Paper-based electronics: Towards sustainable electronics

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Abstract:
The emergence of paper-based electronic devices marks a significant leap forward in the design of flexible, lightweight, and eco-friendly electronics. Paper-based electronic sensors represent a transformative approach to creating flexible, lightweight, and environmentally friendly electronics. This review will discuss recent applications of paper-based electronics, mainly in exploring emergent technologies employed in developing innovative sensors for chemical analysis. Furthermore, the role of paper-based electronics in electrochemical, and physical sensing, specifically addressing relative humidity, temperature, pressure, and strain sensors will be commented. In addition, the integration of paper electronics in energy harvesting and storage is discussed, covering solar cells, tribogenerators, antennas, and supercapacitors. These advancements underscore the versatility and potential of paper-based electronics in diverse applications, from wearable health monitors to sustainable energy solutions, paving the way for the future of recyclable and biodegradable electronic devices.

Keywords: Responsible electronics · Paper-based electronic devices · Paper-based analytical devices · Chemical sensors · Electrochemical sensors.

1. Introduction

The consumption of Electrical and Electronic Equipment (EEE) has been growing enormously in the last years reaching 2.5 million metric tons (Mt) (excluding the contribution of photovoltaic panels) due to the widespread global economic development and due to the impressive technological progress in material science and engineering. Besides traditional electronics, the last decades have also seen the surge of multifunctional flexible electronics able to offer mature low-cost, thin, flexible, stretchable, and conformable devices, including sensors, memories, batteries, light-emitting diodes, energy harvesters, near-field communication/radio

frequency identification and circuitry. This has allowed the penetration of these new devices into many fields of smart electronics: consumer wearable and flexible electronics, interactive skin patches, implantable electronics, flexible lighting and display technologies, architecture, healthcare, robotics, Internet of Things (IoT) related technologies and more recently also in precision agriculture. The growing demand of such versatile and ubiquitous electronics dramatically calls for a reduction in the embodied energy (given by the sum of the energy needed in the production chain and in the storage process) and a higher level of process sustainability, requiring the treatment of the waste obtained at the end of the product life cycle and the implementation of circularity in the product development. In 2019, the world generated a striking 53.6 Mt of e-waste, i.e., 7.3 kg per capita on average, projected to grow to 74.7 Mt by 2030. To tackle this challenge, sustainable flexible electronics could represent a viable solution toward a more sustainable, environmentally friendly, and economically convenient approach thanks to the possibility to i) use biodegradable low-cost substrates; ii) employ low-cost abundant materials for the fabrication of the devices; ii) define technological process with reduced use of materials and lower processing temperatures concerning standard silicon.
Paper-based electronics can play a fundamental role in responsible electronics in such a scenario. This review paper describes the main advances achieved in this field. Paper-based devices are a perfect candidate for the fabrication of electronic sensors due to their unique combination of flexibility, sustainability, and low cost. Paper, as a substrate, offers a lightweight, biodegradable, and widely available platform, making it an eco-friendly alternative to conventional materials. Additionally, its ability to be printed using low-cost printing techniques allows for the mass production of electronic sensors at competitive prices. The porous nature of paper also facilitates the integration of functional materials, enhancing the sensitivity and selectivity of the sensors. These characteristics make paper-based devices not only environmentally responsible but also highly versatile and efficient for applications in various areas, including health, environmental monitoring, and wearable technology.

The paper is structured as follows. First, an introduction to the emergence of paper-based electronic devices is provided in Section 1. In section 2, we describe the use of paper as platform for the fabrication of sustainable electronic devices, discussing its advantages and drawback, also the structure of different type of paper and the impact of it on different fabrication methods. The main technologies used to manufacture electronic components from paper are described in section 3. Then, we comment on the latest achievements in sensors in Section 4, chemical sensors in Section 5, physical sensors in Section 6, and energy devices in Section 7. Finally, we

draw the main conclusions and future perspectives in the field of paper-based electronics in Section 8.

2. Paper as a platform for the fabrication of sustainable electronic devices

The paper has generally been described as a flexible sheet made up of an interlaced network of pressed cellulose fibers with an arbitrary porous membrane that allows fluids to be transported by capillary action. Therefore, the use of paper as a platform for the performance of sustainable devices for analysis has been widespread since the days of litmus paper [1]. Different types of paper can be used as substrates in the development of electronic devices, each with its characteristics for example, standard printing paper is one of the most used substrates with a smooth surface and low porosity which helps control ink absorption and spreading. This type of paper is commonly used for high-resolution printing, including printed circuits and sensors. It is ideal for well-suited inkjet and screen printing techniques. However, it may not be as durable or flexible as specialized papers, and the coating might affect the adhesion of certain types of inks. Two of the most common types of porous membranes that are used for assays are filter paper and nitrocellulose membranes. Filter paper is hydrophilic and is made of cellulose fibers with a high density of hydroxyl functional groups (-OH) and a few carboxylic acid groups (-COOH). Also, a summary of paper structure, properties, and the pros and cons of paper substrates for developing electronic devices is given in Table 1. Also, chromatograph papers are the most widely used cellulose-based papers for developing micropaper analytical devices (microPADs). Nitrocellulose is produced by nitrating cellulose, for example by replacing hydroxyl groups with nitrate groups. The resultant polymer is then cast into membranes with controlled porosity. Nitrocellulose membranes have a high protein binding affinity and are most commonly used in lateral flow assays and western blots for chemical analysis. In fact, in combination with cellulose-based paper and nitrocellulose, there are many other types of porous membranes with a variety of physical and chemical properties that could be used in the development of paper-based microfluidic devices. Coated paper is another type of paper mainly used for electronic device fabrication. This type of paper presents a surface coated with a polymer to create a smooth and uniform surface with low porosity due to the coating which minimizes ink absorption. The coated paper provides a smooth surface for precise ink application. Another common paper substrate used to fabricate electronic devices is recycled paper. The surface of recycled paper can be uneven or have a rough texture due to the inclusion of paper fibers from various sources. Also, recycled paper presents various porous sizes. It is

used in applications where environmental impact is a concern, and high resolution is less critical [2].

Paper, broadly understood as any hydrophilic porous membrane, has several unique advantages as a material for making sustainable electronic devices. It is inexpensive, widely available, and compatible with a wide range of printing and processing techniques. It is easy to store, stack, and transport. There is already an entire industry for the large-scale production of paper products that could be used to make paper-based devices. Paper is available in a variety of thicknesses and porosities. Its surface has a high density of organic functional groups that can be chemically manipulated and used to bind reagents covalently or non-covalently. Furthermore, paper has a high surface area to volume ratio, allowing reagents to be dried and stored on paper for later analysis. Reagents and samples can be easily coated onto paper either manually or using high-speed printing techniques [3]. Paper also has good filtration and chromatographic properties, which could be used to separate chemical compounds from complex samples. Additionally, paper wicks fluids by capillary action, allowing a paper-based device to move fluids and perform an assay without the need for pumps or external power sources. Fluid flow for the first time in a one-dimensional paper channel using the Lucas-Washburn equation, which states that the length of flow is proportional to the square root of time.

Table 1. Summary of paper advantages and drawbacks of using paper as a substrate for the manufacture of sustainable electronic devices.

Pros	**Cons**
○ Biodegradable and sustainable	○ Lack of flexibility
○ Low-cost	○ Less mechanical properties
○ Lightweight	○ Low thermal resistance
○ Esay chemical modification/ integration	○ Absorbs more humidity
○ High biocompatibility and degradability	○ Limited electrical conductivity
○ High specific surface area	
○ Versatile and great flexibility	

As mentioned above, for the development of affordable, portable, disposable, and low-cost tools to improve test equipment, paper-based analytical devices offer an alternative technology. These paper devices are manufactured using one of the nine reported techniques such as wax printing, photolithography, inkjet printing, plasma, and laser processing, flexography, wet etching, screen printing, and wax screen printing. All these techniques are based on the creation

of hydrophobic zones in the paper for the manufacture of the device, but they are very different from each other [4], [5], [6].

- Wax printing. The surface of the paper is printed with solid wax. The wax pattering includes painting with a wax pen, printing with a normal inkjet printer, or direct printing by a wax printer. The fabrication is simple, rapid (5-10 min), inexpensive, environmentally friendly (no use of organic solvents), and easily disposed of by burning. The wax printing technique defines patterns or functional hydrophobic channels onto the paper substrate. This technique is normally used to define regions where conductive inks are deposited to form flexible, low-cost circuits. The wax patterns act as insulating layers allowing multiple conductive paths on the same sheet of paper without causing short circuits. While wax printing is useful for defining large-scale areas or barriers, it may not be suitable for creating very fine, high-precision conductive paths, as it lacks the resolution and fine control offered by techniques like screen printing or photolithography [7].
- Screen printing. It is a versatile printing technique that uses a mesh screen to transfer ink onto a paper substrate, among others. This technique is usually used to deposit conductive inks for electronic functionality. It is a simple process to make conductive traces on paper substrates, also it is very common to fabricate antennas used in radio frequency identification tags, near-field communication labels, and wireless communication devices. Screen printing is ideal for creating low-cost, flexible batteries or supercapacitors. Screen printing allows the deposition of resistive inks, enabling the creation of simple resistive elements in electronic circuits directly on paper. The technique has some limitations in terms of resolution and conductivity. However, its scalability, environmental sustainability and ability to work with a wide range of inks and materials make it a valuable tool in the growing field of printed electronics [6], [8].
- Wax screen printing. This technique is fabricated by two simple steps, printing patterns of solid wax on the surface of paper using a simple screen-printing method and common household supplies, and melting the wax into paper to form complete hydrophobic barriers using a hot plate. The resulting hydrophobic channels are in the range of 1200-1800 mm. The wax is low-cost and can be purchased anywhere, and is also environmentally friendly [9].
- Inkjet printing. The inkjet method of printing recreates digital images by propelling droplets of ink onto paper and other substrates. Inkjet printing involves ejecting tiny droplets of functional ink (such as conductive, resistive or dielectric ink) from the nozzle

of the printer onto the paper substrate. These droplets are placed with high precision, enabling complex patterns like circuits, electrodes, or sensors to be created. The choice of ink is a function of the electrical or sensing properties required. The fabrication of paper devices is relatively inexpensive and produces excellent image quality with accurate color reproduction. This process is ideal for the low-cost, high-volume production of paper-based electronic devices [8], [10].

- Plasma processing. The paper is hydrophobized by silanization with octadecyltrichlorosilane. It is then submitted to plasma treatment. The plasma in the paper is converted into a network of hydrophilic channels. This is due to the degradation of the hydrophobic octadecyltrichlorosilane molecules attached to the paper's cellulose fibers [11].
- Laser processing. The fabrication procedure uses polymerization of photopolymer has guided the flow of fluids and allowed containment of fluids in wells. The CO_2 laser method involves cut a piece of paper by laser according to a predesigned pattern. The method is versatile and allows for controlled through-cutting and ablative etching of paper and nitrocellulose substrates. Also, fabrication is simple and rapid [12].
- Wet printing. The production process consists of two steps: The paper is made hydrophobic using a trimethoxyoctadecylsilane solution as a modulating agent, and a paper mask impregnated with a NaOH solution containing 30% glycerol is applied to the paper, which allows etching. The masked area of the paper becomes very hydrophilic and the unmasked area remains very hydrophobic.
- Photolitogaphy. This precise micro-fabrication technique is widely used in the semiconductor industry for the patterning of intricate designs on a variety of materials, including paper-based substrates and silicon wafers. The production of paper-based plates, in which sheets of paper are patterned into hydrophilic zones surrounded by hydrophobic polymer barriers, can be done using photolithography. Also, this technique uses a low-cost photostable formulation that allows rapid prototyping of paper-based plates (less than 15 min).
- Flexography. This is a simple method based on flexographic printing of polystyrene to form liquid guiding boundaries and layers on paper substrates allowing the formation of hydrophobic barrier structures that completely penetrate through the substrate. The method is compatible with roll-to-roll flexography units found in many printing houses, making it an ideal method for large-scale production of paper-based fluidic structures. Flexography also offers high-speed, large-area production capabilities. It is well suited

to printing functional materials such as conductive inks onto paper substrates. Flexography can also be used to deposit conductive inks, such as silver or carbon-based inks, onto paper to create circuits for simple electronic devices. Some of the advantages of this technique are high scalability, making it an efficient option for large-scale production; Cost-effectiveness, because the process is economical and uses low-cost materials, making it ideal for disposable devices; and Environmentally friendly, because it can be used with biodegradable materials such as paper and water-based ink [8].

3. Technology employed in paper-based electronics

The technology employed in paper-based electronics leverages the unique properties of paper as a versatile and sustainable substrate for creating electronic components. Various techniques, such as inkjet printing, screen printing, and flexographic printing, are used to deposit conductive inks and other functional materials onto paper, enabling the fabrication of a wide range of electronic devices. These methods allow for high precision and scalability, making it feasible to produce components like transistors, capacitors, and resistors directly on paper [13]. Additionally, advancements in material science have led to the development of conductive polymers and nanomaterials that can be seamlessly integrated with paper, enhancing the performance and durability of these devices. This innovative approach not only reduces production costs and environmental impact but also opens up new possibilities for flexible, lightweight, and biodegradable electronics suitable for applications in healthcare, environmental monitoring, and wearable technology. Thus, to produce electronic devices and systems with paper as base material, several techniques are being employed:

- Printed electronics. The use of conventional printing techniques to develop electronics allows the reduction in cost fabrication because the manufacturing conditions are not as strict as clean room processes as well as the modification of any layout is quite simple. Furthermore, printed electronics techniques allow the direct fabrication of flexible substrates as well as the employment of novel materials with innovative features (such as stretchability and/or biocompatibility) [14].
- Thin-film technology. Despite the advantages of printed electronics for IoT solutions, the performance and size of some electronic components make very difficult the fabrication of purely printed devices with a certain degree of complexity and a competitive size. Thin-film technology offers a compatible solution with printed electronics that allows the manufacturing of smaller and more complex electronics [15].

- Hybrid electronics. There are cases where the use of printed electronics and thin film technology is not enough to fulfill the requirements of the final application because further functionalities are needed. In that case, the so-called hybrid electronics is employed, which is the combination of the mentioned techniques with standard silicon technology [16].
- Reconfigurable electronics: An alternative to conventional dedicated circuitry, so-called Application-Specific Integrated Circuits (ASICs), are reconfigurable electronics. They offer different electronic resources that can be interconnected by software as desired. Such solutions allow the reuse and update of silicon circuitry without new fabrication processes [17].

It should be noted that paper and its derivatives have been extensively utilized as substrates for both electronic components and sensing applications, a topic we will explore in the following sections. This innovative use of paper not only provides a sustainable alternative to traditional materials but also enhances the versatility and accessibility of electronic and sensor technologies. As we delve deeper into the specifics, we will highlight the various advancements and techniques that have been developed to harness the potential of paper-based substrates in the field of electronics and sensing.

4. Paper-based electronic sensors

Paper-based electronic devices (EPEDs) are an innovative class of flexible, lightweight, and environmentally friendly sensors that enable new recyclable electronic devices such as paper displays, smart labels, biomedical applications, point-of-care devices, RFID tags, disposable electrochemical sensors, solar cells, and more. These devices can be adapted to diverse surfaces, making them suitable for wearable health monitors, interactive skin patches, and other applications requiring proximity to the human body. Additionally, their integration into intricate systems unlocks the potential for advanced healthcare, robotics, and Internet of Things (IoT) applications, fostering innovative and sustainable solutions across industries. Sophisticated and expensive technologies for physiological monitoring, transdermal stimulation, and therapeutics often impede their use as disposable medical devices. To address this challenge, developing epidermal, EPEDs is imperative for creating affordable wearable and implantable medical solutions [18]. EPEDs offer a budget-friendly alternative that is flexible, easy to apply, and can be disposed of through incineration. Their breathable nature and mechanical resilience, even under stretching, make them ideal for electrophysiological sensors, capable of recording electrocardiograms, electromyograms, and electrooculograms, even underwater. Furthermore,

EPEDs can deliver thermotherapeutic treatments to joints, map temperature distribution, and function as wirelessly powered implantable devices for stimulation and therapeutics. Their applications span continuous health monitoring, prosthetics, implantable devices, and advanced robotics.

Skin-mountable electronics include a diverse range of sensors that conform closely to the skin, enabling accurate detection of pressure, strain, temperature, and electrophysiological signals. These electronics exhibit outstanding electrical stability even when subjected to bending, twisting, or stretching. This stability is achieved through the use of conductive electrodes, which are usually created by crafting thin layers of flexible metals into intricate patterns such as serpentine paths, fractal geometries, or self-similar designs. This design approach helps avoid mechanical impedance mismatches with soft biological tissues, ensuring seamless integration and reliable performance. Several non-conventional methods to fabricate stretchable electrodes using nanocomposites comprising percolating nanoparticles, [19] conducting nanowires [20] or nanomeshes, [21] and microfluidic channels filled with ultralow modulus conductive materials [22], [23], [24] have been proposed to enhance the mechanical performance of skin-mountable devices and to provide them with self-healing properties and improved resistance to scratches and fatigue. The fabrication techniques typically used to generate skin-mountable electronics often involve high capital equipment and operating costs, hazardous chemicals, and several processing steps [25] limiting their practical utilization.

A quick, straightforward, and scalable method for producing cost-effective, disposable EESs with mechanical reinforcement would be highly advantageous. Such a process could expedite the advancement and market availability of wearable and implantable biomonitoring sensors and actuators. It could also facilitate their utilization in clinical environments, particularly in regions with limited resources. Paper-based devices are simple to fabricate, without requiring expensive materials or cleanroom facilities, using tools compatible with large-scale manufacturing processes [26]. Unfortunately, the mechanical and electrical properties of paper-based electronics are severely affected by environmental humidity [27] because of the hygroscopic nature of paper, making its use difficult as a low-cost substrate for the fabrication of skin-mountable devices.

Recently, it has been demonstrated the use of fluoroalkylated trichlorosilanes ($RFSiCl_3$) to alter the surface chemistry of cellulose fibers, rendering paper omniphobic resistant to wetting by aqueous solutions and organic liquids with surface tensions as low as 25 mN m^{-1} while preserving the mechanical flexibility, strength, and breathability of untreated paper [28]. Omniphobic RF paper has been used to fabricate a variety of low-cost electronics [29] and

microfluidic devices for environmental monitoring, [30] point-of-care diagnostics, [31] and biomedical fluid handling [32]. Unfortunately, because of its limited stretchability and lack of adherence to skin, RF paper has never been considered suitable for the development of skin-mountable devices.

5. Paper-based electronics for chemical analysis

Paper-based electronic devices for sensing have emerged as an innovative and accessible solution in the field of chemical analysis. Thus, PADs offer an economical and portable alternative to conventional analytical methods, and they are presented as an attractive solution by providing low-cost, flexible, simple, and portable devices because they allow for testing liquid samples through capillary action, eliminating the need for expensive and complicated equipment. These devices, typically composed of patterned paper, use microfluidic channels for testing liquid samples with capillary action, eliminating the need for external pumps. Their versatility makes them suitable for a variety of applications, from analyzing biological samples and food to environmental monitoring. Over the past decade, PADs have been extensively explored for applications in biological samples, food testing, and environmental analysis, utilizing chemical reactions in the detection zone. Their accessibility and ease of use make them promising tools for chemical analysis in diverse and resource-limited environments.

These paper-based devices are presented as ideal tools for chemical analysis compared to conventional analytic methods which are highly sensitive and selective, they often necessitate expensive equipment and extensive training [33]. The incorporation of electronic components onto cellulose paper platforms has given rise to a spectrum of applications, ranging from healthcare to environmental monitoring. In particular, chemical sensors stand out as a promising field within paper-based electronics, given their sensitivity, selectivity, and versatility. Some of the most outstanding examples in the development of paper-based electronic sensors for chemical applications are commented on below.

5.1. Fluorescence sensors

Paper-based analytical devices (PADs) for chemical analysis, specifically in the realm of luminescence, have gained significant attention and interest. These devices offer several advantages, making them an attractive platform for various applications. Luminescence-based detection methods on paper provide a portable, low-cost, and user-friendly alternative to traditional analytical techniques [34]. One notable feature of luminescent PADs is their versatility in detecting a wide range of analytes. Researchers have successfully developed

luminescent sensors for ions [35], small molecules, biomolecules [36], and various environmental pollutants [37]. The ability to tailor these devices for different applications showcases the flexibility and adaptability of luminescent PADs. Integration with advanced materials and nanotechnology has significantly enhanced the performance of luminescent PADs. Incorporating luminescent nanoparticles, quantum dots, or other nanomaterials as sensing elements has improved these devices' sensitivity, selectivity, and overall analytical performance [38]. Several fluorescent nanostructured materials have been implemented mostly in μPAD for the detection of clinical biomarkers and compounds of interest in the food and environmental field. Thus, carbon dots, metal-organic frameworks, and grapheme carbon dots have shown great potential for designing sensors so their implementation as receptors for building assays on cellulose platforms is very promising. Luminescent nanoparticles possess quantum yields much higher than those of conventional organic fluorophores as well as enhanced chemical and photoluminescent stability. In this sense, I. Ortiz-Gómez, et al. [39] developed a highly sensitive microfluidic paper-based analytical device for the determination of total carbohydrates in clinical and food samples. The device utilizes a flexible, portable, and reusable laser-induced graphene (LIG) heater to synthesize fluorescent silicon nanodots through a redox reaction between (3-aminopropyl) triethoxysilane (APTS) and carbohydrates, employing glucose and fructose as model compounds. The resulting fluorescence intensity is utilized for rapid carbohydrate screening, facilitated by a UV LED and a smartphone. The method exhibits a low detection limit (0.80 μM for glucose and 0.51 μM for fructose) and a linear response (10–200 μM for glucose and 10–100 μM for fructose).

These advancements pave the way for the development of highly efficient and reliable luminescent sensors on paper. In terms of fabrication, the simplicity of PADs is a key strength. The use of paper as a substrate allows for cost-effective and straightforward manufacturing processes. Techniques such as wax printing, screen printing, or inkjet printing enable the creation of well-defined patterns for luminescent sensing regions on the paper surface. These fabrication methods contribute to the scalability and mass production potential of luminescent PADs. The integration of luminescent PADs with smartphones for detection and quantification has opened up new avenues for on-site and point-of-care applications. Smartphone cameras can capture luminescent signals, and the data can be processed using dedicated applications, making these devices accessible and user-friendly for non-experts. Despite their numerous advantages, challenges remain in optimizing the sensitivity and specificity of luminescent PADs. The stability of luminescent materials on paper, the reproducibility of fabrication methods, and the interference from complex sample matrices are areas that require further exploration and

improvement. In this line, Ali. M. Yehia, et al. [40] have been working on a paper-based microfluidic device fabricated by wax printing to combine potentiometric, fluorimetric, and colorimetric detection zones. This trimodal paper chip is designed for on-site detection of ketamine hydrochloride (KET), a date rape drug, in beverages. The device utilizes polyaniline nano-dispersion as a conducting polymer in ion-sensing paper electrodes with a USB plug connector. Fluorescence and color detection zones employ carbon dots-gold nanoparticles and cobalt thiocyanate, respectively. The on-site detection is facilitated by a cellular phone's camera for fluorimetric and color detection. The trimodal detection system demonstrates specificity for KET in the presence of beverage interferences. The innovative sensor meets WHO criteria for point-of-care devices, making it suitable for rapid on-site drug diagnostics, not only for KET but potentially for other similar drugs.

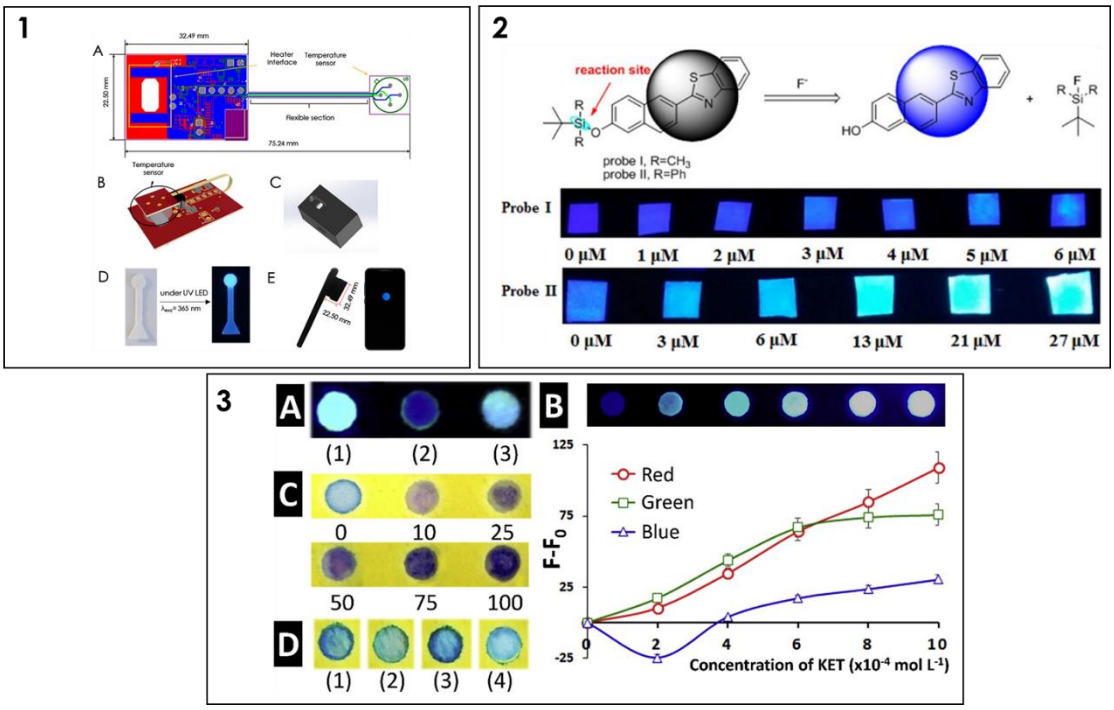

Figure 1. 1) Microfluidic paper-based analytical devices for fluorescent determination of glucose in clinical and food samples by fluorescence measurement under 365 nm excitation using UV-LED. 2. Paper-based device for fluoride ions determination in drinking water under 254 nm ultraviolet light. 3) Fluorescent spots of C-dots on a paper substrate under 395 nm excitation using UV-LED to determine ketamine. Adapted with permission from I. Ortiz et al., «In situ synthesis of fluorescent silicon nanodots for determination of total carbohydrates in a paper microfluidic device combined with laser prepared graphene heater», Sensors and Actuators B: Chemical, vol. 332, pp. 129506, April 2021), X. Wu, et al., «Highly sensitive ratiometric fluorescent paper sensors for the detection of fluoride ions», ACS Omega, vol. 4, pp. 4918-4926, 2019, A. M. Yehia, et al.,« A novel trimodal system on a paper-based microflu

idic device for on-site detection of the date rape drug “ketamine” », Anal. Chim. Acta, vol. 1104, pp. 95-104, april, 2020.

Furthermore, well-established method for protein detection has been miniaturized and incorporated into PADs, enabling the detection of specific biomolecules. They are particularly valuable for point-of-care applications, environmental monitoring, and resource-limited settings where traditional laboratory infrastructure is not readily available. Challenges remain, such as optimizing reaction conditions, ensuring stability, and addressing sample complexity, but the ongoing advancements in this field hold significant promise for enhancing chemical detection capabilities through the use of genetic material (RNA and DNA) immobilized onto paper-based platforms. For example, a luminescent-based analytical method for sensitive fluorometric detection of a milk allergen in food products has been recently developed based on using DNA bioconjugated to paper substrate [41]. Optimal performance is achieved at 37 °C for 40 minutes. The device successfully detects β-lactoglobulin reaching detection limits of 0.6 ng mL^{-1}. This fluorescence-based approach offers a simple and effective solution for detecting milk traces in food by utilizing a µPAD to specifically target β-lactoglobulin. The proposed platform is low-cost, portable, instrument-free, and highly sensitive, with easy sample pretreatment. Combined with a smartphone and electronic platform which includes a UV LED and dark housing, it allows for quantitative β-lactoglobulin detection based on fluorescence changes. The device achieves a detection limit of 0.6 ng/mL, surpassing previous paper-based sensors and commercial kits. This µPAD could be highly valuable to the food industry, offering a portable, affordable, and efficient tool for allergen detection and contamination control.

Table 2. Comparative table of paper-based fluorescence sensors.

Reference	**Measurement Method**	**Material**	**Sensitive range**	**Sensitivity**	**Response Time**
[39]	Fluorescence	Silicon nanodots	10-200 µmol L^{-1} for Glucose 10-100 µmol L^{-1} for Fructose	0.80 µmol L^{-1} for Glucose 0.51 µmol L^{-1} for Fructose	30 minutes
[40]	Fluorescence	Carbon dots-gold nanoparticles	$2x10^{-4}$ – $1x10^{-3}$ mol L^{-1}	$3.2x10^{-6}$ mol L^{-1}	20 seconds
[41]	Fluorescence	CQDs-DNA	1.8-500 ng mL^{-1}	0.6 ng mL^{-1}	40 minutes

5.2. Electrochemical sensors

Whitesides and colleagues [42] pioneered the use of patterned paper as a substrate for novel lab-on-a-paper systems, termed PADs. These systems combine the simplicity, portability, disposability, and low cost of paper strip tests with the multiplex analysis capabilities of conventional lab-on-a-chip devices [43], [44]. Recent advances in electronics have spurred research into the manufacture of electronics and circuits. This progress is further facilitated by cost-effective manufacturing techniques without compromising product quality. One notable application is ePADs, which use electrochemical principles to increase the sensitivity of microfluidic paper devices. Carbon and noble metals are commonly used as electrode materials in ePADs, with carbon being particularly attractive due to its low cost, ease of fabrication, simple chemical modification, and wide potential window in aqueous solvents.

Inexpensive and easy-to-use diagnostic tools for fast health screening are imperative, especially in the developing world, where portability and affordability are a necessity. An example is the work developed by Eloïse Bihar et al. [45] where a disposable analytical device that can measure physiologically relevant glucose concentrations in human saliva based on enzymatic electrochemical detection was carried out. In this work, all components of an enzymatic glucose sensor are inkjet printed onto disposable paper substrates. The process involves four steps of automated deposition of vertical films and is easily adaptable to the roll-to-roll process. The device has an operating range of 0.025 mM to 0.9 mM and exhibits good sensitivity for detecting glucose concentrations in saliva, suitable for screening for abnormal levels. When stored at room temperature under vacuum, the sensors retain their functionality for up to one month after manufacture, with only minor performance degradation (<25%). To incorporate the biorecognition element, an aqueous solution containing glucose oxidase (GOx) and a ferrocene (Fc) complex was printed onto the working electrode. Fc, a commonly used electron mediator in enzymatic sensors, serves as a co-substrate to replace oxygen. However, as Fc has weak adsorption properties on surfaces and its leakage may raise toxicity concerns, Fc is applied in a solution mixed with the polysaccharide chitosan. Finally, a thin layer of Nafion was printed over the sensing area, covering the working, counter and reference PEDOT: PSS electrodes. As a polyanion, Nafion acts as a barrier to interfering species present in the complex biological environment or generated by non-specific redox reactions during electrode operation. Thus, the developed work provides a low-cost paper device for the electrochemical determination of glucose using inkjet printing technology. The paper sensor is produced rapidly and inexpensively, making it highly suitable for widespread use, especially in resource-limited settings. The fabrication process of this paper device is also easily scalable to roll-to-roll

manufacturing. In addition, the design of this paper device reduces the amount of enzyme and material waste during production.

Furthermore, M. Punjiya et al. [46] present a microfluidic paper-based three-dimensional electrochemical device (3D-μPED) for electrochemical immunoassays as an ideal example of paper-based microfluidic devices with screen-printed electrodes for low-cost point-of-care electrochemical sensing. This work not only presents a low-cost, highly useful paper-based point-of-care device, but also allows simultaneous detection of glucose, pH and dopamine using potentiometric, amperometric and voltammetric modalities. Additionally, the 3D structure addresses several issues in paper-based sensor development, including variation in the electroactive area between sensors, non-specific analyte absorption leading to inconsistent analyte volumes, and paper pore size limiting particle size detection. Thus, the developed device features a three-dimensional paper-based microfluidic structure with screen-printed electrodes (SPEs), making it ideal for electrochemical sensing applications. The 3D-μPED was fabricated using a simple process that includes wax-printing, screen-printing, and folding. Additionally, the device integrates a custom-designed, miniaturized potentiostat IC, enabling versatile electrochemical measurements of biochemical analytes such as dopamine, glucose, and pH. Considering other practical applications, Quingpeng Cao et al. [47] were working on the development of a three-dimensional paper-based microfluidic electrochemical integrated device (3D-PMED) for real-time monitoring of sweat metabolites. The 3D-PMED was constructed by screen-printing wax patterns on cellulose paper and folding the pre-patterned paper four times to create five stacked layers: the sweat collector, vertical channel, transverse channel, electrode layer, and sweat evaporator. Integration of a screen-printed glucose sensor on a polyethylene terephthalate (PET) substrate with the fabricated 3D-PMED realized a sweat monitoring device. Sweat flow in the 3D-PMED was simulated using red ink to illustrate its ability to collect, analyze, and evaporate sweat, facilitated by the capillary action of the filter paper and the hydrophobic nature of the wax. The glucose sensor was designed with high sensitivity (35.7 mA mM^{-1} cm^{-2}) and low detection limit (5 μM) to account for the low concentration of glucose in sweat. In summary, this wearable 3D-PMED is presented as an effective and practical paper-based device for monitoring glucose levels in human sweat. Its notable advantages include high simplicity, portability, and ease of use, along with the capability for real-time and non-invasive monitoring.

In addition, the latest advancements in paper-based electronic devices for sensitive point-of-care applications show great promise for detecting a wide range of biomarkers with high sensitivity and accuracy. For example, Panpan et al. [48] developed a three-dimensional paper-

based electrochemical immunodevice using multi-walled carbon nanotube (MWCNT)-functionalized paper for the sensitive, simultaneous detection of two tumor markers—cancer antigen 125 and carcinoembryonic antigen—in serum samples. The microfluidic, paper-based three-dimensional electrochemical device consisted of a wax-patterned paper layer and a screen-printed electrode layer on a transparent polyethylene terephthalate (PET) substrate. The wax patterns formed reservoirs for the electrochemical cells, while the screen-printed electrodes were connected by aligning the wax-patterned electrochemical cells after stacking. MWCNTs were employed to modify the porous structure of the paper working zones, enhancing the electronic conductivity of the electrochemical cell on the microfluidic paper-based analytical devices (μPADs). Immunoreactions were carried out on antibody-modified paper working zones, which were prepared using chitosan coating and glutaraldehyde cross-linking. The device demonstrated a linear detection range of 0.001-75.0 U/mL for cancer antigen 125 and 0.05-50.0 ng/mL for carcinoembryonic antigen.

Table 3. Comparative table of paper-based electrochemical sensors.

Reference	Measurement method	Material	Sensitivity range	Sensitivity	Response time
[45]	Chronoamperometry	PEDOT:PSS	0.025 - 0.9 mM	-	0.1 seconds
[46]	Chronoamperometry	CMOS	5 – 17.5 mM	0.34 μA/mM	60 seconds
[47]	Amperometry	Chitosan/Nafion/GOx	0-2 mM	5 μM	-
[48]	Voltamperometry	glutaraldehyde/chitosan /MWCNTs	0.001-75.0 U mL^{-1}	0.2 mU mL^{-1}	-

6. Paper-based physical sensors

6.1 Relative humidity sensors

Humidity sensors have become a global phenomenon with the rapid growth of their applications in many fields. To mention some examples, in the agriculture industry, humidity sensors are used to monitor soil moisture, optimize water use, and promote healthy plant growth. In the field of medicine, they are used for monitoring parameters related to patients [49] and medical equipment such as sterilizers, incubators, and other machines that need humidity control [50]. Furthermore, these sensors are also present in the food storage and food processing industry [51]. Given the wide range of applications in which these types of sensors are involved, they must be designed to be versatile, cheap, and adaptable to any surface, which makes the use of paper a good candidate. For this reason, paper-based humidity sensors have been widely studied in recent years, where different works showed that paper can be employed as either a flexible

substrate or as an active layer, due to its porous fiber nature, that can be modified by infiltration with electronic materials, or it can be directly used as the sensing layer.

To illustrate a recent example where paper is used as the substrate, Lim, W. Y. et al. reported RH sensors by inkjet-printing graphene in the shape of interdigital electrodes on glossy paper [52], where changes in relative humidity are manifested in changes in the capacity of the electrode (Figure 3a). Their device exhibited a capacitive sensitivity of 0.03 pF/%RH in the range of 10-70 %RH with good stability and high intraday and interday repeatability. In the same line, Xie, M. Z. et al. [53] presented a paper-based inductance-capacitance (LC) humidity sensor with a single spiral coil, fabricated directly on A4 printing paper. They screen-printed a coil with nano-silver paste on ordinary printing paper. The sensor coil showed a sensitivity and response time of 120 kHz/%RH and 60 minutes respectively, with a hysteresis error of less than 0.9% in the humidity range of 15%RH to 75%RH.

Related to the use of paper as an active layer by infiltration of electronic materials, Zhu, P. et al. [54] designed its flexible paper-based substrate for humidity sensors based on conductive 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized cellulose fibers/carbon nanotubes (TOCFs/CNTs) conformal fibers network. The conductive fibers were produced through an electrostatic self-assembly process between previously positively charged CNTs absorbed by the surface of negatively charged TOCFs. The hydrophilicity of TOCFs offers more adsorption sites for water molecules and enhances the transfer of electrons from water molecules to CNTs, allowing the sensor to be highly responsive to humidity (Figure 3b). Taking advantage of this specific layout, the sensor exhibits a maximum response value of 87.0% $\Delta I/I0$ and excellent linearity ($R^2 = 0.995$) between 11 and 95% relative humidity (RH).

The simplest approach is accomplished in [55] where a humidity sensor using conventional printing paper as both, the humidity-sensing material and the substrate of the sensor is developed (Figure 3c). The current through a piece of paper covered with adhesive tape (to make it foldable) was measured while applying a bias voltage. This sensor showed an excellent response and recovery time of ~472/~19 seconds and even though a good linearity of 0.9549 is achieved in a working range of 41.1% to 91.5%, no obvious response was found below 72.0% RH. Despite of this, the sensor could be suitable for some applications involving high RH like breath rate or baby diaper wetting detection.

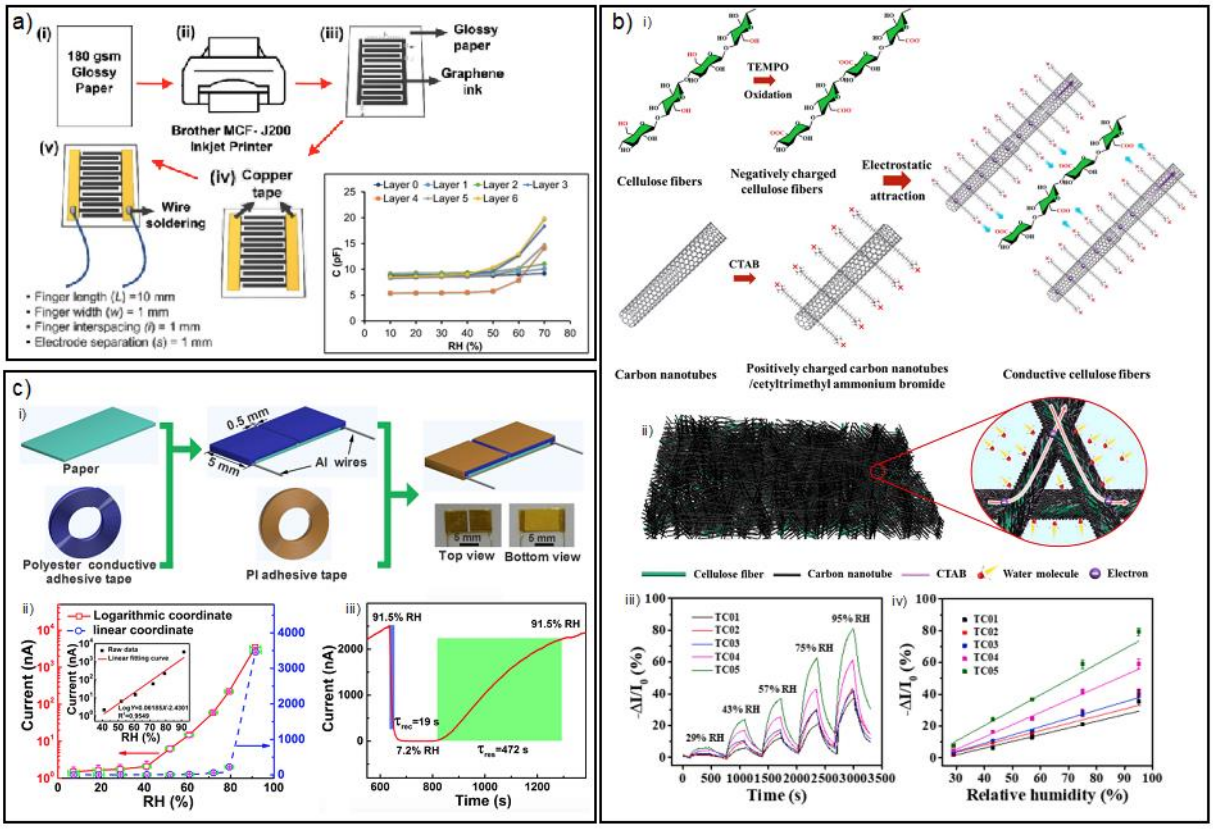

Figure 2. a) Schematic illustration of the fabrication process for the inkjet-printed-paper sensor with graphene printing ink [52] and performance of the sensor for changes on RH% depending on the number of printed layers. b) Illustration of the process to form the conductive cellulose fibers and the fibers network structure of the obtained paper sensor [54]. i) Illustration of the formation of conductive cellulose fibers by electrostatic attraction of negatively charged TOCFs and positively charged CNTs, ii) schematic illustration of the paper network based on the conformal conductive TOCFs/CNTs fibers, iii) dynamic response and recovery curves of the paper sensors with various TOCFs-to-CNTs ratios between 11% and 29, 43, 57, 75, and 95% RH. iv) The response of the paper sensors with various TOCFs-to-CNTs ratios as a function of RH. c) i) Fabrication process of the paper-based humidity sensor proposed in [55], ii) current versus RH curves (the inset shows the linear fitting curve of current versus RH) and iii) is the amplified response and recovery curve in the linear coordinate system. (Adapted with permission from Z. Duan et al., «Facile, Flexible, Cost-Saving, and Environment-Friendly Paper-Based Humidity Sensor for Multifunctional Applications», ACS Appl. Mater. Interfaces, vol. 11, n.o 24, pp. 21840-21849, jun. 2019).

Table 4. Comparative table of paper based humidity sensors.

Reference	Measurement Method	Material	Sensitivity	Response/Recovery Time
[52]	Capacitive	Inkjet-printed graphene on glossy paper	0.03 pF/%RH (10-70%RH)	< 5 s
[53]	Inductance-Capacitance (LC)	Screen.printed Nano-silver paste on A4 paper	120 kHz/%RH	60 min / 40 min
[54]	Resistive	TEMPO-oxidized Cellulose Fibers/Carbon Nanotubes	87.0% ΔI/I0	333 s / 523 s
[55]	Resistive	Conventional Printing Paper	Linear between 41.1%-91.5% RH	~472 s / ~19 s

6.2. Temperature sensors

Temperature monitoring plays a crucial role in a lot of applications, not only is it important in the agriculture sector or climatization monitoring but also can be vital in applications such as maintaining the cold chain in food logistics or helping diagnose diseases in humans. In consequence, the study of flexible temperature sensors has been tackled in the past few years, most of them based on nanomaterials and conductive polymers [56]. The main substrates used for this type of sensor are polyimide (PI), polydimethylsiloxane (PDMS), polyurethane (PU), and polyethylene terephthalate (PET), but when it comes to the paper substrate, the problem of its porous structure influencing electrical properties arises, making it difficult to perform measurements. A paper-based temperature sensor is presented in [57] where the paper was covered by dipping it into poly(3,4-ethylene dioxythiophene): poly(4-styrene sulfonate) PEDOT: PSS solution and adhered to a band-like body-attachable thermometer for wearable purposes (Figure 4a). The sensitivity achieved in the range of 25-55 ºC was 658.5 Ωcm^{-1} with a linearity of 99.43%. For higher temperatures, although linearity was reduced, it remained over 99% and sensitivity lowered to 286.6 Ωcm^{-1} in the range of 55 ºC to 75 ºC and to 148.8 Ωcm^{-1} in the range of 75 ºC to 100 ºC. Also, they demonstrated the enhancement in temperature sensitivity of using paper substrate compared to polyimide substrate. An electrode-based sensor is exposed in [58] where a resistive temperature sensor was printed using inkjet printing of silver nanoparticles on uncoated paper substrates. Within the temperature range of interest (20 °C - 80 °C), all sensors showed a good linear temperature dependency, minimal hysteresis,

and low baseline drift. However, the accuracy and overall quality of the sensors decreased in an extended temperature range (−25 °C to 150 °C) and exposed to humidity (0%RH–80%RH). A different approach is performed in [59], where Tao, X. et al. presented a paper thermometer based on an ionic liquid thermosensitive fluid deposited on the paper substrate by pen writing or inkjet printing (Figure 4b). The thermosensitive ink was trapped within the cellulose fibers of a paper matrix and by shortening the heat exchange distance between ionic liquid and samples, it took only 8s for the thermometer to reach an electrical equilibrium at a given temperature. The thermal response of the paper thermometer reached over 60% by increasing temperature from 25 ºC to 45 ºC, keeping unvarying after multiple cycles of heating and cooling. In addition, a study of the bending and folding performance of this sensor was carried out, proving good performance in both proceedings.

Table 5. Comparative table of paper-based temperature sensors.

Reference	**Measurement Method**	**Material**	**Sensitivity range (ºC)**	**Sensitivity**	**Response Time**
[57]	Resistive	PEDOT: PSS	30 – 42 55 – 75 75 – 100	625.9 Ω/ºC 286.6 Ω/ºC 148.8 Ω/ºC	-
[58]	Resistive	Silver nanoparticles	20 -80	1.63×10^{-3} K^{-1} (TCR)	-
[59]	Resistive	Ionic liquid	25-45	60.5 %	8 seconds

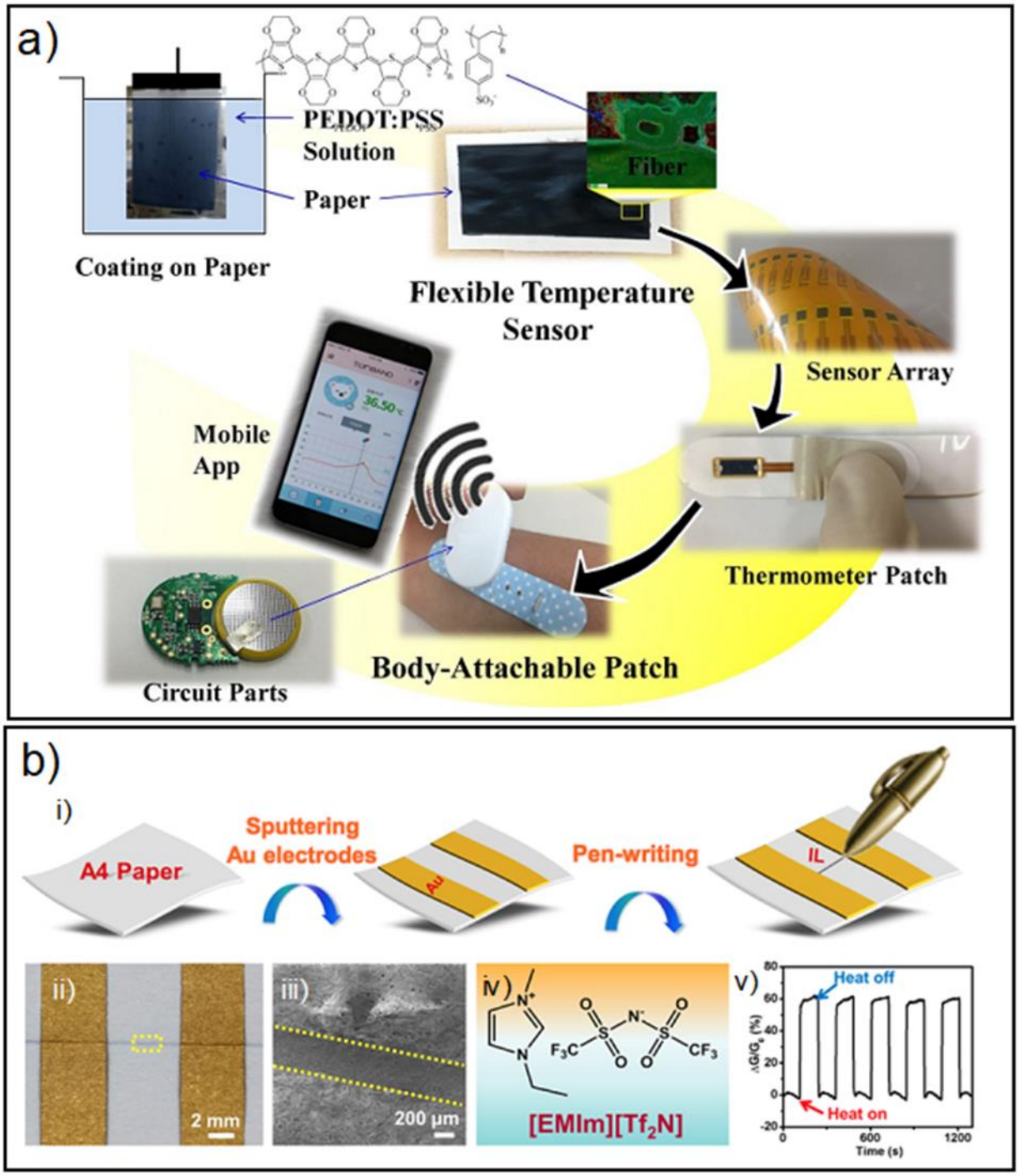

Figure 3. a) Fabrication process and application of PEDOT:PSS and paper based temperature sensor in [57]. b) i) Schematic illustration of the fabrication process of pen-written ionic liquid paper chip [59]. ii) Picture of ionic liquid based paper chip. The yellow dotted box region marked is magnified as a SEM image in iii). iv) Molecular structures of ionic liquid [EMIm][Tf_2N] serving as ink. v) On-off cycles of thermal response of paper chip operated between 45 ºC and 25 ºC. (Reprinted and adapted with permission from X. Tao, H. Jia, Y. He, S. Liao, y Y. Wang, «Ultrafast Paper Thermometers Based on a Green Sensing Ink», ACS Sens., vol. 2, n.o 3, pp. 449-454, mar. 2017).

6.3 Pressure sensors

In the matter of pressure sensors, applications in health care systems include pulse rate and blood pressure monitoring, breath monitoring, foot plantar pressure monitoring or intraocular and intracranial pressure monitoring [60]. Moreover, pressure sensors applied to the field of robotics and human-machine interfacing devices have also been reported. However, achieving high sensitivity over a wide pressure range still a challenge, and the demand for pressure sensors with high sensitivity, wide measurement range and low costs is growing so fast [60]. Pressure sensors should have a good sensitivity in the range of 0−20 kPa when applied in wearable applications. Traditional pressure sensors cannot achieve both a high sensitivity and a large working range simultaneously, which results in their limited applications in wearable fields. Therefore, there is an urgent need to develop a pressure sensor to make a breakthrough in both sensitivity and working range. In [61] Lee, T. et al. demonstrated a paper-based resistive tactile sensor using carbon nanotube- and silver nanoparticle-printed mulberry paper as the pressure-sensing element and electrodes, respectively (Figure 5a). The rough surface and high inner porosity of the mulberry paper showed high sensitivity to pressure, exceeding 1 kPa^{-1} in an applied pressure range of 0.05-900 kPa. The sensor also presented a fast response/relaxation time, low detection limit, high resolution, high durability, and high flexibility. Gao, L., et al. [62] developed a paper-based piezoresistive (APBP) pressure sensor based on a tissue paper coated with silver nanowires (AgNWs) as a sensing material, a nanocellulose paper (NCP) as a bottom substrate for printing electrodes, and nanocellulose paper (NCP) as a top encapsulating layer (Figure 5b). A high sensitivity of 1.5 kPa^{-1} in the range of 0.03 − 30.2 kPa was achieved by the APBP sensor as well as retaining an excellent performance in the bending state. Furthermore, the sensor was successfully applied as a skin electrode to monitor physiological signals (arterial heart pulse and pronunciation from the throat) and responded well to external pressure. In [63] a strategy based on a special multilayered cellulose paper structure composed of alternate layers of plain and corrugated sheets coated with 2D tin-monosulfide (SnS) was employed (Figure 5c). With this paper structure design, a high sensitivity up to 14.8 kPa^{-1} was achieved as well as a wide working range of 0−120 kPa and a maximum time response of 1ms. Tao, L. Q. et al. [64] presented a graphene-paper-based pressure sensor that showed good performance in a range of 0 – 20 kPa and a sensitivity of 17.2 kPa^{-1} (0 – 2 kPa). They also showed demonstration of use in breath and wrist pulse detection, movement monitoring, and voice recognition (Figure 5d). Finally, in [65] an increase in sensitivity (1911.4 KPa^{-1}) is achieved with an all-tissue-based piezoresistive pressure sensor based on a bottom interdigitated.

Table 6. Comparative table of paper based pressure sensors.

Reference	Measurement Method	Material	Sensitivity Range (kPa)	Sensitivity	Response / Recovery Time
[61]	Resistive	Carbon Nanotube, Silver	0.05 – 100 100 – 300 300 -900	6.67 kPa^{-1} 2.80 kPa^{-1} 1.19 kPa^{-1}	20 ms / 80 ms
[62]	Piezoresistive	Silver Nanowires, Tissue Paper	0.03 – 30.2	1.5 kPa^{-1}	90 ms
[63]	Capacitive	Multilayered Cellulose, SnS	0 – 10 10 – 40 40 – 120	14.8 kPa^{-1} 8.4 kPa^{-1} 3.28 kPa^{-1}	1 ms
[64]	Piezoresistive	Graphene-Paper	0 – 2 2 - 20	17.2 kPa^{-1} 0.1 kPa^{-1}	120 ms / 60 ms
[65]	Piezoresistive	All-tissue-based	-	1911.4 kPa^{-1}	4 ms / 3 ms

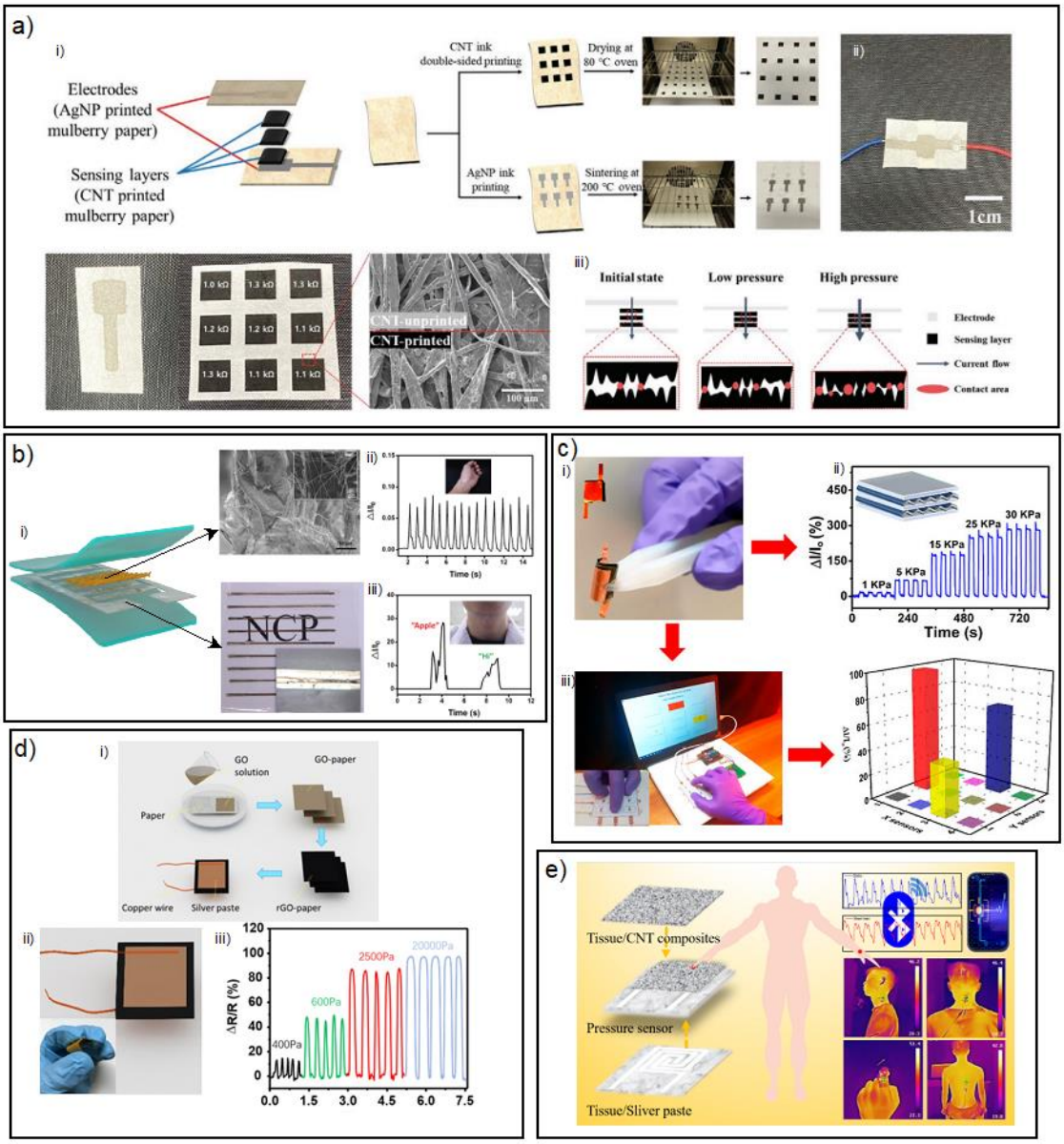

Figure 4. a) i) Schematic illustration of proposed tactile pressure sensor using inkjet-printed mulberry paper [61] ii) Preparation process of inkjet-printed mulberry paper. iii) Picture of fabricated tactile sensor iv) Picture of AgNPs- and CNT-printed mulberry paper. v) Pressure-sensing mechanism of tactile sensor. b) i) APBP pressure sensor with a sandwich structure [62]. ii) Detection of arterial heart pulse on the wrist iii) Responsive curve of the pressure sensor mounted on the throat to monitor pronunciation. (Reprinted and adapted with permission from L. Gao *et al.*, «All Paper-Based Flexible and Wearable Piezoresistive Pressure Sensor», *ACS Appl. Mater. Interfaces*, vol. 11, n.º 28, pp. 25034-25042, jul. 2019). c) i) Flexible and foldable pressure sensor [63] ii) dynamic response cycles to different load values of 1,5,15,25, and 30 kPa. iii) Photograph of three fingers pressing onto the paper-based keyboard connected to a computer showing in different colors the amount of pressure at different positions. (Reprinted and adapted with permission from N. Sakhuja, R. Kumar, P. Katare, y N. Bhat, «Structure-Driven, Flexible, Multilayered, Paper-Based Pressure Sensor for Human–Machine Interfacing», *ACS Sustain.*

Chem. Eng., vol. 10, n.º 30, pp. 9697-9706, ago. 2022). d) i) Fabrication process of the graphene-based pressure sensors with the paper substrate [64]. ii) Picture of the pressure sensor iii) Response test of the sensor at different pressure values. (Reprinted and adapted with permission from L.-Q. Tao *et al.*, «Graphene-Paper Pressure Sensor for Detecting Human Motions», *ACS Nano*, vol. 11, n.º 9, pp. 8790-8795, sep. 2017). e) Schematic illustration of the fabrication procedure of all-tissue-based pressure sensors [65].

6.4 Strain sensors

Strain is defined as the ratio of change in dimension to the initial dimension. It is the measure of a material inability to completely regain its original shape and size. Usually, strain sensors transduce external mechanical stimuli into electrical signals [66]. Similarly, to pressure sensors, applications for strain sensors include wearable health monitoring devices, detection of human body movements for sports performance monitoring, interactive gaming, and virtual reality or soft robotics and neuromechanics [67]. When it comes to paper-based strain sensors, the strain sensitivity is mainly caused by the piezoresistive properties of a conductive material deposited on the paper substrate. Changes in the conductive material network due to mechanical deformation produce an alteration in its resistance which can be related to the amount of strain presented in the paper substrate [68].

Based on a paper substrate dip-coated in an aqueous suspension of carbon black (CB) and carboxymethyl cellulose (CMC), Liu, H. et al. [69] proposed a flexible and degradable strain sensor, obtained by assembling the coated paper and wires with silver paste (Figure 6a). The sensor was capable of monitoring human movements with high stability over 1000 cycles, displaying a gauge factor of 4.3 and a responsive time of approximately 240 ms. In [70] Liao, X et al. reported a paper-based strain sensor built from graphite glue (graphite powder and methylcellulose). Sensitivity was improved by exposing the graphite glue to thermal annealing to produce surface micro/nano cracks, which are very sensitive to compressive or tensile strain. The device presented a gauge factor of 804.9, a response time of 19.6 ms, and a strain resolution of 0.038%. Shen, L. et al. [71] presented a strain sensor based on a laser-induced carbonization electrode fabricated by direct laser writing over filter paper (Figure 6b). By adding lignin and applying bending cycles, the gauge factor was boosted, which enabled the sensor to satisfy high sensitivity for weak tension and compression strain. Moreover, the sensor was proven to be applied for monitoring physiological activities like finger gestures, pulsing, swallowing, and eye blinking. A different approach was tackled in [72] where a photo paper with 1mm separated slits was covered by an Ag thin film and finally treated with a superhydrophobic coating to make it water resistance (Figure 6c). When the sensor was exposed to stress, the quantity of the

connections between Ag nanoparticles (Ag NPs) and multiwalled carbon nanotubes (MWCNTs) distributed on the two walls of each groove, causing an increase or decrease in the conductive pathways that leaded to a change in resistance. As a result, the strain sensor exhibited a gauge factor of 263.34, a resolution of 0.098% and a response time of 78 ms, as well as a good stability over 12,000 cycles.

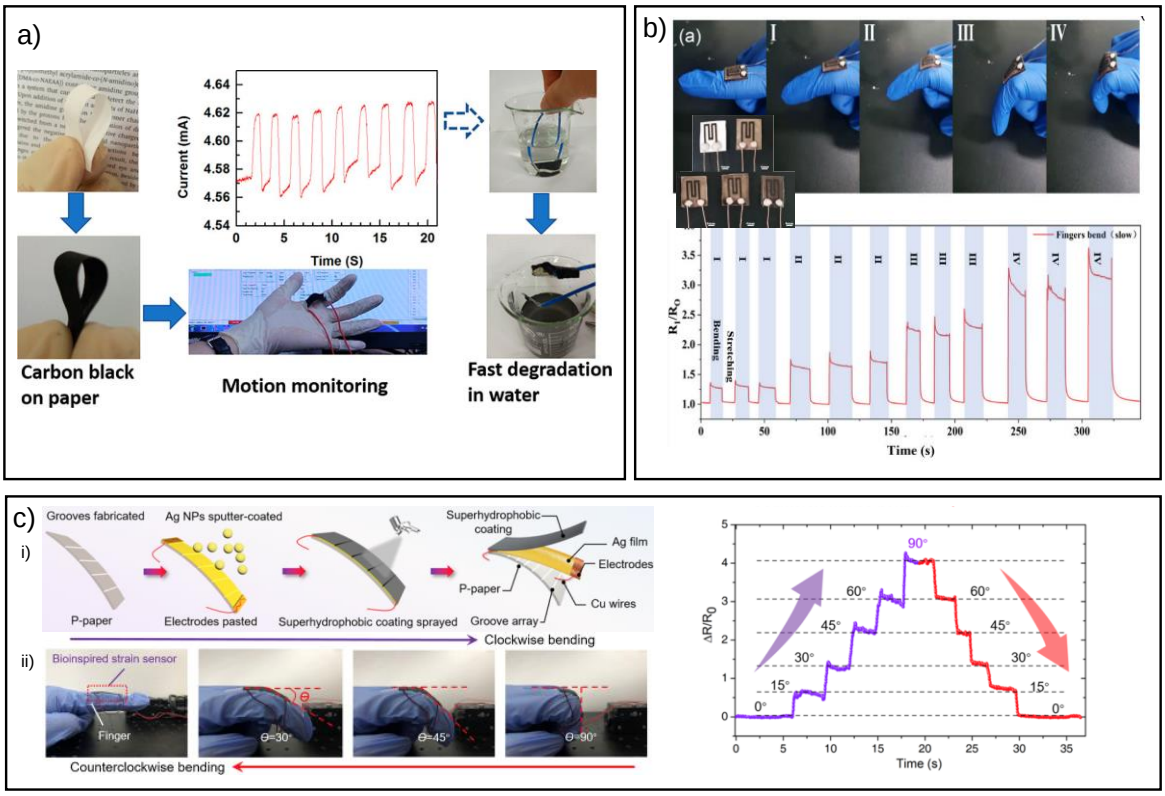

Figure 5. a) Life cycle of strain sensor in [69] and current change at 3V for finger flexing. (Reprinted and adapted with permission from H. liu, H. Jiang, F. Du, D. Zhang, Z. Li, y H. Zhou, «Flexible and Degradable Paper-Based Strain Sensor with Low Cost», *ACS Sustain. Chem. Eng.*, vol. 5, n.º 11, pp. 10538-10543, nov. 2017). b) Strain sensors in [71] with different lignin concentrations and finger movement monitoring. c) i) Design concept and fabrication process of the bioinspired strain sensor based on the strategy of coupling bionics described in [72] ii) sensor fixed on a finger with different bending angles and its relative change in resistance under these different bending angles iii). (Reprinted and adapted with permission from L. Liu *et al.*, «Bioinspired, Superhydrophobic, and Paper-Based Strain Sensors for Wearable and Underwater Applications», *ACS Appl. Mater. Interfaces*, vol. 13, n.º 1, pp. 1967-1978, ene. 2021).

Table 7. Comparative table of paper based strain sensors.

Reference	Measurement Method	Material	Gauge factor	Response / Recovery Time
[69]	Piezoresistive	Carbon Black, CMC	4.3	~ 240 ms / -
[70]	Piezoresistive	Graphite glue (thermally treated)	804.9	22.3 ms / 19.6 ms
[71]	Resistive	Laser-induced carbon, lignin	408 (tensile) 91 (compressive)	0.165 s / 0.265 s 0.631 s / 0.394 s
[72]	Resistive	Silver film, Ag nanoparticles, MWCNTs	263.34	78 ms / -

7. Paper-based energy harvesters and storage elements

7.1 Solar cells

Paper-based solar cells have been the subject of extensive research in recent years, with different perspectives not only about which part of the cell the paper or cellulose is used but also the type of photovoltaic technology employed [73]. For this reason, in this section, the main parts of solar cells, the development of the different generations, and the most recent trends are explained.

Solar cells are usually comprised of an active layer and two electrodes, with the requirement of one of them to be transparent. The latter electrodes are commonly made of transparent conductive oxides (TCOs) unlike back electrodes which are usually made with metallic contact of gold (Au), silver (Ag), aluminum (Al), or platinum (Pt). On the other hand, solar cells are typically divided into three categories. The first-generation photovoltaic (PV) cells are based on mono- and multicrystalline silicon solar cells which are usually characterized by high efficiency and durability but are mechanically vulnerable and incompatible with cellulose substrates. The second generation includes thin films, usually amorphous silicon (a-Si) and copper indium selenide (CIS), and have less thickness (<10 μm) as compared to first-generation. Finally, third-generation PV cells embrace material-engineered thin-film technologies, specifically organic photovoltaics (OPV), quantum dot photovoltaics (QDPVs), dye-sensitized solar cells (DSSCs), and perovskite solar cells (PSCs). PV on paper could be applied as a “disposable” power source for gadgets, electronic labeling, or remote sensing systems. For this reason, efforts on manufacturing good electrodes are of great importance, and since paper has a rough surface, this characteristic has led to the following approaches when it comes to overcome the problem of designing electrodes over cellulose:

1) Optimization of bottom electrode deposition avoiding or exploiting the surface structure. In [74] Bella, F. et al. used natural cellulose fibers as photoanodes and polymer electrolytes in dye-sensitized solar cells (DSSCs) (Figure 7a). The prepared paper-DSSCs assembled with the cellulose-based electrodes and electrolytes guaranteed sunlight conversion efficiency as high as 3.55 and 5.20% at simulated light intensities of 1 and 0.2 sun, respectively, along with an excellent efficiency retention of 96% after 1000 h of accelerated aging test.
2) Modification of the paper surface by using planarization layers. As proposed in [75], to overcome the problem of paper roughness, UV curable lacquer was employed to planarize the surface of a plain 80 grams of printer paper, taken as a substrate. The lacquer smoothens the rough surface of the paper such that a designed nanostructure can be imprinted for light scattering. Thus, single junction amorphous silicon solar cells with a hot wire chemical vapor deposition (HWCVD) intrinsic layer were processed on paper. The cell performance was proven to be comparable to that of reference cells grown on stainless steel, verifying that solar cells can be deposited on paper substrates without sacrificing performance (Figure 7b).
3) Use of nanocellulose substrate with smooth surface. In [76] transparent and flexible cellulose-based nanocomposite papers were fabricated for its use as solar cell substrates. O-(2,3-Dihydroxypropyl) cellulose (DHPC) synthesized by homogeneous etherification was transparent and flexible but poor in mechanical properties, so rigid tunicate cellulose nanocrystals (TCNCs) were introduced to reinforce the cellulose. The flexible cellulose-based polymer solar cells exhibited a PCE of 4.98%, showing enormous potential for optoelectronic devices (Figure 7c).
4) Development of conductive cellulose substrates. Based on optically transparent nanofiber paper containing silver nanowires [77] Nogi, M. et al. demonstrated organic solar cells that achieved a power conversion of 3.2% (Figure 7d). The transparent conductive nanofiber paper showed high electrical durability in repeated folding tests, due to dual advantages of the hydrophilic affinity between cellulose and silver nanowires, and the entanglement between cellulose nanofibers and silver nanowires and their optical transparency and electrical conductivity were as high as those of based on indium tin oxide (ITO) glass.

A third-generation perovskite solar cell (PSC) is developed in [78], based on cellophane paper and using oxide/ultrathin Ag/oxide (OMO) and perovskite as electrodes and absorbers,

respectively e). The perovskite solar cell (PSC) on cellophane exhibited a PCE of 13.19%, the highest among all the paper-based solar cells.

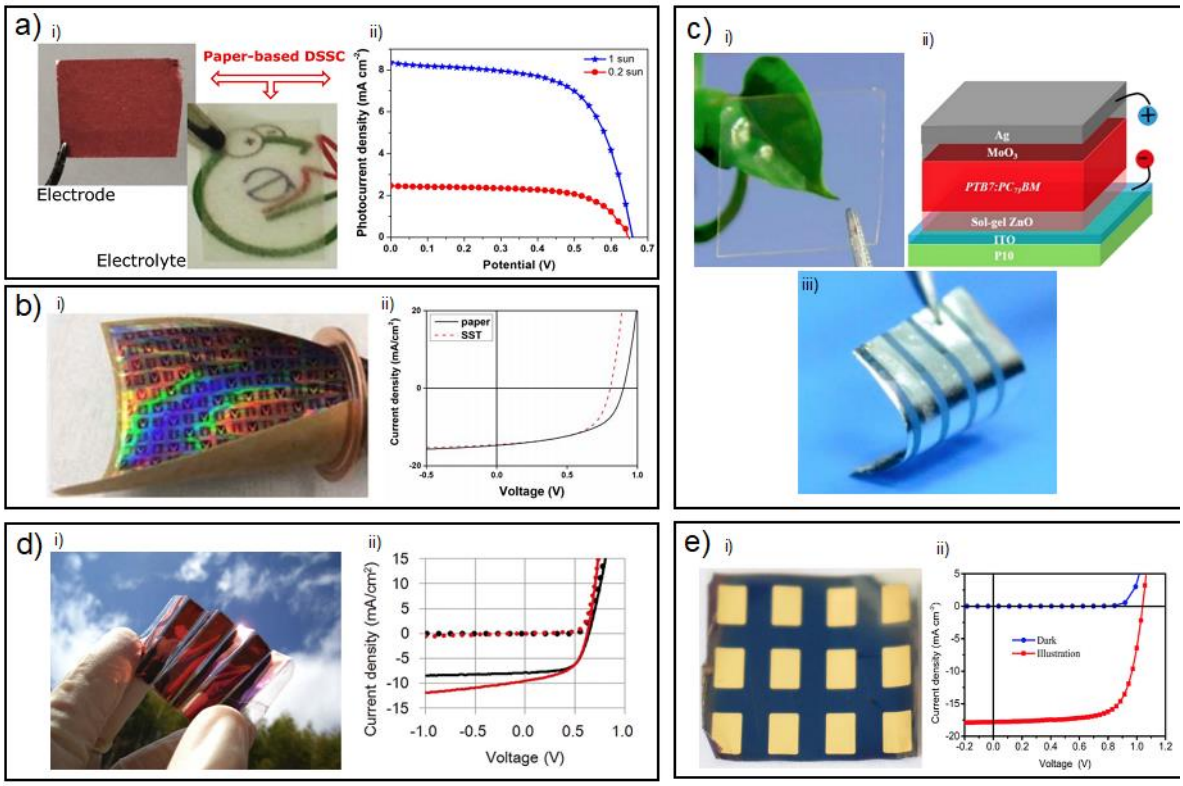

Figure 6. a) Paper-based photoanode electrode in its sensitized version and paper-based polymer membrane (electrolyte) before the activation in the iodine-based redox couple solution ii) photocurrent density-potential curves measured under different irradiation intensities. b) i) photograph of the completed cells (10x10 cm^2) rolled up in a (CF-40) copper gasket and ii) J-V curve of the best cell on paper and on stainless steel (SST). c) i) DHPC/TCNC nanocomposite paper ii) structure of an inverted polymer solar cell and iii) Photograph of a solar cell [76]. (Reprinted and adapted with permission from Q. Cheng *et al.*, «Construction of Transparent Cellulose-Based Nanocomposite Papers and Potential Application in Flexible Solar Cells», *ACS Sustain. Chem. Eng.*, vol. 6, n.º 6, pp. 8040-8047, jun. 2018). d) i) Photograph of the portable solar cells [77] based on foldable and lightweight transparent conductive nanofiber paper and ii) current-voltage characteristics of the organic solar cells (P3HT/PCBM) in the dark (broken lines), and under 100 mW/cm^2 of AM 1.5G illumination (solid lines); red plot: transparent conductive nanofiber paper-based solar cells; black plot: indium tin oxide glass-based solar cells. e) i) perovskite solar cell on cellophane [78] and ii) dark and illuminated J-V curves of PSCs on cellophane.

Table 8. Comparative table of paper-based solar cells.

Reference	Photovoltaic Technology	Substrate	Electrode	Efficiency
[47]	DSSCs	Cellulose fibers	Cellulose-based polymer electrolytes	3.55 % 5.20 %
[75]	a-Si	Printer paper	ITO	6.7 %
[76]	Polymer solar cells	Cellulose nanocomposite	ITO	4.98 %
[77]	OPV	Nanofiber paper	Ag NWs	3.2 %
[78]	PSCs	Cellophane paper	OMO	13.19%

7.2 Transistors

Transistors are one of the key components of modern electronics, playing the role of the main building block in countless devices from computing systems to telecommunications. The development of printed transistors on paper traditionally includes both Electrochemical Transistors (ECTs) and Field-Effect Transistors (FETs). ECTs operate by modulating ionic conductivity through an electrolyte, making them suitable for flexible and paper-based applications, but their performance is often worse than that of other transistor types. FETs, on the other hand, are a broader category of transistors in which an electric field controls the current flow through a semiconductor channel. FETs can be further classified into organic (OFETs) and inorganic types based on the materials used for the active layer. Thin-film transistors (TFTs), a subtype of FETs, are widely used in applications of flexible electronics but typically require costly fabrication techniques like sputtering and chemical vapor deposition, which limit their potential for low-cost production.

Several key challenges must be addressed to optimize the performance of printed transistors, including charge carrier mobility, the on/off current ratio (Ion/Ioff), and reducing the operating voltage. This is particularly important for low-power electronics, such as wearable devices, where minimizing operating voltages is essential for energy efficiency. However, many existing printed transistors operate at voltages of several tens of volts, which is impractical for such applications. To address this, increasing the capacitance of the dielectric material is necessary. This can be achieved by using a very thin dielectric layer with high permittivity while simultaneously preventing issues such as leakage currents or short circuits across the dielectric.

OFETs typically offer poorer performance compared to inorganic ones, especially in terms of carrier mobility and stability. However, the main advantage of organic materials is their high compatibility with traditional printing techniques. Unlike inorganic materials, their solubility makes them easier to process and integrate into printing inks, which helps lower production costs. Additionally, organic materials exhibit superior resistance to mechanical stresses typically encountered in flexible electronics, such as bending, stretching, and folding.

Concerning paper-based transistors, which is the primary focus of this review, the type of paper used -ranging from standard paper to specialized cellulose-based and nanopaper- usually affects transistor performance. Furthermore, paper is not always compatible with the typical fabrication techniques used for transistors. Consequently, it is sometimes necessary to treat the paper with coatings to prevent it from being adversely affected during the manufacturing process of these devices.

7.2.1. Paper-based transistors

As research progressed on printing transistors on plastic substrates [80], there was a growing interest in shifting toward more environmentally friendly alternatives. This led to the exploration of biodegradable substrates, with paper emerging as a promising candidate. The first transistor printed on paper was reported in 2002 by Andersson et al. [81], who demonstrated a pixel array controlled by ECTs on a polyethylene-coated cellulose-based fine paper. The reason why ECTs were the first type of transistors printed on paper is due to the resistance of this kind of device to the surface roughness of the paper substrate.

Andersson's work inspired further research into fabricating transistors on paper substrates, which led to the report of the first organic thin-film transistors (OTFTs) on paper in 2004 by Eder et al. [82]. These devices were built with a bottom-gate bottom-contact structure on commercially available hot-pressed cotton-fiber paper with a surface pre-coating treatment. The titanium gate electrode, along with the gold source and drain electrodes, was deposited through radiofrequency (RF)-sputtering, followed by patterning via photolithography and wet etching. The gate dielectric consisted of polyvinylphenol (PVP) mixed with poly(melamine-co-formaldehyde) (PMF), deposited via spin coating, thermally crosslinked, and patterned by photolithography. Pentacene was used as the organic active layer. Their device achieved a carrier mobility of 0.2 cm^2/Vs, an Ion/Ioff ratio of 10^6.

That same year, Kim et al. [83] also presented organic transistors on paper using a similar bottom-gate bottom-contact structure. In their work, a parylene protective layer was vacuum deposited to protect the substrate from subsequent wet chemical processes. Their device used Ni as the gate electrode, a dual layer of polyimide/SiO_2 as the gate dielectric, Cr/Au as the

source/drain electrodes, and P3HT as the organic material. They reported a mobility of 0.086 cm^2/Vs and an Ion/Ioff ratio of 10^4. Fabrication techniques included RF-sputtering, thermal evaporation, electron-beam deposition photolithography and wet etching for patterning.

In 2008, Fortunato et al. [84] demonstrated the first use of paper as both the dielectric and substrate for inorganic transistors. A cellulose-fiber-based paper was used without any surface treatment. On one side of the paper, a 160 nm film of indium zinc oxide (IZO) was deposited by RF sputtering as the gate electrode, while on the other side, a 40 nm gallium indium zinc oxide (GIZO) semiconductor layer was deposited. After depositing the GIZO layer, the aluminum source and drain electrodes were patterned on top of it using e-beam evaporation. The device exhibited a mobility of over 30 cm^2/Vs and an Ion/Ioff ratio of 10^4.

In 2009, Lim et al. [85] introduced an amorphous InGaZnO4 thin-film transistor on cellulose paper. They used sputtered indium tin oxide (ITO) for the gate electrode and indium zinc gallium oxide (IZGO) as the semiconductor. A mobility of 35 cm^2/Vs and an Ion/Ioff ratio of 10^4 were achieved. Also in 2009, Sun et al. [86] improved the mobility of transistors on paper with an electric double-layer (EDL) design. Their device utilized ITO electrodes, microporous SiO_2 as the gate dielectric, and Sb-doped SnO_2 as the active channel. This device exhibited a mobility of 47.3 cm^2/Vs, with a reduced threshold voltage of 0.06 V, making it compatible with low-voltage applications for wearable electronics.

In 2011, Jiang et al. [87] introduced the first in-plane TFT on paper. They demonstrated that ITO electrodes and channels could be deposited simultaneously onto paper substrates in a one-step magnetron sputtering process with a nickel shadow mask, avoiding the need for photolithography and alignment processes. Their device achieved a mobility of 22.4 cm^2/Vs using a SiO_2-based solid electrolyte.

In 2013, Huang et al. [88] demonstrated the first OFET with high transparency on nanopapers made from nanofibrillated cellulose (NFC). To maintain the transparency feature of the device they used transparent n-type organic semiconductor naphthalene tetracarboxylic diimide derivative (NTCDI-F15), single-walled carbon nanotubes (SWCNTs) as the gate electrode, and poly(methyl methacrylate) (PMMA) as the dielectric, achieving mobility of 4.3×10^{-3} cm^2/Vs and an Ion/Ioff ratio of 200. Drain and source electrodes were deposited by thermal evaporation of silver while Meyer rod coating and spin-coating methods were employed as the deposition techniques for SWCNTs and PMMA respectively. A later study carried out by Liu et al. [89] in 2015 demonstrated the fabrication of inorganic dielectric-based aluminum oxide (Al_2O_3) TFTs on photo paper substrates making use of SWCNTs as the active layer.

Subsequent studies focused on the use of nanopaper as a substrate for transistors. Gaspar et al. [90] advanced this approach by utilizing transparent nanocrystalline cellulose (NCC) as both the substrate and gate dielectric, achieving a mobility of 7 cm^2/Vs and an Ion/Ioff ratio above 10^5 on its inorganic GIZO transistors. Pereira et al. [91] showed that gate leakage current in paper-based FETs could be reduced by employing a dense microfiber/nanofiber cellulose paper as the dielectric material.

To reduce the costs of fabrication and offer low-cost production, Minari et al. [92] proposed an improved silver (Ag) nanoparticle ink, which can be printed at room temperature. Using this ink, they fabricated OTFTs by combining screen-printing, spin-coating, solution-based printing, and patterned wettability techniques on commercially available photo paper (inkjet printer paper) coated with a perylene surface layer. The estimated mobility for these devices was 2.5 cm^2/Vs. Hyun et al. [93] introduced the first foldable TFTs on paper, fabricated using low-cost techniques as well. They utilized untreated glassine paper as the substrate, screen-printed graphene electrodes in a side-gate configuration, aerosol-jet printed poly(3-hexylthiophene) (P3HT) as the semiconductor, and inkjet-printed ion-gel for the gate dielectric. The devices demonstrated excellent foldability, with stable performance maintained over 100 folding cycles, confirming their mechanical robustness and suitability for flexible electronics.

Since 2015, the use of nanomaterials and van der Waals materials, such as CNTs, graphene, and transition metal dichalcogenides (TMDs) like molybdenum disulfide (MoS_2), gained significant attention. In 2018, Sahatiya et al. [94] first reported graphene-MoS_2 FET transistors using biodegradable cellulose paper as both the substrate and gate dielectric, achieving a mobility of 18.7 cm^2/Vs. In 2020, Conti et al. [95] demonstrated MoS_2 transistors using hexagonal boron nitride (hBN) as the gate dielectric, achieving a mobility of 26 cm^2/Vs and an Ion/Ioff ratio of 5×10^4. Pimpolati et al. [96] carried out a study of the 1/f noise in bilayer MoS2 transistors on paper with inkjet-printed Ag electrodes and hBN dielectric. The results suggest that the noise properties of the investigated devices remain stable under substrate bending and are primarily influenced by the printing of the dielectric, with minimal impact from the use of the paper substrate.

Table 6 presents a comparison of the information discussed regarding paper-based transistors, while Figure 8 shows illustrative images of some of the devices mentioned.

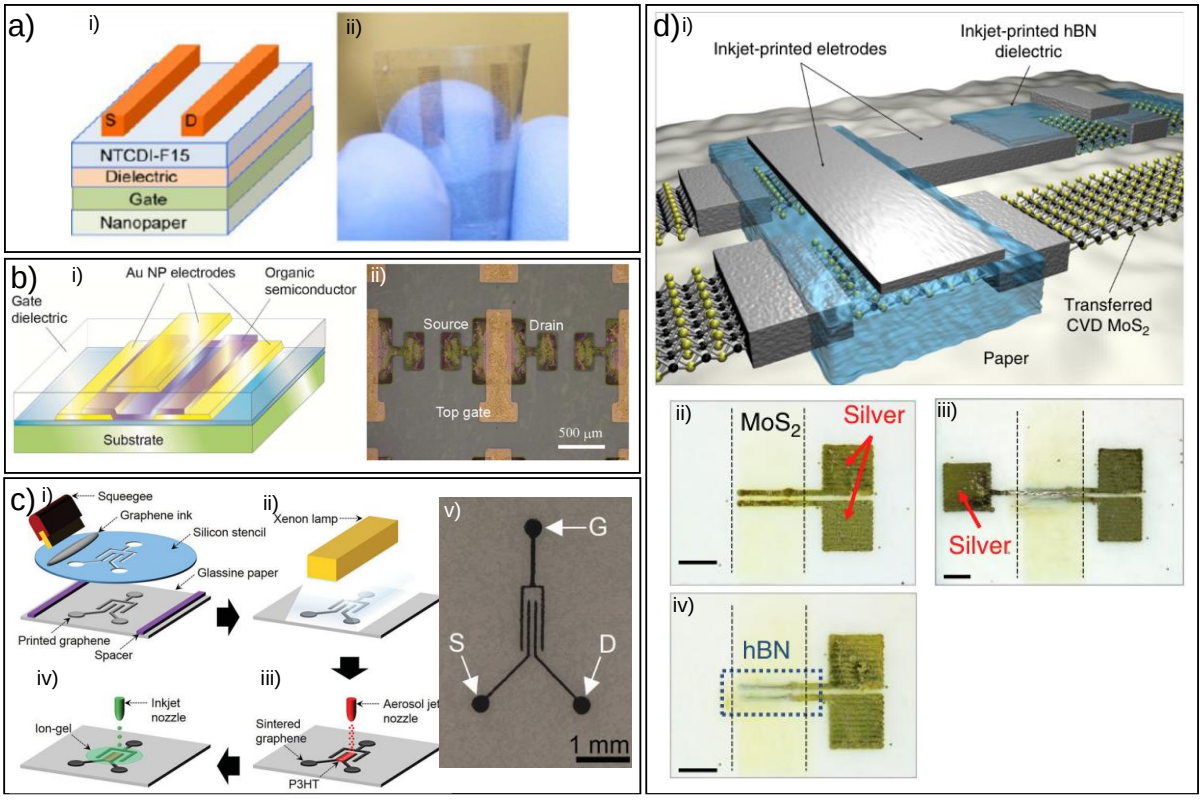

Figure 7. a) i) Schematic structure of OFET on nanopaper [88]. ii) Picture of the fabricated transistor. (Reprinted and adapted with permission from X. Fan, J. Chen, J. Yang, P. Bai, Z. Li, y Z. L. Wang, «Ultrathin, Rollable, Paper-Based Triboelectric Nanogenerator for Acoustic Energy Harvesting and Self-Powered Sound Recording», *ACS Nano*, vol. 9, n.º 4, pp. 4236-4243, abr. 2015). b) i) Schematic illustration of transistors fabricated by Minari et. al. ii) Picture of the fabricated transistors [92]. (Reprinted and adapted with permission from T. Minari *et al.*, «Room-Temperature Printing of Organic Thin-Film Transistors with π-Junction Gold Nanoparticles», *Adv. Funct. Mater.*, vol. 24, n.º 31, pp. 4886-4892, 2014, doi: 10.1002/adfm.201400169.) c) On the left, is the fabrication process for all-printed organic TFT on a glassine paper substrate, and on the right is a picture of the obtained device [93]. (Reprinted and adapted with permission from W. J. Hyun, E. B. Secor, G. A. Rojas, M. C. Hersam, L. F. Francis, y C. D. Frisbie, «All-Printed, Foldable Organic Thin-Film Transistors on Glassine Paper», *Adv. Mater.*, vol. 27, n.º 44, pp. 7058-7064, 2015, doi: 10.1002/adma.201503478.) d) i) Schematic of the inkjet-printed circuit on paper with a CVD-grown MoS_2 channel [95]. ii) Inkjet-printing of silver source and drain contacts. iii) Inkjet printing of the hBN dielectric layer. iv) Inkjet-printing of top-gate contact.

Table 9. Comparative table of paper-based transistors.

Reference	Transistor Type	Substrate	Active layer	Dielectric	Mobility (cm^2/Vs)
[81]	OTFT	Hot-pressed cotton-fiber	Pentacene	PVP mixed with PMF	0.2
[82]	OTFT	Photo paper	P3HT	Polyimide/ SiO_2	0.086
[83]	TFT	Cellulose-fiber	GIZO	(same as substrate)	< 30
[84]	TFT	Cellulose	IZGO	(same as substrate)	35
[85]	TFT	Paper	Sb-doped SnO_2	Microporous SiO_2	47.5
[86]	TFT	Paper	ITO	SiO_2	22.4
[87]	OFET	NFC	NTCDI-F15	PMMA	$4.3x10^{-3}$
[88]	TFT	Photo paper	SWCNTs	Al_2O_3	3
[89]	TFT	NCC	GIZO	NCC	7
[91]	OTFT	Photo paper	C_8-BTBT	Parylene	2.5
[92]	OTFT	Glassine paper	P3HT	Ion-gel	0.14
[93]	FET	Cellulose paper	Gr/MoS_2	Cellulose paper	18.7
[94]	TFT	Paper	MoS_2	hBN	26

7.3 Tribogenerator

Triboelectric generators (TEGs) generally involve multiple layers of polymers and conductor which differ on its tendency to lose or gain charges. This way, harvesting energy is possible from triboelectric potentials generated by different sources such as human activities, mechanical vibration, rotating tires, etc [97]. Making these generators flexible and suitable for large-scale manufacturing makes them ideal for self-powered wearable electronics. The main problem related to TEGs lies on its manufacturing process, since the output voltage depends on the contacts area, the inner layer of small TEGs (<8 cm2) requires to be micro or nanopatterned to generate high voltage peaks. At this point is when triboelectric nanogenerators (TENGs) are introduced. The TENGs' operating principle to generate electricity in an external circuit is coupling the triboelectric effect and electrostatic induction between two materials with different tribo-polarities in periodic contact and separation. The abundant oxygen atoms of cellulose allow the paper to lose electrons and positively charge easily, which makes it a perfect tribo-positive candidate material for TENGs [98].

In 2013, the first paper-based TENG was reported by Zhong, Q. et al. [99]. Their device was composed of commercial printing paper prepared in two different ways, the first one coated by

thermal evaporation of Ag and the second one spin coating of polytetrafluoroethylene (PTFE) over the already Ag coated paper. The Ag-paper and the PTFE-Ag-paper were assembled to build the paper-based nanogenerator, which was demonstrated to produce a power density of 90.6 mW cm2 at a voltage of 110 V. Chaoxing Wu et al. [100], developed an ultrasoft paper-based triboelectric nanogenerator using commercial tissue paper coated with silver nanowires (Ag NWs) (Figure 8a). Their designed P-TENG consisted on two pieces of conductive paper acting as electrodes, sandwiched between two pieces of tissue paper and a polyvinyl chloride (PVC) thin film attached to one side of the prepared Ag NW/paper. This simple structure made possible to change its size and shape without negatively impacting its proper functionality. The achieved open circuit voltage peaks (Voc) when using the P-TENG to pat different insulator materials such as glass, printer paper, cotton fabric, wood, and PET are 17, 40, 25, 20, and 100 V, respectively. When wiping the same materials, the corresponding peaks of the open-circuit voltages were measured to be around 5, 11, 4, 19, and 15 V. In [101] a paper-based triboelectric nanogenerator (TENG) for harvesting sound wave energy is presented by Xin Fan et al., providing a maximum power density of 121mW/m2 and 968 W/m3 under a sound pressure of 117 dBSPL (Figure 8b). The electricity generated can be used to charge a capacitor at a rate of 0.144 V/s. A novel cellulose paper-based drum-like triboelectric nanogenerator (D-TENG) with self-cleaning superhydrophobic features to harvest water drop energy was designed by Shuangxi Nie et al. [102] (Figure 8c). The provided output power reached 16 μW per droplet, which is more than 13.3 times that generated from other TENGs based on the electrostatic induction of water drops.

To illustrate the promising potential of TEGs as a support platform for other applications Aniket Pal et al. [103] presented a self-powered paper-based electrochemical device (SPEDs) (Figure 8d). A cellulose paper as the top layer with patterned hydrophilic domains to perform electrochemical detection with colorimetric assays was placed over a triboelectric generator (TEG). This device could harvest electric energy from the user's interaction with the SPED through a rechargeable handheld potentiostat fabricated to interface with the SPED, enabling the accurate quantitative electrochemical detection of glucose, uric acid, and l-lactate. The battery powering the potentiostat could be recharged by the user, using the sequential discharge of a capacitor previously charged with the TEG built into the SPED. TEG built into the SPED could produce peaks of ≈300 V every time it is tapped by the user. The short-circuit currents generated by the paper-based TEG reached 59 μA. The power density of the TEG reached a maximum value of 63 μWcm2 for an impedance matching load of 1.5 MΩ.

Table 10. Comparative table of paper based tribogenerators.

Reference	Type of Tribogenerator	Materials	Output Voltage	Peak Power Density
[104]	Paper-based TENG	Commercial printing paper, Ag, polytetrafluoroethylene (PTFE)	110 V	90.6 mW/cm²
[105]	Ultrasoft Paper-based TENG	Silver nanowires, tissue paper, PVC	40 V	-
[106]	Sound Wave Energy Harvesting TENG	Multiholed paper, Cu, PTFE	-	121 mW/cm²
[107]	Drum-like TENG	Cellulose paper, superhydrophobic coating	21.6 V	16 μW
[108]	Self-powered Paper-based Electrochemical Device (SPED)	Cellulose paper, hydrophobic domains, embedded TEG	170 V	63 μW/cm²

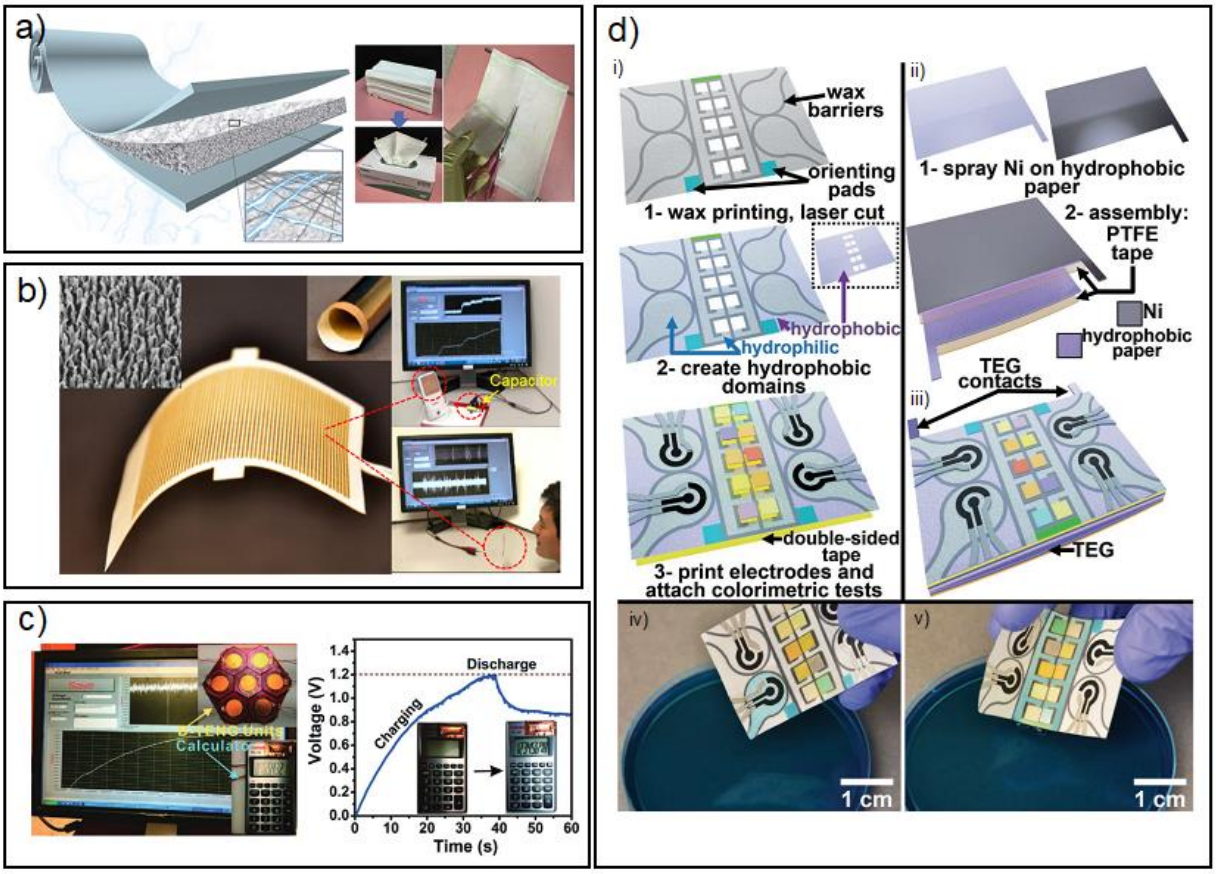

Figure 8. a) Schematic of the conductive paper layout and pictures of the P-TENG [104]. b) Picture of the paper-based TENG and its application in sound wave energy harvesting and self-powered sound recording [106]. (Reprinted and adapted with permission from X. Fan, J. Chen, J. Yang, P. Bai, Z. Li, y Z. L. Wang, «Ultrathin, Rollable, Paper-Based Triboelectric Nanogenerator for Acoustic Energy Harvesting and Self-Powered Sound Recording», *ACS Nano*, vol. 9, n.º 4, pp. 4236-4243, abr. 2015). c) Picture of the D-TENG units to power a calculator and charging and discharging processes to power the calculator [107]. d) Fabrication process and pictures of the self-powered paper-based electrochemical device (SPED) [108].

7.4 Antennas

Antennas are worldwide used in every application which requires a communication system interface, from satellites, PCs, RFID technology, etc [109]. Given its integration in such this wide range of different areas, characteristics and purposes, flexible antennas can boost the growth of this technology, making it possible to reach more applications for example the ones involving environments with more limited space in which conventional antennas cannot be used. The use of paper as a substrate for housing antennas has a lot of advantages such as its foldability, printability, lightweight, and minimal cost. However, its rough surface is the main drawback in this field as it produces significant transmission losses. For this reason, coating has been proposed as a solution to give the paper a smoother surface, with the handicap of losing foldability instead. R. A. G. de Carvalho et al. [110] reported a screen-printed paper-based and Ag-ink antenna improved with a magnetoactive layer of PVA-Fe_3O_4 into the antenna structure: This way, they improved its reflection coefficient (S11) from −8 to −56 dB and maximum transmission range from 208 m to 256 m at a 2.4 GHz tunning (used in ZigBee, BLE and WiFi communications) (Figure 9a).

Yan Wang et al. [111], using inkjet printing combined with surface modification and electroless deposition (ELD), also introduced a highly adhesive flexible metal antenna with low resistivity for RFID tags on paper substrates. A colloidal solution of hydrolyzed stannous chloride and chitosan solution was used to modify the surface and reduce the penetration rate of ink on paper at the same time as it further increased the adsorption amount of silver ions. This could create a catalytic activating layer to catalyze the subsequent ELD of a conductive deposited metal antenna (Figure 9b). A simpler approach is performed by Xiaotian Li et al. in [112], where they demonstrated the proper operation of an HF RFID antenna reader system where four loop antenna elements screen printed with Ag ink onto commercial photo paper as a substrate.

Table 11. Comparative table of paper-based antennas.

Reference	Type of Antenna	Material	Frequency Range	Key Features
[110]	Dipole	Conductive ink, Paper	2.4 GHz	Flexible, suitable for wearable applications
[111]	Microstrip Patch Antenna	Silver nanoparticle ink, dielectric substrate	300 MHz – 3 GHz (UHF Band)	High gain, low profile, used for communication devices
[112]	RFID Antenna	Graphene ink, Paper substrate	13.56 MHz (HF Band)	High conductivity, used for tracking and identification

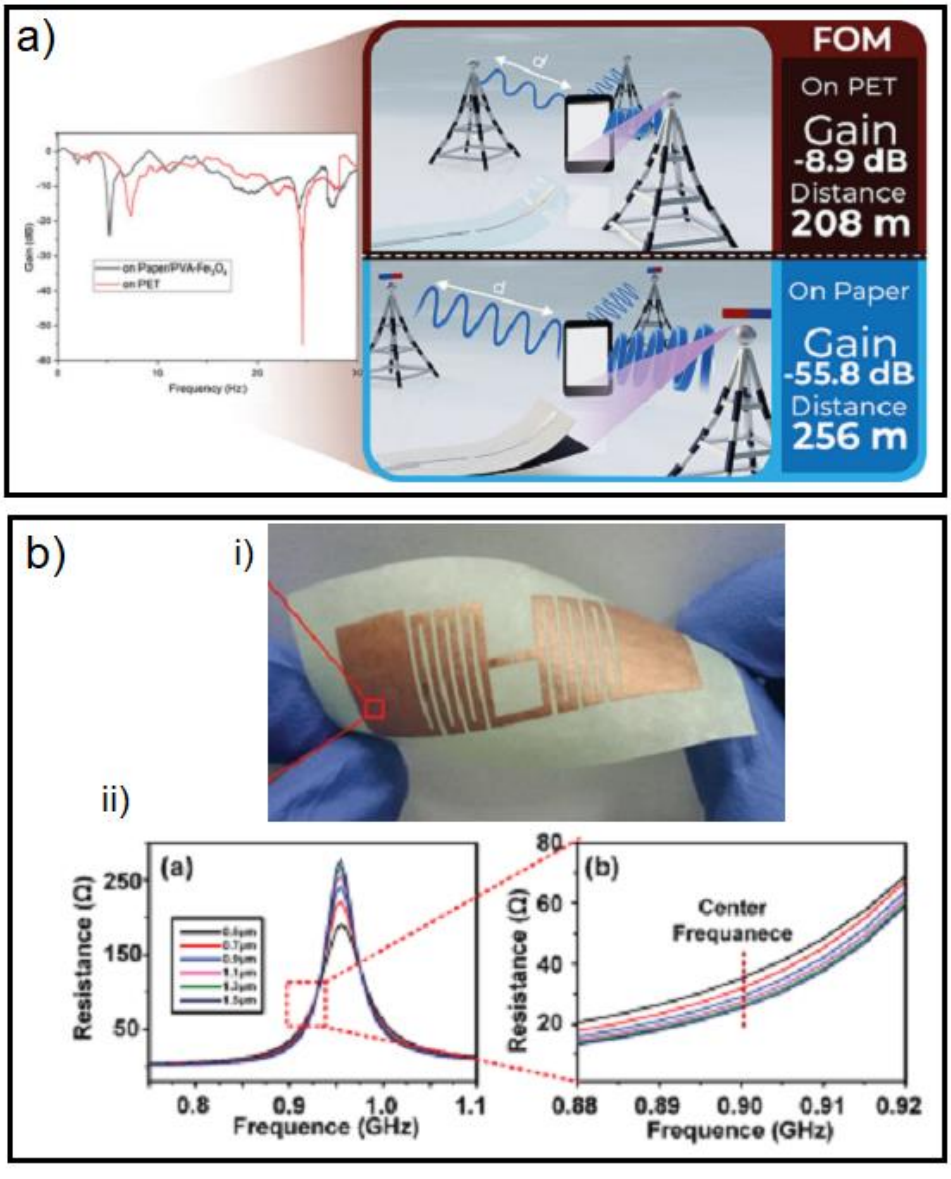

Figure 9. a) Comparative results obtained for the printed antenna on PET and Paper/PVA-Fe_3O_4, showing the best performance of the latter [110]. b) i) Image of the printed RFID tag and ii) Performance of the antenna, showing the resistance of the RFID tag with different thickness of copper layer [111].

7.5 Supercapacitors

In the field of flexible supercapacitors, cellulose has attracted widespread interest due to its rich content and its outstanding mechanical properties. In addition, supercapacitor electrodes based on cellulose paper have a controllable porous structure and distribution which is crucial for the rapid transfer of electrolyte ions during the energy storage process. This feature along with its low cost and sustainability makes paper an ideal material for its application in the supercapacitor field. The research hotspot in this field is the design and preparation of the material and structure of the electrode. As this is directly related to its electrochemical performance, it can boost the supercapacitor efficiency.

Mehmet Girayhan Say et al. demonstrated a spray-coating approach to fabricate paper PEDOT:PSS cellulose nanofibril (CNF) electrodes for printed supercapacitors [112] (Figure 10a). Their device showed an areal capacitance of 9.1 mF cm^{-2} and an equivalent series resistance (ESR) of 0.3 Ω, due to the improved contact and homogeneous electrodes. Chuanyin Xiong et al. [117] solved the problem associated to poor flexibility and mechanical strength by introducing a water-soluble synthetic polymer Poly(vinyl alcohol) (PVA) and $ZnCl_2$ into carbonized paper. The obtained supercapacitor had excellent adaptability to the environment, even in a low-temperature environment below zero, still maintaining a high volumetric specific capacitance of 3438.45 mF cm^{-3}.

A different approach is proposed by Zihang Xiong et al. [113] to improve the electrochemical performance of polypyrrole (PPy)-coated cellulose paper-based supercapacitor by increasing the reaction interface between hydrophobic pyrrole and hydrophilic $FeCl_3$ solution. A pre-treatment of paper with regenerated cellulose (RC) increased the specific surface area of the electrode, enhancing the reaction interface between pyrrole and $FeCl_3$ in the following polymerization while providing more adsorption sites for PPy. Due to the effective utilization of the chemical properties of PPy, the resulting supercapacitor assembled with these electrodes presented a high areal capacitance of 589 mF cm^{-2} at 2 mA cm^{-2}, an energy density of 33.1μWh cm^{-2} at a power density of 0.60 mW cm^{-2}. In [114] an air-laid paper is used as a flexible substrate to boost mechanical performance. PPy and reduced graphene oxide (rGO) were used as the electrochemically active materials to fabricate the flexible electrode assembled into the

supercapacitor. The developed paper electrodes had a specific capacitance of 1685 mF cm^{-2} at 2 mA cm^{-2} and the all-solid-state supercapacitor assembled was proven to provide an areal capacitance of 1408 mF cm^{-2} and a high areal energy density of147μWh cm^{-2}.

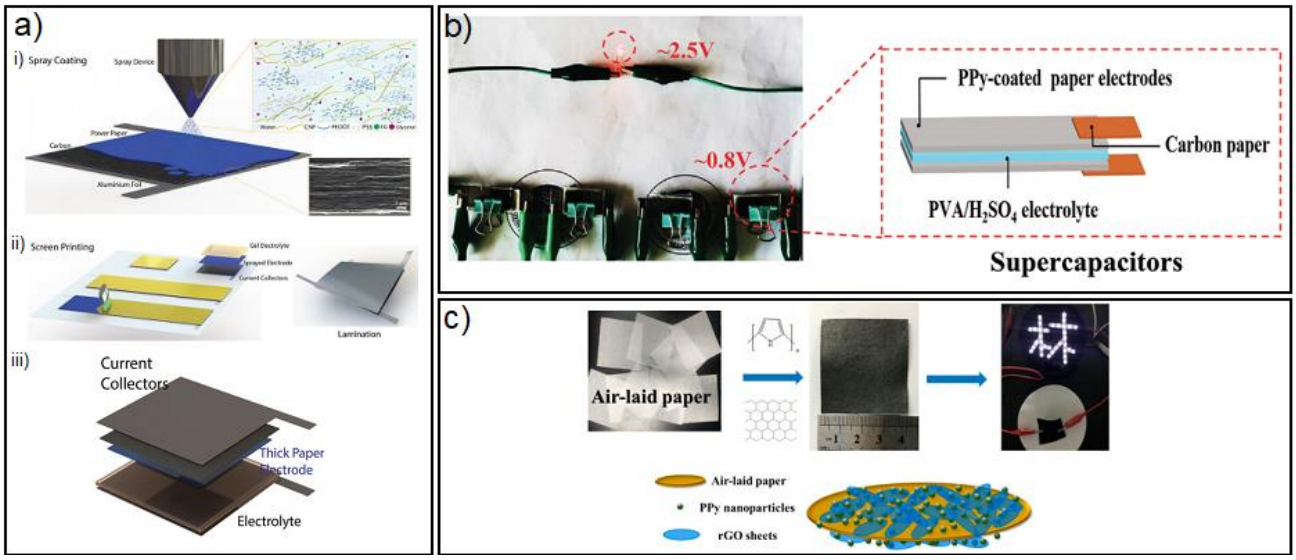

Figure 10. a) Schematic illustration of the fabrication process for the mixed ion-electron conductor paper electrode and supercapacitor [111]. i) Spray coating of the electrode, inset: cross-sectional SEM image of the spray-coated electrode ii) screen printing of the gel electrolyte and lamination of printed layers to form a flexible paper-based supercapacitor and iii) schematic representation of the spray-coated thick paper electrode. b) Four solid-state PCP-based supercapacitors [112] connected in series to light a red LED, inset: configuration of the solid-state supercapacitors fabricated by sandwiching PVA/H_2SO_4 electrolyte between two symmetric PCP electrodes. c) Schematic process of air-laid-paper-based electrodes and diagram of rGO/PPy/air-laid paper [114]. (Reprinted and adapted with permission from C. Ma, W.-T. Cao, W. Xin, J. Bian, y M.-G. Ma, «Flexible and Free-Standing Reduced Graphene Oxide and Polypyrrole Coated Air-Laid Paper-Based Supercapacitor Electrodes», *Ind. Eng. Chem. Res.*, vol. 58, n.º 27, pp. 12018-12027, jul. 2019).

Table 12. Comparative table of paper-based supercapacitors.

Reference	Type of Supercapacitor	Materials	Capacitance	Energy Density
[111]	Electrochemical Supercapacitor	Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and cellulose nanofibril (CNF)	9.1 mF/cm^2	0.453 μWh/kg

[112]	Hybrid Supercapacitor	PVA, $ZnCl_2$ and carbonized paper	5764.48 mF/cm^3	44 mWh/cm^3
[113]	Pseudocapacitor	Conductive polymers, cellulose paper	589 mF/cm^2	33.1 μWh $/cm^2$
[114]	Hybrid supercapacitor	Polypyrrole (PPy), graphene oxide (rGO) and air-laid paper	1685 mF/cm^2	1.0 mWh/cm^3

8. Conclusions and future perspectives

This review paper highlights recent advances in paper-based electronics, a promising field with the potential to move towards more sustainable and environmentally friendly approaches in electronics production and waste management. Using paper as a substrate for electronic devices has many advantages, including its low cost, biodegradability, flexibility, and ease of manufacturing. Although paper presents considerable potential as a platform for developing sensors, additional enhancements in fabrication and analysis techniques are required to reach the performance of conventional sensors available on the market. Nevertheless, the progress of paper-based sensor development is still in its early stages; hence, limitations persist in terms of sensitivity, accuracy, repeatability, and stability of paper-based sensors. In this work, the different technologies employed to produce electronic components using paper, such as printed electronics, thin-film technology, and hybrid electronics were reviewed. Special emphasis was given to the specific applications of paper-based electronics in biosensors, chemical sensors, and physical sensors.

Some shortcomings are common regarding paper in any application, such as its durability and stretchability, which are generally poor. Hence, significant efforts should be made to handle studies on this topic, as well as the optimization of the surface and mechanical properties of paper materials themselves.

Related to the field of biosensors, the paper's porosity enables capillary action, enabling fluid flow without the need for external pumps in μPADs. However, paper may not be compatible with certain chemicals or biological samples, restricting its use in some applications. To overcome this limitation, efforts are in progress to develop paper treatments or coatings that expand the range of compatible analytes and samples for uPAD applications. Additionally, as it has been shown, colorimetric and fluorescence detection methods on paper offer rapid and visual analysis of target analytes, excluding the need for complex instrumentation. However, paper properties and background interference may affect the accuracy of colorimetric and fluorescence assays, leading to false-positive or false-negative results. Achieving consistent

results across different paper-based devices can be challenging due to variations in paper properties and manufacturing processes. Therefore, establishing standardized protocols for the fabrication, calibration, and validation of these paper-based devices is essential to ensure reproducibility and reliability. Additionally, exploring the integration of digital imaging and smartphone-based analysis platforms with colorimetric and fluorescence paper-based devices for automated and quantitative analysis of results holds promise for enhancing their utility in various applications.

In the same way, the porous nature of paper also serves as an advantage for humidity sensors and energy harvesting devices, such as supercapacitors and triboelectric generators. Regarding humidity sensors, the paper has shown potential not only as a substrate but also as a humidity-sensing material. PB humidity sensors typically exhibit a notably high sensitivity, but studies reviewed in this paper have proven that its main drawback is their slow response and recovery time, which decreases the accuracy of their measurements during cyclic changes in humidity. Given this, reducing the distance between the electrodes, optimizing the structure, and reducing the thickness of humidity sensitive layer is proposed. Concerning energy harvesting devices, paper's pores have a significant surface area, which can enhance energy capture and charge transfer in such devices. The main challenges concerning them areas mentioned, are related to enhancing the durability and mechanical strength of paper to ensure its long-term performance under various environmental and usage conditions. Researching materials and coatings that can improve resistance to other adverse environmental factors would thereby increase the lifespan and reliability of the devices.

However, the paper's porousness does not always help, as in the case of temperature sensors and antennas, where coating techniques are generally used to improve the behavior of the paper in such applications. For this reason, developing advanced deposition techniques or surface treatments to enhance the smoothness and uniformity of paper substrates might be needed, aiming to improve devices' accuracy and reliability. Variations in paper composition, thickness, and environmental factors like humidity can pose challenges in achieving consistent calibration and accurate temperature measurements. On the other hand, paper-based antennas usually have limited bandwidth and operating frequencies because of material constraints and fabrication techniques, so researching conductive ink formulations or incorporating conductive additives into paper substrates to enhance conductivity, enabling improved antenna performance and signal transmission should be considered.

Concerning pressure and strain paper-based sensors, the research included in this paper presents lower sensitivity compared to traditional strain gauges, limiting their ability to detect very small pressures or strains.

Finally, related to the field of solar cells, a wide range of possibilities opens regarding the use of paper. Focusing on the development of good electrodes, the main approaches include optimization of bottom electrode deposition avoiding or exploiting the surface structure, modification of the paper surface by using planarization layers, use of nanocellulose substrate with smooth surface, and development of conductive cellulose substrates.

In summary, even though paper-based electronics presents exciting new directions for environmentally friendly electronics, addressing issues with sensitivity, uniformity and durability remains crucial for the growth of this field. Despite these challenges, ongoing research and development efforts continue to push the boundaries of what paper-based electronics can achieve.

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