assembled into a supercapacitor by sandwiching a cellulose membrane as a separator between them	355.6 mF cm^{-2}	0.054 W cm^{-3}	• Environmentally benign • Easy to scale up • This method can be applied to fabricate planar solid-state supercapacitors • Portable and wearable	
Hu et al. [134]	Loading TiO_2 into the wet graphene papers to fabricate highly flexible graphene-TiO_2 hybrid papers	122–200 mA h g^{-1}	–	• High capacity • High mass loading • Excellent electrochemical performance • Excellent cyclic stability	
Zhi et al. [107]	Combination of silver nanowires, graphene paper, and OMC (Ag–GF–OMC)	213	5.04	• High electrical conductivity (up to 762 S cm^{-1}) • High mechanical robustness • Long-term stability (9% capacitance decay over 10,000 charge/discharge cycles) • The transport of the electrons in the bulk electrode can be easier using the graphene paper • No considerable difference between current- potential (CV) curve with and without bending • Wearable	
Xiao et al. [139]	Growing $NiCo_2S_4$ on CFP and coat it with different electroactive materials	2.86 F cm^{-2}		• High surface area sue to creation of hollow structure • Fast electron conduction and ion diffusion • High cycling life • High flexibility • Low toxicity	• The conductivity of the typical pseudocapacitors is low
Li [6]		216	–	• No need to binders	

Table 1 (*continued*)

	Authors, reference	Fabrication method	Specific capacity [F g^{-1}]	Power density [mW cm^{-2}]	Advantages	Limitations
		Combination of BC/PPy and MWCNT in ordre to increase the efficiency of supercapacitor			• Excellent cycling stability • It is possible that the membrane is directly cut into desired shapes • It can be simply connected in series or parallel	
Printable circuit boards	**Authors, reference**	**Fabrication method**	**Specific capacity [F g^{-1}]**	**Power density [kW kg^{-1}]**	**Advantages**	**Limitations**
	Siegel et al. [65]	Circuit boards printed on paper	33	250	Inexpensively and easily manufactured Paper-based electronic circuits are thin and lightweight Useful for applications in consumer electronics and packaging Applicable in the military and homeland security Useful for paper-based microelectromechanical systems in medical sensing or low-cost portable diagnostics Device can be used for applications related to textiles The fabrication procedure also develops electrically conductive pathways on both sides of the substrate. The result is an alternative to the more common rigid printable circuit boards.	• To maintain its flexibility it requires the use of other flexible conductors, limiting the variety of suitable metals. The wires present some mechanical/electrical fatigue when the device is folded
Paper in Li-ion batteries	**Authors, reference**	**Fabrication method**	**Specific capacity [F g^{-1}]**	**Power density [kW kg^{-1}]**	**Advantages**	**Limitations**
	Hu et al. [135]	Conductive CNT-cellulose aerogels coated with Si (using plasma-enhanced CVD (PECVD) method)	1200 mA h g^{-1}	–	• Lightweight • Earth-abundant materials (Si and Nanocellulose) are used • Scalable processes • Excellent electrical conductivity of the nanocomposite • By Substituting CNT with graphene, the cost of the conductive nanopaper could be decreased • The device demonstrates a better cycle stability when compared to normal Si strips on paper	• Regular paper does not demonstrate a cycle stability as good as the SI-conductive paper, as its capacity decays rapidly
Paper in alkaline batteries	**Authors, reference**	**Fabrication method**	**Specific capacity [F g^{-1}]**	**Power density [kW kg^{-1}]**	**Advantages**	**Limitations**
	Gallo [137]	Cellulosic fibers such as TENCEL, Viscode, and Pulp blended with synthetic PVA fibers and water soluble PVA binders	–	–	• Using TENCEL® as battery separators provides appropriate porosity and great dimensional stability	• The effects of using TENCEL® on the power and energy density of battery is not clear

[161] (a device based on solar-power) which present an ingenious method for alternative energy generation. Nonetheless, paper-based microfluidics are currently most often used to power small devices, and few are actually used for larger scale energy generation. This limits their influence in the energy sector. However, μPADs should not be underestimated as they do demonstrate possibilities in large-scale energy storage devices, primarily though hybrid combinations of μPADs and other technologies. The conversion of paper into a highly conductive material [39], for example, does demonstrate how its combination with metal nanowire strips as global current collectors can be used in large-scale energy devices. Taking advantage of paper's natural characteristics, such as its high solvent absorption and strong bindings with nanomaterials, allow for easy and scalable coating procedures, while its use with other solution-processed nanoscale materials could lead to new developments in advanced energy storage and conversion applications.

Paper's flexibility does present several new opportunities in the energy sector. Although these devices present several novel

benefits, they do present some disadvantages, which would make their manufacturing more difficult in more resource-deficient areas. Microfluidic biofuel cells, for example, often require meticulously manufactured microelectrodes [157], while also demonstrating a relatively lower life span and power density when compared to galvanic cells used in μPADs. Solar-power based devices require specific technical equipment and knowledge, which hinders their mass production. Paper-based microfluidics present simpler, low-cost fabrication methods for energy-based devices, while also using a widely available substrate. Nonetheless, paper-based microfluidics is still in its infant phase, requiring the evolution of its capabilities to fully develop into a novel and essentially practical device [56], as they are still primarily capable of powering small devices. Paper-based microfluidics' varying range of applications and fabrication methods do present a startling amount of flexibility to its users' needs.

There are several different methods to fabricate μPADs, each with their own benefits and hindrances. Wax printing is one of the least complicated approaches due to its quick and easy use, which also aids its viability as a fabrication method. This makes it well suited for mass production in developing economies. As demonstrated in Table 1, the fabrication methods do require a varying amount of technical expertise and materials, which in turn leads to a varying amount of difficulty for the implementation of that specific device. Some interesting ways to fabricate paper-based microfluidics are by using customized fabrication methods which are usually developed by implementing modifications to the more "traditional" patterning methods (such as wax printing and plasma etching) to better suit a users' specific needs [36,162–165]. Amongst these are 3D origami paper-based analytical devices [36]; the use of layered paper and tape as a fabrication method [42]; the use of fast lithographic activation sheets (FLASH) [164]; and the installation of functional elements onto the device [165].

Although such methods are usually associated with the assay processing aspect of the μPADs, they do present some interesting aspects that might benefit the energy sector as well. The use of a 3D origami like structure could lead to developments in energy processing, as the division of the device into multiple sectors could allow the fabrication of a hybrid energy storage device, with different areas controlling different energy fabrication methods. In purely fabrication terms, the construction of a device using only layered paper and tape is a highly scalable procedure, which is easy to implement in resource deficient regions. The same can be said about using FLASH, which, although being based upon the concepts of photolithography, does not require clean rooms nor special facilities, as its materials are composed of a simple hot plate and UV lamp, further facilitating the manufacturing of the device.

Even with the added ease of fabrication methods for paper-based microfuidics, they sometimes require materials or technical knowledge which are harder to acquire in developing or remote regions; hindering the mass-production of these devices. To use such fabrication techniques in more resource-deficient locations, further research needs to be done in order to either simplify the fabrication process or to find alternative, more readily available materials to use during the manufacturing of the device.

Further studies are still needed to reach a full understanding of the capacities and potential of commercial paper. In order to give paper-based microfluidics that necessary "push" to transform them into fully fledged novel devices, several studies still must be pursued: advances in the control over the structural properties of paper, the complete integration of novel material in the devices, further development in hydrophobization printing techniques, further studies on the substrate's surface energy, and how environmental conditions influence the performance of the devices [56].

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