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Digitally Printed Eco-Friendly Electronic Devices

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ABSTRACT

The sustainable life cycle of electronics is one of the major challenges of modern society. Moreover, the manufacturing of conventional electronics significantly impacts the environment due to the extensive consumption of raw materials, energy, and the release of harmful byproducts. In this regard, manufacturing of electronics using eco-friendly materials through digital printing routes represents a promising and sustainable solution. This review discusses the state-of-the-art research in this field, exploring various digital printing techniques such as inkjet, electro–hydro–dynamic jet, aerosol-jet, and extrusion printing techniques, focusing on resource efficient manufacturing with minimal material wastage and their environmental impacts. Various eco-friendly ink formulation strategies are discussed together with their practical application. Additionally, the review addresses various end-of-life management strategies such as repair, recycling, and biodegradation. Finally, major challenges associated with digital printing of eco-friendly electronics to compete with conventional electronics are explored and future perspectives in this emerging area are outlined.

1 | Introduction

Technologies related to the fields of flexible, wearable electronics, remote sensing, and robotic systems are set to revolutionize the way we interact with each other, systems and devices in the coming years [1–5]. However, these technologies still rely heavily on conventional microfabrication methods that are material-intensive, energy-demanding, and dependent on hazardous chemicals, introducing significant environmental burdens even before a device reaches its end of life [6–8]. As the demand for these technologies increases, device manufacturing is also expected to rise sharply, further amplifying these fabrication-related impacts. Additionally, current trends and envisioned application scenarios of these technologies will predictably continue promoting the design of products with short lifecycles [9–13]. These trends, have currently produced an unsustainable production of electronic waste (e-waste) which is only partially recovered, reused, or recycled. According to the *Global E-waste Monitor (2024)*, 62 million metric tons (MMT) of e-waste were generated globally in 2022, while only the 22.3% (13.8 MMT) of the mass was properly collected and recycled. Projections estimate that by 2030 global e-waste generation will reach 82 MMT, where the formal recycling rate is predicted to decrease to 20% and at ambitious scenario of policy changes could potentially raise this recycling rate to 60% [14]. Furthermore, untreated e-waste is commonly transferred to countries where it is loosely regulated, generating disastrous socioeconomic, environmental and healthcare consequences for their inhabitants [15, 16].

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The most direct way to mitigate negative sustainability issues in most end-of-life phases would be the utilization of inherently sustainable materials and processes from the very conception of a technology. This approach could decisively contribute to more sustainable electronic applications which simplify, reduce, or eliminate the economic, energetic, and environmental burden on regulation, recovery, or recycling of e-waste [17–20]. During the past decade, the scientific community has increasingly committed to investigate new material concepts for creating more sustainable electronic systems [20–26]. Naturally and synthetically acquired biodegradable materials as well as byproducts of agriculture or the food industry have been investigated as passive and active components in various electronic devices [27–33]. Materials such as silk, paper, and cellulose diacetate have been used as substrate alternatives to plastic foils [34–38]. Biodegradable materials with semiconducting, dielectric, and/or electrochemical properties (e.g., DNA, β-carotene, keratin, gelatin, etc.) have enabled transistors, sensors, batteries, actuators, and light-emitting devices with reduced environmental impact [39–49]. These devices have just started to show the possibilities that eco-friendly functional materials could offer in the fields of sensing, security, packaging, or bioelectronics at a lab scale. However, the development of truly sustainable processing methods translatable to economically viable fabrication schemes is indispensable to ensure the sustainability of the entire value chain.

For sustainable manufacturing, digital printing fabrication methods offer an effective alternative to conventional manufacturing, that can provide the personalization, cost-efficiency, and high throughput needed in thin-film electronics applied in fields such as wearables, sensing, healthcare, or robotics [50–52]. These digital techniques are characterized by intrinsic sustainability advantages. For example, they are highly resource efficient as the material is only deposited where and when needed, thus eliminating waste associated with overproduction. By being digital, all processes to fabricate a given device use the same equipment, decreasing the use of additional physical space or the need for fabricating further forms, masks, or molds. As solution-based methods, they typically consume less energy compared to thermal evaporation-based processes in vacuum. Furthermore, there is a large variety of green solvents available from other chemical industries which are currently being investigated in printed electronics [53–58]. Finally, the industrial maturity of the techniques can facilitate the eventual technology transfer of research results without investing time and energy in establishing additional production steps and approaches [59].

Advances have been demonstrated in vacuum-processed and lab-scale solution-processed eco-friendly devices, whereas printing technology has progressed more slowly. This is due to the additional challenges posed by printing processing, which requires ink formulations and postprocessing suitable for the specific technique that yield precise control of film homogeneity, micromorphology, and interfaces that promote efficient optical, electronic, or electrochemical processes [60–67]. These challenges lead to interesting scientific questions and engineering challenges to develop technologies that reach an optimum compromise between processability and performance, which adds to the compromise of already using eco-friendly materials presenting lower performance than their conventional counterparts.

This review highlights the recent advances in eco-friendly electronics enabled by digital printing techniques, with a particular focus on research developments from the past decade. It complements existing reviews on eco-friendly electronics [6, 29, 68, 69–73], which predominantly emphasize material selection, device concepts, or end-of-life management. By narrowing the scope to digitally printed electronic devices, this work provides a distinct perspective on how additive, printing-based fabrication methods can enable environmentally sustainable electronic systems across the entire lifecycle, from manufacturing to end-of-life management. Within the scope of this review, "biodegradable" refers to materials or devices capable of undergoing biologically mediated degradation, whereas "transient electronics" denote systems designed to physically dissolve or disintegrate after a defined operational period under specific environmental triggers. The term "green" is used specifically for the choice of chemistry and materials, for example, using renewable feedstocks and synthesis routes with high atom economy. The term "eco-friendly" is used in this review to describe materials that avoid hazardous substances and reduce the generation of harmful by-products. The term "sustainable" is used more broadly, to describe fabrication approaches that reduce environmental impact through resource- and energy-efficient processing, or responsible end-of-life pathways [74].

The review firstly discusses major digital printing techniques, including inkjet, electro–hydro–dynamic jet (EHD), aerosol-jet (AJP), and extrusion-based printing, with a focus on their inherent sustainability advantages and limitations. Eco-friendly materials and the corresponding ink formulations are subsequently reviewed, covering green solvents, eco-friendly functional materials, and additives. Various potential applications of such printed materials, including active and passive devices, sensing, actuation, energy harvesting, and storage, were comprehensively reviewed. We further discuss various end-of-life management strategies widely employed in the field. Furthermore, we identify critical research gaps and opportunities, discussing possible future strategies and end-of-life scenarios to offer an outlook for the development of truly eco-friendly digitally fabricated electronics. Figure 1 provides a schematic overview of digitally printed electronics. The coverage of this topic is timely given the rapidly growing issues associated with resource-intensive electronic fabrication practices and the management of e-waste. This review provides researchers and practitioners across academia and industry with a critical evaluation of current trade-offs and a prioritized roadmap for advancing digitally printed eco-friendly electronics toward industrially relevant, environmentally responsible technologies.

2 | Digital Printing Techniques for Eco-Friendly Electronic Manufacturing

Digital printing techniques enable the direct deposition of materials in computer-defined patterns onto a wide range of substrates. As outlined above, these techniques form the foundation for the eco-friendly devices discussed throughout this review. These

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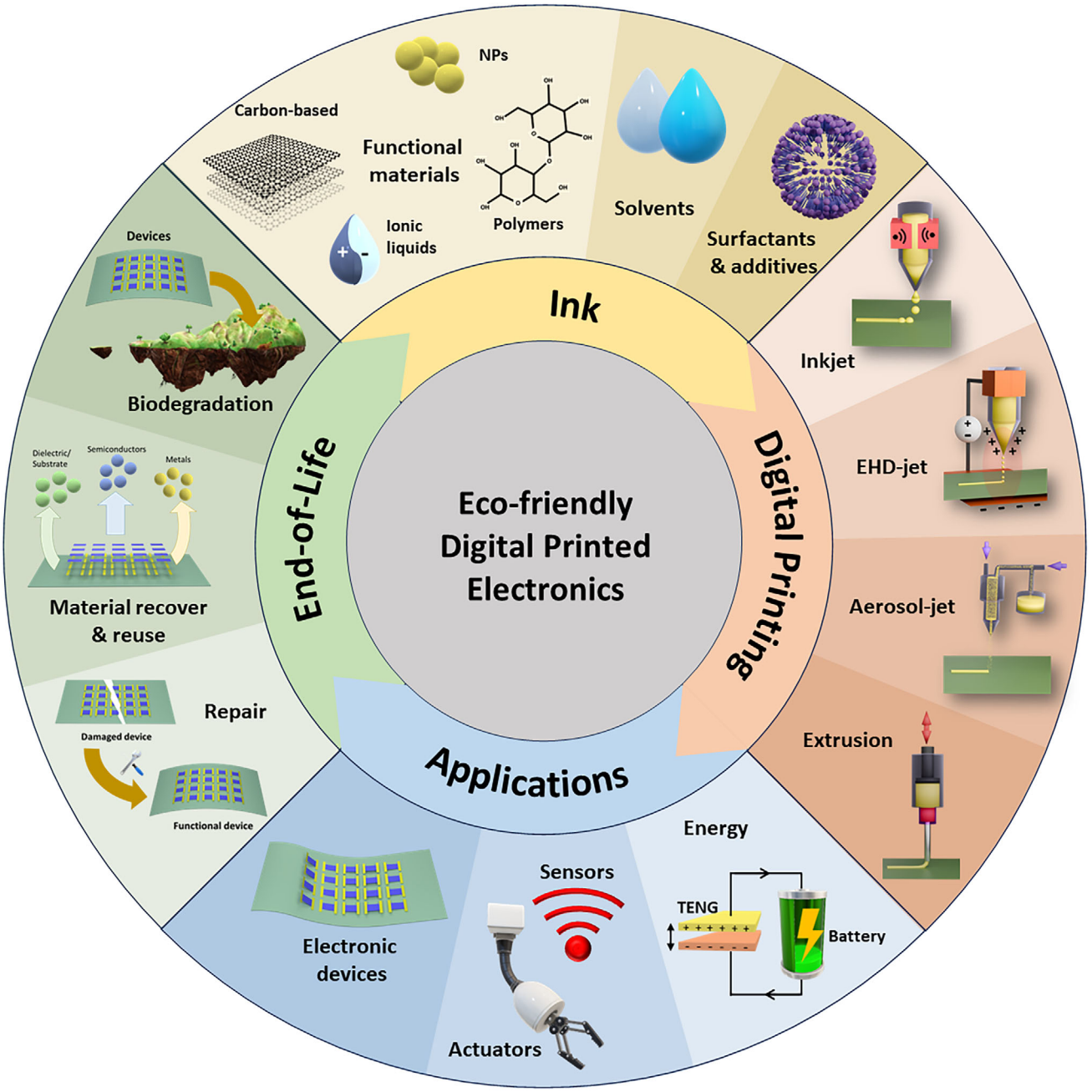


FIGURE 1 | Overview of eco-friendly digitally printed electronics, highlighting key aspects including green ink formulations based on sustainable materials, resource-efficient digital printing processes, potential applications, and end-of-life management strategies for devices after they become nonfunctional.

methods eliminate the need for physical printing forms, masks, or subtractive steps typical for conventional printing technologies, or lithographic techniques, thereby reducing material waste, processing time and complexity, and energy consumption. The most widely used techniques for the manufacturing of (opto)electronic devices and sensors include inkjet, electrohydrodynamic jet, aerosol jet, and extrusion printing (Figure 2) [72, 73, 75, 76]. The sustainable digital printing of electronics demands not only the use of eco-friendly materials, green solvents and low processing temperatures but an integrated approach that considers functional performance and environmental impact across all stages of the fabrication process. The process typically begins by selecting a printing technology based on the suitability of the functional materials to be formulated as inks within the technique's processing window, together with the substrate in which printing will occur. This section reviews relevant digital printing techniques, emphasizing their processing window, and sustainability aspects.

2.1 | Inkjet Printing

Inkjet printing is a widely used digital printing technique in which small droplets of functional ink are ejected from a

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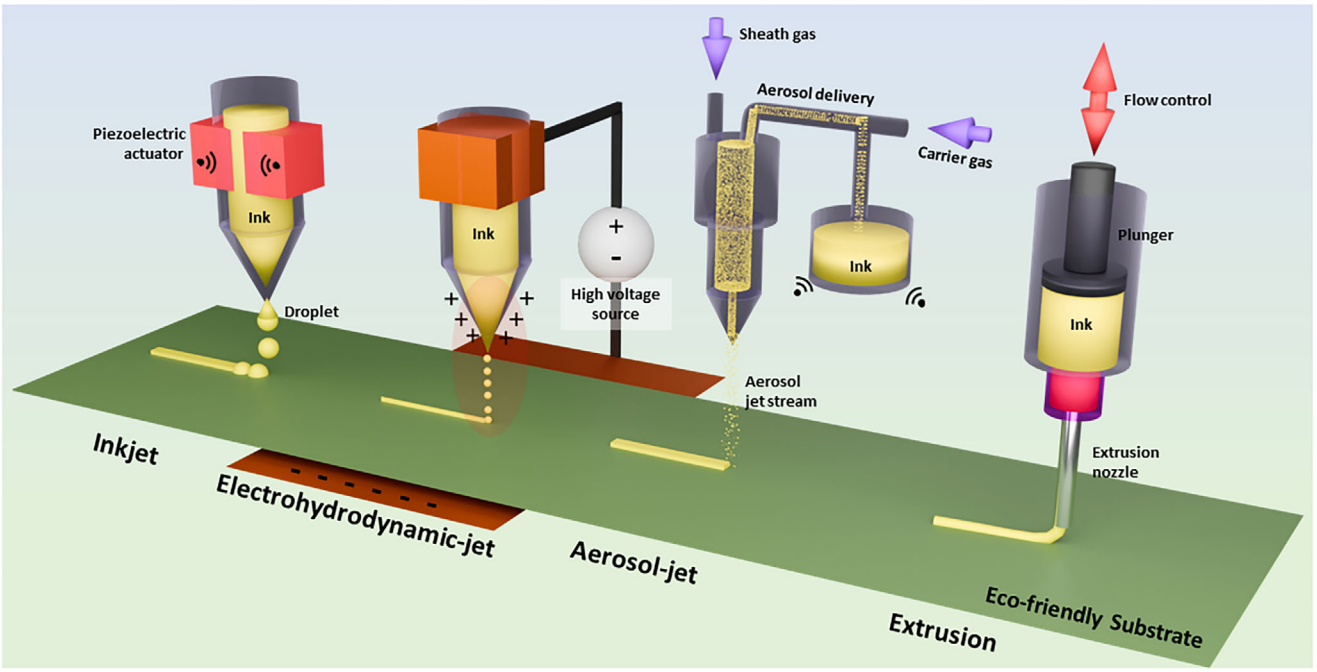


FIGURE 2 | Schematic illustration of various digital printing techniques: inkjet printing, employing piezoelectric printheads for controlled droplet generation; electrohydrodynamic-jet printing, where a high electric field induces jet formation from a Taylor cone; aerosol-jet printing, in which atomized ink droplets are transported and focused using carrier and sheath gases; and direct ink writing, an extrusion-based printing technique in which viscous inks are deposited through a nozzle via pressure-driven flow.

printhead onto a substrate in a noncontact manner. It supports line resolutions of 30–50 μm, depending on printhead specifications, and enables controlled film thicknesses from ~100 nm to several micrometers owing to precise, low-volume deposition [75]. Based on droplet ejection control, inkjet systems can be classified into continuous inkjet (CIJ), or drop-on-demand (DoD) systems [72]. In CIJ, a continuous stream of droplets is generated and unused droplets are redirected into a gutter for recycling whereas in DoD droplets are produced only when required, reducing material waste and energy consumption, therefore being considered the more sustainable option.

Ink formulation plays a central role in determining the electrical and mechanical performance of inkjet printed films [58]. A variety of functional additives such as nanoparticles (NPs) are frequently incorporated to introduce electrical, optical, or other functional properties [72]. Examples include metallic NPs, polymers, carbon-based materials, and ionic liquids, which are discussed in detail in the following section. The nozzle diameter dictates droplet size and imposes a critical limit on the maximum allowable particle dimension in the ink. For example, dispersed particles should be no larger than ~1/50 th of the nozzle diameter to avoid clogging and unstable jetting [77, 78]. Solvent choice governs ink volatility, drying kinetics, and film uniformity, with increasing emphasis on low-toxicity, water-based, or bio-derived formulations to support eco-friendly processing. Key parameters, such as viscosity, surface tension, density, and pH, can critically influence droplet formation, spreading, and final film properties [79]. For inkjet processes, viscosity between 2 and 15 mPa.s typically enables stable droplet generation, values below this range can cause uncontrolled flow, while higher values hinder droplet ejection [77]. Temperature control is commonly used to adjust viscosity. Surface tension determines how easily an ink forms droplets and should typically be ≥35 mN m^{-1} [77, 80]. Ink jetting behavior can be described using dimensionless numbers, such as Ohnesorge (Oh), Weber (We), and Reynolds (Re), which are functions of viscosity, surface tension, density, and nozzle diameter [81]. The Oh, which represents the ratio of viscous to surface tension forces, should lie between 0.1 and 1 for inkjet printing. If Oh < 0.1, satellite droplets may form, while Oh > 1 indicates excessive viscosity that hinders droplet ejection [82]. The Re relates inertial to viscous forces. For successful ejection, the condition (Oh(Re) ≤ 2) ensures sufficient kinetic energy to overcome surface tension. After ejection, the droplet should land without splashing, which occurs when $Oh(Re)^{(5/4)} \geq 50$ [83]. Therefore, stable inkjet printing requires ink properties to be formulated within these parameter limits. Secondary components such as surfactants, dispersants, and rheology modifiers are often introduced to regulate ink stability and substrate wetting. However, excessive use may leave insulating residues or persistent byproducts, with implications for both device performance and environmental impact. Therefore, eco-friendly formulation design requires balancing printability, functionality, and ecological safety. In DoD systems, the printhead generates droplets by inducing pressure pulses within the ink chamber via electrostatic, piezoelectric, or thermal mechanisms [72]. Once ejected, the ink-substrate interactions govern the quality of film formation [84]. Surface hydrophilicity, for example, affects spreading and adhesion, while nonuniform drying phenomena such as the "coffee-ring effect" can result in inhomogeneous particle distribution [85]. Careful tuning of ink composition and substrate surface energy is therefore required to achieve uniform films. Post-deposition treatments, including thermal annealing,

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Ultraviolet (UV) curing, or chemical sintering, are often necessary to improve film continuity, remove solvent residues, and activate functional materials. To align with sustainable processing, these steps should minimize energy consumption and remain compatible with biodegradable or recyclable substrates.

2.2 | Electrohydrodynamic-Jet Printing

In EHD printing, droplets are generated not by extrusion pressure, as in inkjet printing, but by applying a strong electric field that draws ink from the nozzle toward the substrate. This technique offers sub-micrometer resolution and accommodates a wide viscosity range (1–10^5 mPa.s) [86]. The resolution arises from three factors: (i) the use of nozzles with smaller inner diameters (<100 nm) than those in conventional inkjet printing, which are micro-sized, (ii) the ability of electrohydrodynamic forces to generate droplets smaller than the nozzle itself, and (iii) the electrical-field induced focusing effect of the applied electric field, which enables precise deposition with minimal lateral spreading [87].

The basic principle of EHD printing is conceptually simple; however, the process is highly complex and strongly dependent on ink physicochemical properties. The ink experiences extreme stretching and shearing force under high electric field and hence the dynamic rheological properties are important [88]. Viscosity and viscoelasticity mainly control jet stability and deformation. The electric field must overcome viscous resistance to form a stable jet. Higher viscosity generally improves jet stability and produces more uniform droplets, while very low viscosity can lead to unstable jets and spreading after deposition. Viscoelasticity also plays an important role, moderate elasticity helps stabilize the jet and widens the operating voltage range, but very high elasticity can cause jet thickening or loss of resolution [88, 89]. Surface tension, on the other hand, controls jet initiation and droplet breakup. The applied electric field must overcome surface tension to form the Taylor cone and initiate jetting [90]. Lower surface tension facilitates jet formation and can support continuous jetting, while higher surface tension resists jet formation and may even prevent it. However, if surface tension is too low, satellite droplets can form and reduce printing accuracy [89, 91]. Therefore, careful selection of surfactants, and solvents is essential. Ink conductivity also plays a decisive role, since charge mobility governs key parameters such as Taylor cone formation time, charge relaxation time, and the voltage required to initiate flow [90]. To enhance ionic or electronic conductivity, dilute acids, salts, or other charge carriers are often added. Optimization of EHD printing conditions is commonly guided by the ratio of charge to hydrodynamic relaxation time. When this ratio is below unity, the fluid contains sufficient charge carriers with high electrical mobility and flows slowly enough to maintain quasi-equilibrium, thereby enabling stable jetting. Thus, precise control over ink formulation, together with careful adjustment of system parameters such as flow rate, applied voltage, nozzle size, and standoff height, is crucial to achieving high-resolution, uniform, and reproducible printing. Despite its outstanding resolution, the scalability and throughput of EHD printing remain limited, making it most suitable for high-value applications requiring nanoscale precision [92].

2.3 | Aerosol-Jet Printing

AJP is a digital printing technique that utilizes atomization and a stream of inert gas to deposit inks as focused aerosol sprays, being capable of producing features with resolutions below 10 μm [93]. In this method, functional materials are dispersed in solvents to prepare inks with viscosities ranging from 1 to 1000 mPa.s. The ink is atomized into fine droplets using high-frequency ultrasound (typically in MHz range), which induces capillary waves at the ink surface and generates aerosols of uniform dimension [94]. The ink is then transported by a carrier gas (typically N_2) toward the nozzle (atomizer flow). At the nozzle, a sheath gas surrounds the aerosol stream, preventing droplet accumulation on the nozzle walls and focusing the stream onto the substrate (sheath flow). For selective deposition, a mechanical shutter can interrupt the aerosol flow when needed, although this makes the process more material and energy consuming compared to other digital printing techniques. Due to its relatively large standoff distance (>3 mm) and the jetting behavior of the aerosol stream, AJP can conformally print on nonplanar surfaces with complex curvatures, high roughness, or multidirectional geometries. Printing is achieved by moving either the nozzle or the substrate stage, enabling the deposition of both fine lines and complex patterns [93].

AJP relies on a high-velocity carrier gas to atomize inks into fine aerosol droplets, imposing distinct rheological requirements compared with inkjet printing. Printable inks must satisfy three key criteria: appropriate viscosity, shear-thinning behavior, and controlled surface tension [95]. AJP is compatible with a broad viscosity range, where low viscosity (<10 mPa.s) facilitates atomization but may lead to overspray, while high viscosity (>500 mPa.s) can hinder droplet formation and stability [95, 96]. Effective atomization requires inertial forces to overcome surface tension, and the resulting droplet size is primarily governed by surface tension, density, and ultrasonic frequency, while viscosity controls capillary wave growth. Shear-thinning behavior is advantageous, as it promotes droplet breakup under shear while maintaining dispersion stability at rest. Surface tension must be carefully balanced (typically 25 to 40 mN m^{-1}) to enable efficient atomization and controlled spreading after deposition [97]. Solvent selection is critical to ensure stable dispersion of functional materials [94, 98]. During transport to the printhead, droplets repeatedly interact with dry gas streams, which can accelerate solvent evaporation and lead to significant droplet shrinkage or premature drying. To mitigate this, a low-volatility co-solvent (~10%) is often added to stabilize the droplet size during flight [99]. However, additional challenges arise when droplets collide with internal walls, causing partial deposition inside the printing system [94]. Over time, this accumulation can lead to process drift, contamination, or even clogging of the printhead, ultimately reducing printing reliability and lifetime. Additionally, it also results in material waste, lowering the overall sustainability of the process. These limitations contribute to the relatively low scalability of AJP, as extended operation is prone to inconsistencies and deviations in performance. As a solution, recently, an unconventional aerosol-on-demand technique has been demonstrated, in which ink atomization occurs directly inside the printhead. By generating aerosol only when required, this approach eliminates the need for mechanical shutters for

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selective flow control and significantly reduces material waste, making the technique more advantageous for future aerosol-jet applications [100].

2.4 | Extrusion Printing

Extrusion printing is another popular digital printing technique, that generally comprises two main approaches: fused deposition modeling (FDM) and direct ink writing (DIW) [101]. In FDM, thermoplastic filaments are heated and extruded layer by layer to fabricate 3D objects. By contrast, DIW involves the controlled extrusion of liquid-phase inks or pastes through fine nozzles, enabling the precise deposition of functional materials. While FDM is widely employed for additive manufacturing of structural 3D objects, DIW has gained prominence for the fabrication of electronic devices and functional architectures. DIW typically supports resolutions in the range of 100 µm, depending on nozzle diameter and ink rheology, while film thicknesses span from 10 µm and above. Compared to inkjet or aerosol jet printing, DIW enables the direct fabrication of thicker films and 3D architectures, making it particularly well suited for applications requiring mechanical robustness or high material loading. DIW also supports multinozzle configurations for multimaterial extrusion, which facilitates the rapid fabrication of complex, heterogeneous, and layered devices [102].

The extrusion mechanism in DIW can be pneumatic or mechanical. Pneumatic systems use compressed gas to regulate ink flow, while mechanical systems employ pistons or screws for displacement-controlled extrusion [103]. These mechanisms enable DIW to process materials across a substantially broader rheological window than inkjet, AJP, or EHD systems. Consequently, DIW can accommodate highly concentrated suspensions, pastes, and gels with higher viscosities exceeding 10^6 mPa·s under relevant shear conditions. This versatility allows the use of colloidal suspensions, polymers, composites, and even high-aspect-ratio nanomaterials such as carbon nanotubes (CNTs) and metallic nanowires, which often clog finer nozzles in other printing techniques [104–106].

Although DIW can accommodate highly viscous inks, not all material formulations are inherently printable, and their rheological properties must be carefully engineered to enable stable extrusion and reliable pattern formation [101]. A DIW printable ink must exhibit an elasto–visco–plastic behavior rather than pure viscous or viscoelastic characteristics, it should flow smoothly under applied pressure yet retain sufficient structural integrity to maintain its shape after deposition. A finite yield stress is essential to prevent flow under gravity and maintain the shape of deposited features. At the same time, a shear-thinning behavior facilitates extrusion through the nozzle by reducing viscosity under applied pressure. Furthermore, an appropriate balance between the elastic and viscous responses, commonly described by the storage modulus (G′) and loss modulus (G″), is critical. Rapid structural recovery after extrusion is essential to ensure shape retention and minimize the material spreading [106–109]. Therefore, recent DIW printability frameworks increasingly move beyond simple viscosity ranges and consider coupled material-process parameters, including yield stress, shear-thinning behavior, viscoelastic recovery, filament continuity, shape fidelity, and interlayer buildability. The nozzle diameter determines both the resolution and the maximum particle size in the ink. High solid loading is often required to minimize drying-induced shrinkage and cracking, while the resulting particle network must withstand capillary stresses during solvent evaporation. As in other printing techniques, additives such as dispersants, binders, and rheology modifiers are commonly employed to stabilize suspensions, tailor yield stress, enhance shear-thinning behavior, and promote structural recovery, while preventing phase separation [101]. However, the choice of these additives must consider both printability and sustainability, as excessive use of synthetic binders may hinder recyclability or biodegradability.

2.5 | Comparison of Digital Printing Techniques

Table 1 summarizes the functional characteristics of the various digital printing techniques.

Printability requirements: Printability is governed by the interplay between ink formulation and the underlying deposition mechanism, and these requirements differ significantly across techniques. Inkjet printing relies on relies on low-viscosity, near-Newtonian fluids that can be ejected as droplets within a narrow rheological window. This constraint limits material loading and demands precise control over solvent volatility, surface tension, and viscosity to avoid nozzle clogging or satellite droplet formation. By contrast, EHD depends on electrohydrodynamic forces to form a stable Taylor cone from which ultrafine jets are ejected. Here, viscosity, viscoelasticity, surface tension, and electrical conductivity must be carefully balanced to enable jet stability and prevent satellite droplets or unstable jetting. AJP is controlled by atomization and gas-driven transport, requiring inks that can efficiently form aerosols under shear while maintaining dispersion stability. Viscosity, surface tension, and shear-thinning behavior govern droplet breakup and transport fidelity. DIW accommodates highly viscous, particle-rich pastes and gels with yield-stress behavior, enabling shape retention after deposition. This versatility allows for concentrated inks, including those with high-aspect-ratio nanomaterials. Thus, while low-viscosity techniques (inkjet, EHD) prioritize fluid dynamics and jet stability, high-viscosity methods (DIW) rely on rheological structuring, and intermediate methods (AJP) balance both regimes. This highlights a fundamental trade-off that increasing formulation flexibility generally comes at the expense of resolution and process simplicity.

Resolution: Printing resolution is primarily governed by the mechanism used to generate and control material deposition. EHD printing achieves the highest resolution (<1 µm) due to electrically driven jet thinning and field focusing, which enable droplets smaller than the nozzle itself. However, this precision comes at the cost of process robustness and throughput, as stable jet formation requires high electric fields and low flow rates. Inkjet offers moderate resolution (20–50 µm), constrained by droplet size and fluid dynamics, while maintaining stable and reproducible operation. AJP improves resolution compared to inkjet by focusing aerosol streams, but this depends strongly on atom-

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TABLE 1 | Functional characteristics comparison of various digital printing techniques.

Technique	Viscosity range	Resolution	Film thickness	Scalability	Printing speed	Suitable materials	Printability requirements
Inkjet	2–20 mPa·s	20–50 µm	Thin films (~100 nm – few µm)	High	High (Multinozzle arrays)	Small molecules, polymers, NPs, bio-inks	Stable droplet formation Clog-free
EHD	1–10,000 mPa·s	<1 µm	Ultrathin, precise films (tens of nm – µm)	Low	Low (1–10 Hz jetting, serial writing)	NP inks, conductive polymers, biomaterials	Stable tailor cone Controlled EHD jetting
Aerosol Jet	1–1000 mPa·s	10–120 µm	Thin to medium (~100 nm – 10 µm)	Medium	Moderate (tens of mm/s, limited by atomization)	Low-aspect ratio CNT dispersions, metal NPs, nanowires (NWs), polymers	Efficient atomization, stable aerosol, no agglomeration
Extrusion (DIW)	10^2–10^6 mPa·s	100–300 µm	Thick films (>10 µm – mm)	Medium	Low–Moderate (1–10 mm/s)	High-aspect ratio CNTs, graphene pastes, biopolymers	Yield stress + shear-thinning; rapid post-extrusion recovery

ization quality and gas flow control. By contrast, extrusion-based DIW is fundamentally limited by nozzle diameter and ink rheology, resulting in the lowest resolution. Thus, higher resolution typically demands tighter process control and reduced material throughput, highlighting a direct trade-off between precision and productivity.

Scalability: Inkjet printing is the most scalable due to its use of multinozzle arrays and drop-on-demand operation, enabling high-speed, large-area fabrication. By contrast, EHD printing is inherently limited by its serial nature and low jetting frequency, making large-area manufacturing challenging despite its high precision. AJP offers moderate scalability, faces challenges related to atomization efficiency, carrier-gas-induced material loss, and wall deposition within the printhead, which can introduce process drift and reduce reliability during extended operation. DIW scales well for thick films and 3D structures but is constrained by slow deposition speeds due to viscous ink formulations and mechanical extrusion mechanisms. Overall, techniques that achieve high resolution tend to sacrifice scalability, whereas highly scalable methods impose stricter material and processing constraints.

Sustainability: From a sustainability perspective, material efficiency, energy consumption, and process complexity must all be considered. Inkjet printing is generally the most material-efficient due to drop-on-demand deposition, though its reliance on dilute inks can increase solvent use and drying energy. EHD printing minimizes material waste through precise deposition, but requires high electric fields, increasing energy demand per printed feature and reducing overall efficiency due to low throughput. AJP introduces additional material loss during atomization and transport, as well as higher energy use from carrier and sheath gases, reducing its sustainability despite good material compatibility. DIW enables the use of highly concentrated and bio-based inks, which can lower solvent usage and support sustainable materials; however, its lower resolution reduces material efficiency in fine-feature applications, and mechanical extrusion increases energy demand. Therefore, sustainability does not align with a single technique but emerges from balancing material use, energy input, and process efficiency, depending on the application.

3 | Sustainable Materials for Digital Printing

Thus far, we have analyzed the trade-offs among various digital printing techniques. While each offers unique advantages for fabricating functional devices, their potential in sustainable electronics depends not only on process efficiency but also on the materials used. Unfortunately, many conventional inks and precursors rely on nonrenewable, toxic, or energy-intensive materials, posing significant sustainability challenges. In this section, we critically examine the traditional materials commonly used in digital printing, alongside their limitations and commented on the sustainable materials that are commonly used in the different applications. This discussion paves the way for Section 4, where we will focus on specific sustainable components employed for the device fabrication.

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3.1 | Challenges with Conventional Materials for Printed Electronics

Digital printing stands out for its high-resolution, design and material versatility, and cost-effective fabrication of (opto)electronic devices. Traditionally utilized inks comprise metallic and inorganic nanoparticles, functional precursors and synthetic organic compounds, along with organic solvents and additives. When adequately formulated, these materials provide excellent printability and enable multilayer architectures and high-performance devices, however sustainability aspects are not always considered.

From the early stages of printed electronics conductive materials have been widely explored toward flexible, lighter, and cheaper devices. Silver inks are the most used conductive materials in printed electronics because of their high electrical conductivity, as well as the relatively conductive nature of its oxides, which helps maintain electrical performance even after partial oxidation. However, it is considered a precious element, meaning a costly and nonrenewable material, although it can be recycled [110]. Interestingly, the electrical and electronic industry accounted for the 40.5% of Ag global demand in 2024, where the annual global recovery and supply do not meet the current annual demand and only the 18.7% originates from recycling [111]. Currently, some inks are reported as sustainable Ag inks because of the low loadings (~7 wt.% of Ag) [112], avoiding of organic solvents and surfactants, and reduced sintering temperatures [113]. Au inks are chemically more stable but extremely expensive, hindering the scalability. Therefore, Au is ~49 times less used than Ag, where only the 8% of the demand is dedicated to electronics [114]. Other metals such as copper, aluminum, zinc, magnesium, or nickel are more abundant and affordable alternatives; however, their high sensitivity to form insulating oxides during and after printing and ease of cracking of the layers on flexible substrates limits long-term performance, often requiring processing in inert atmosphere and the use of protective layers, both increasing the fabrication steps and complexity [115]. An alternative that introduce circularity to metals is the biogenic synthesis of Ag, Au, and Cu nanoparticles [116, 117] (and other metals). However, there is no mature industrial production of biogenic metal. Transparent conducting oxides achieve the unusual combination of optical transparency and metallic-like conductivity because they are wide-bandgap semiconductors (>3 eV) that are degenerately doped, resulting in the Fermi level being shifted into the conduction band [6]. Therefore, in addition to metallic conductive layers, several metal oxides, such as antimony tin oxide (ATO), fluorine tin oxide (FTO), indium tin oxide (ITO), aluminum zinc oxide (AZO), and gallium zinc oxide (GZO) have been employed as transparent conductive materials due to their excellent electrical conductivity and optical transparency. Nevertheless, their adoption in flexible and printed electronics remains limited because of their relatively poor mechanical flexibility and the challenges associated with their processing and deposition under printing-compatible conditions [75].

Beyond conductive materials, semiconductors are also essential in the performance of printed electronics such as transistors, photodetectors, sensors or light emitting diodes (LEDs) among others. Inorganic semiconductive materials, such as silicon, tin oxide (SnO_2), indium oxide (In_2O_3), zinc oxide (ZnO), gallium oxide (Ga_2O_3), indium gallium zinc oxide (IGZO), and molybdenum disulfide (MoS_2) have been processed via printing either in form of nanoparticles or organometallic precursors. These oxides have produced high-performance electronics however they have a nonrenewable origin and typically are not biodegradable materials [75]. Beside inorganic semiconductors, organic semiconductors such as poly(3-hexylthiophene) (P3HT) have also constituted a major class of active materials, enabling the development of organic field-effect transistors (OFETs), organic LEDs (OLEDs), organic photovoltaics (OPVs), and a wide variety of sensing applications manufactured via printing technology. In inorganic semiconductors, conductivity arises from a bandgap separating filled and empty electronic states, while in conjugated organic semiconductors such as P3HT, charge transport instead relies on the overlap of π-orbitals between adjacent polymer chains [118]. Although several of these technologies have succeeded as commercial products, their conjugated backbones entail high chemical and thermal stability that led to uncertain end-of-life scenarios because each semiconductor has specific recycling requirements [119, 120].

Likewise, other nonbiodegradable polymers have been extensively used as insulators or binders when combined with functional additives, such as polymethyl methacrylate (PMMA) or poly(4-vinylphenol) (PVP). Finally, although it is out of the scope of this work, most of the flexible substrates employed in digitally printed electronics are nondegradable polymers such as poly(ethylene terephthalate) (PET), polyimide (PI), and polydimethylsiloxane (PDMS), which, after the printing of the functional layers, become challenging to separate, recover, reuse, or dispose [121].

In short, most of the aforementioned material systems, despite producing high-performance printed devices, are also associated with critical drawbacks such as scarcity, toxicity, high energy consumption during processing, and uncertain end-of-life outcomes (e.g., poor biodegradability and limited recyclability). To address these challenges, research has increasingly focused on sustainable alternatives, including carbon-based particles, biosourced polymers, and recyclable active materials in recoverable systems. Besides, ionic liquids are being introduced in printed electronics as functional counterpart. In parallel, greener solvents and water-based ink formulations have emerged, where low-temperature sintering supports a low environmental footprint of printed devices. These approaches enable opportunities for more sustainable electronic devices with benign end-of-life pathways, facilitating easier recyclability, reusability or biodegradability of the produced devices while presenting exciting materials science and engineering challenges.

3.2 | Emerging Functional Materials for Sustainable Digitally Printed Electronics

Carbon-rich materials, such as graphite, graphene, CNTs, carbon black, and activated carbon, are widely used in functional inks for digital printing due to their excellent electrical, optical, thermal, and mechanical properties and superior chemical stability compared to metals. Therefore, these materials have emerged

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as a good alternative to metal NPs in printed electronics. These fillers have been extensively investigated in printed electronics, sensors, and energy devices on flexible substrates through the various digital printing techniques that explained in previous section [122].

Graphite consists of micro-sized planar particles of stacked carbon atoms with sp^2 hybridization arranged in a honeycomb lattice, exhibiting an electrical conductivity in the range of 10–10^3 $S{\cdot}m^{-1}$ when forming printed layers [123, 124]. It has been extensively used as electrode material due to its high availability, purity, low cost, and stability [125]. However, the relatively high size in the micrometer scale can limit the printing of graphite by techniques that employ small nozzles like inkjet printing, EHD or AJP. Conversely, few layers of graphene (defined as a single layer of graphite) can be obtained in nanometre sized when the graphite is ball-mill, shear, or liquid phase exfoliated [123, 126]. Graphene presents diverse morphologies, and its high surface area allow the tailoring of ink formulations and higher electrical conductivity when printed ($\sim 10^3$–10^4 $S.m^1$) [127, 128]. However, graphene is not well dispersed in many solvents; therefore, some polymers can be used as dispersants. Figure 3a shows aerosol jet spray printed micro-supercapacitors on paper printed with a graphene-based ink. Graphene, was exfoliated by liquid phase from graphite mixed with polypropylene carbonate (PPC), a synthetic and biodegradable polymer, as dispersant aids. The prints were then annealed at 220°C for 30 min and the PPC is degraded at such temperature leading to a graphene layer with an electrical conductivity of 2.05×10^4 $S{\cdot}m^{-1}$ [129]. Although not implemented at great scale yet, graphene can be obtained from various natural sources such as glucose, rice husk, hemp, disposable paper cups, coal, cockroach legs, cookies, and grass, which provide a renewable origin to the material [130].

Another strategy to enhance dispersion is the formulation of inks using graphene oxide (GO), consisting in the same lattice highly decorated with oxygen-rich moieties (epoxide, hydroxyl, carbonyl, and carboxyl). GO increases hydrophilicity and dispersibility, which facilitates the printing process, however resulting in lower conductivity or even insulating properties. Typically, it is used as a precursor for conductive inks because GO can be partially deoxygenated via post-printing reduction to form reduced graphene oxide (rGO), which shows higher conductivity (10^3 $S{\cdot}m^{-1}$ for inkjet printed layers) [131] as it increases sp^2 hybridized carbon to oxygen ratio, but does not reach the pure graphene range [132]. In 2010, an ink of rGO for inkjet printing was published for the first time. That ink was an aqueous dispersion of rGO at 3 mg mL^{-1}, where isopropanol (IPA), polyethylene glycol 200 and Triton-X100 in small amounts were added for the proper printing and dispersion. The material was deposited onto a poly (ethylene terephthalate) (PET) substrate and employed to reversibly detect NO_2 and Cl_2 vapors in air samples at parts per billion level [133].

Besides graphene and GO, CNTs offer exciting properties attractive for printed electronics such as lightweight, high surface area, high chemical and thermal stability, and high electrical conductivity. They consist of cylindrical structures formed by rolled up graphene sheets, where a single layer of carbon atoms forms single walled CNTs (SWCNTs), while several concentric layers form multi walled CNTs (MWCNTs) [138, 139]. A single CNT can exhibit a conductivity between 10^3–10^7 $S{\cdot}m^{-1}$ while it decreases to 10^2–10^5 $S.m^{-1}$ for 2D deposited layers. The broad range in the electrical properties is ascribed to the thickness of the layers, their CNT–CNT contact, and the type of CNTs, where MWCNTs are semiconductive and SWCNTs have metallic behavior and higher conductivity [140, 141]. A random network of CNTs in thin films leads to semiconducting properties, consequently, they have been applied as active layer in printed film transistors [126, 127, 142]. Similarly, MWCNTs are semiconductive due to interwall electronic coupling among layers with different chiralities whereas SWCNTs can be truly metallic with higher conductivity because their electronic properties are determined by a single wall's chirality, allowing purer and more efficient electron transport and thereby higher conductivity. In addition, CNTs have been employed to develop transparent conductive films substituting ITO due to their high extrinsic optical transparency, better stretchable films formation, easier and versatile deposition and scalable production in bulk quantities [6, 75, 143]. Composites comprising low amount of CNT have been applied in pressure and strain sensors, and transparent electrodes [144]. The high aspect ratio and strong van der Waals interactions of CNTs cause aggregation, poor dispersibility, and high viscosity, risking printhead clogging. Wrapping with high-molecular-weight polymers mitigates this but increases material loading and can reduce the printed network's conductivity [145]. CNTs and graphene show excellent performance comparable to metallic materials, but their costs are high, and their dispersion still remains a challenge for digital printing technologies [146, 147].

Finally, activated carbon and carbon black consists of spherical nanoparticles of amorphous carbon with graphitic domains, were activated carbon present high porosity. Traditionally, carbon black was obtained from incomplete combustion of hydrocarbons, but currently can be obtained from renewable feedstocks such as bamboo and natural fibers or from industrial wastes [148, 149]. Meanwhile, activated carbon have been obtained from a plethora of biomass feedstock such as grains and starch crops, agricultural residues, food waste, or animal by-products [150]. They present lower conductivity compared with CNTs and graphene, (10–10^4 $S{\cdot}m^{-1}$), but this is offset by its low cost, large scale production, and good dispersibility in solvents. Consequently, activated carbon and carbon black have been used as a conductive filler in inks for printing electrodes in various applications [151, 152]. Damasceno et al. formulated a conductive inkjet printable ink using activated carbon particles, with a diameter between 35 and 70 nm from coconut shells, ethyl cellulose, ethanol, and terpineol for printed resistive sensors (Figure 3b). The ink demonstrated high colloidal stability, while the printed conductive patterns showed a conductivity of 15.15 $S{\cdot}m^{-1}$ [136]. Overall, ink formulations containing carbonaceous fillers are extensively used, enabling applications such as wearable or piezoresistive sensors, conductive electrodes and traces, and printed energy storage components [75].

Beyond processability, carbon-based materials, are considered degradable/biodegradable, particularly for oxygen rich structures. For example, hydrogen peroxide (H_2O_2) can be used to attack C—C bonds generating a hole in the lattice and the surrounding carbon atoms are more susceptible to this attack increasing the hole size until complete degradation [153]. In the

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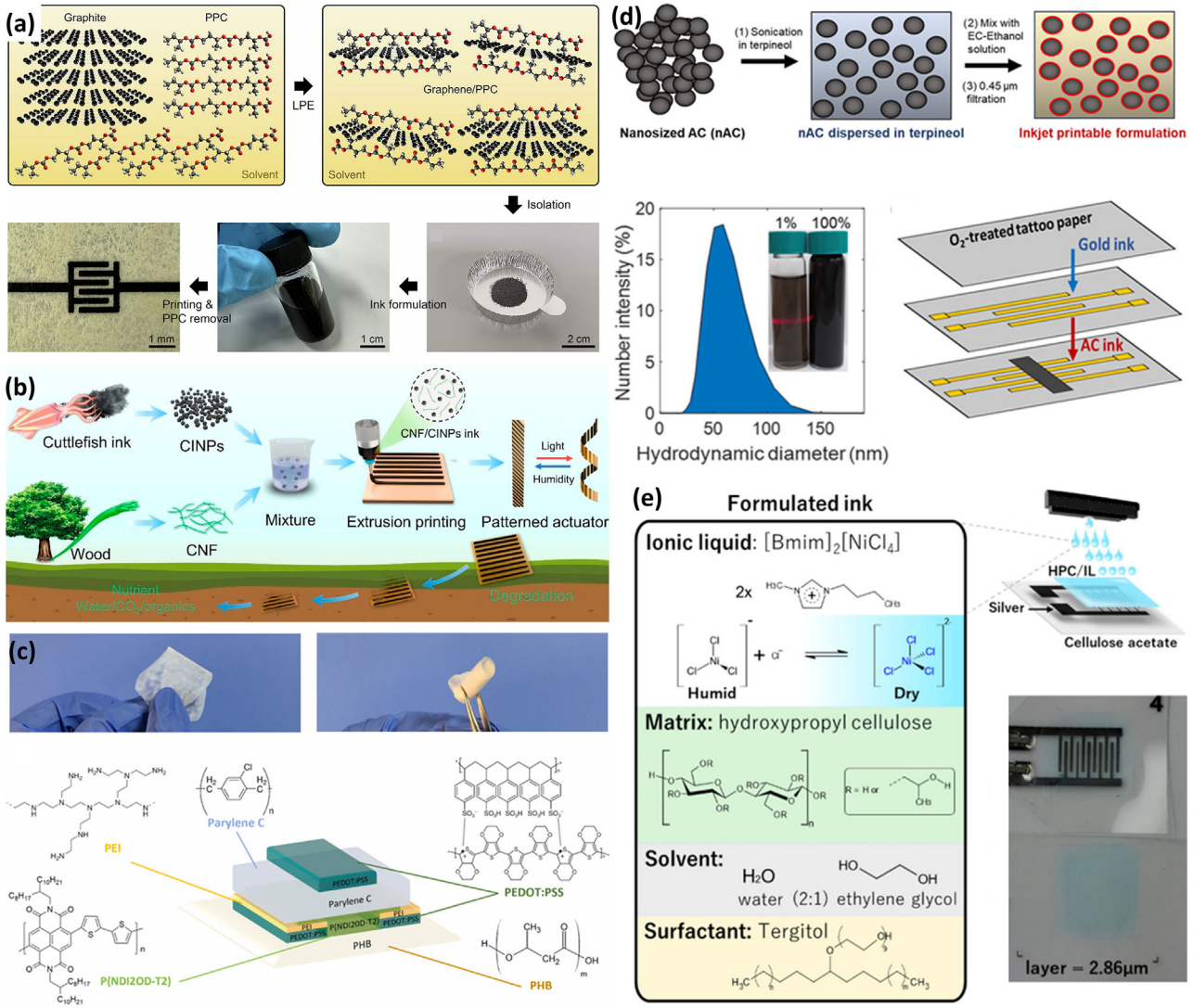


FIGURE 3 | (a) Scheme showing the liquid-phase exfoliated graphene from graphite using PPC as a dispersant aid, followed by the ink of graphene/PPC preparation and the subsequent printed micro-supercapacitor electrodes deposited by aerosol jet printing. Reproduced under the terms of the CC-BY license [129]. Copyright 2025, Springer Nature. (b) Illustration showing the preparation of CNF/CINP ink and the fabrication process of the soft actuators. Reproduced with permission [134]. Copyright 2024, American Chemical Society. (c) Images of PHB film used as substrate and representation of the OFET and the chemical formulas of the used polymers. Reproduced under the terms of the CC-BY-NC-ND license [135]. Copyright 2024, Elsevier. (d) Schematic representation of activated carbon and ethyl cellulose preparation. Average DLS data over for the ink diluted to 1% in a terpineol–ethanol mixture (1:1 volume). Printed activated carbon ink over gold electrodes perpendicularly oriented as proof-of-concept of resistive sensor. Reproduced under the terms of the CC-BY-NC-ND license [136]. Copyright 2025, American Chemical Society. (e) Schemes of the formulated ink based on $[Bmim]_2[NiCl4]$ and hydroxypropyl cellulose, together with the inkjet printing process and image of printed humidity sensor in blue color at a relative humidity below 30%. Reproduced with permission [137]. Copyright 2024, American Chemical Society.

Fenton reaction, CNTs and GO are exposed to H_2O_2 and ferrous salts to generate highly reactive hydroxyl radicals (·OH) that oxidize the structure and when combined with ultraviolet irradiation (i.e. photo-fenton reaction), the exposure accelerates the production of the radical groups [154]. Biodegradation methods for this family of materials mainly include enzymatic degradation, cellular degradation and bacterial degradation [155, 156]. Enzymatic degradation, the most studied approach, also uses H_2O_2. H_2O_2 is a naturally occurring ubiquitous compound in the human body that can be produced by several enzymes in biological systems, specially through peroxidases such as lignin peroxidase (LiP), horseradish peroxidase (HRP), neutrophil myeloperoxidase (MPO), eosinophil peroxidase (EPO), and lactoperoxidase (LPO) [157, 158].

In addition to carbon-based materials, some polymers present conductivity and have been used as conductive layer without the presence of conductive fillers or additives such as poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), polyaniline (PANI), and polypyrrole (PPy). These polymers are widely used in printed electronics for their electrical conductivity, printability, and environmental stability. However, they are not natural sourced or biodegradable. Among them, PEDOT:PSS is the most widely used p-type polymer due to its high electrical

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conductivity and has been employed in stretchable electronics because it resists high mechanical strain maintaining functional performance. A whole polymeric organic field-effect transistor (OFET) has been printed on polyhydroxybutyrate (PHB) substrate, a bioderived and biodegradable biopolymer, using PEDOT:PSS as for the source and drain contacts (Figure 3c), whose working principle is explained in next section [135]. Interestingly, some studies have reported that the incorporation of biodegradable materials to PEDOT:PSS lead to a degradation of the stack material, such as PEDOT:PSS in combination of Graphene/PVA [159], Silk Fibroin [160], or montmorillonite [161].

On the other hand, biopolymers (natural sourced, biodegradable, and biocompatible) are also attractive for added value materials development as they combine mechanical lightweight, low-cost, natural abundance, circularity, versatility to functionalize, and for enabling manufacture on flexible materials like plastics, textiles, and paper. In general, biopolymers, such as cellulose, chitosan, gelatin, shellac, and silk (protein-based biopolymer) are widely used in 2D printable inks for applications in sensors, thermal interfaces, dielectric layers, smart (stimuli responsive) materials, and printed electronics [162]. Cellulose is the most abundant biopolymer available and forms the main structural component of plant cell walls. It has excellent mechanical strength, but low density due to its network of strong inter- and intra-chain hydrogen bonds, which give cellulose its high tensile strength and insolubility. Cellulose nanocrystals (CNCs), which are needle-shaped nanoparticles, have also been used as printable dielectric layers [163]. The surface of cellulose contains high density hydroxyl (—OH) groups, which can polarize strongly under an applied electric field [164]. As a result, cellulose exhibit good dielectric properties and typically has a dielectric constant between 3 and 10 depending on its molecular structure. Environmental conditions can also affect the dielectric behavior of cellulose [162]. Some cellulose derivatives such as hydroxypropyl cellulose (HPC) and carboxymethyl cellulose (CMC) are soluble in water and present good film forming capacity to be employed as substrates or binders. While cellulose derivatives nonsoluble in water such as cellulose diacetate and ethyl cellulose are commonly used as transparent and resistant substrates, substituting PET or PMMA as is described in the applications section.

Environmental conditions can also affect the dielectric behavior of cellulose [162]. Since cellulose is hygroscopic and it can absorb and holds moisture, humidity changes can alter its dielectric properties. This makes cellulose useful for humidity sensors [165]. Moreover, because cellulose can expand or contract with humidity changes, it is also a candidate material for mechanical actuation applications [166]. In Figure 3d, cellulose nanofibers (CNFs) are shown, a higher aspect ratio cellulose structure with crystalline and amorphous domains, combined with cuttlefish ink (CINP), which consists of natural melanin nanoparticles to develop an DIW ink for light and humidity bending actuators [134]. Similarly, an ink to design anti-counterfeiting system was formulated based on poly(amidoamine) (PAMAM) dendrimer modified with microcrystalline cellulose decorated with coumarin [167].

Gelatin and alginate are water-soluble biopolymers that have been used for the formation of hydrogel layers, which can even be printed as self-standing patterns. Gelatin is a polypeptide derived from collagen's partial hydrolysis sourced from pig and cuttle skins, hooves and bones, and fish [168]. Very soft hydrogels based on gelatin are principally printed using DIW and EHD due to its water solubility, good adhesion, and reversible sol–gel transition, which are combined with biocompatibility, degradability, and mechanical flexibility [169]. In printed electronics liquid and solid fillers have been combined with gelatin to be used as electrolyte layers in electrochromic devices [170], electrodes when comprising CNTs [171], and ionic electrodes [172].

Conversely, alginate is an anionic polysaccharide located in cell wall of brown algae. Inks are typically based on sodium alginate due to its solubility in alkaline media, its nontoxicity, film-forming capacity, strong gelation, and cross-link with cations. Therefore, when sodium alginate is exposed to polyvalent cations, frequently Ca^{2+}, is formed calcium alginate gel with cross-linked bond because Ca^{2+} displaces part H^+ and Na^+. The cross-linking process can be produced by printing sodium alginate with the tip of DIW printer on a calcium solution. Contrary to gelatin, pure alginate gels are rigid and fragile [173]. However, there has been printed free-standing soft actuators [174]. Interestingly, it have been reported as edible biopolymer a printed hydrogel comprising alginate and gelatin as matrix, and CNC as a reinforcement filler to be used as sensor in food industry [175].

Similarly, shellac, a natural biopolymer resin secreted by the lac insect (Kerria lacca), have been inkjet printed, exhibiting good insulating properties and relatively low dielectric loss over broad frequency ranges [176]. It forms robust, smooth films with high dielectric breakdown strength, used as a bio-derived dielectric component in low-voltage organic transistor structures and as a binder in printable electronic inks [177]. The molecular structure of shellac presents a hydrophobic long-chain of fatty acids with free carboxyl (—COOH) and hydroxyl groups that facilitate film formation and adhesion [178], which makes it insoluble in water but dissolves in alcohols and alkaline solutions. In addition, the density of polar functional groups in shellac contributes to dielectric behavior, enabling solution processing with low-toxicity, volatile-organic-solvent-based inks compatible with printed electronics [177].

Although publications in nanotechnology have grown for more than 30 years, concerns remain about risks and human health impacts from prolonged NP exposure. The behavior of NPs is governed by their size, shape, and surface reactivity with surrounding tissues [179]. The extremely small size of nanomaterials facilitate entry into the human body and cells. Once inside the body, the fate of NPs remains insufficiently understood; for example, nondegradable or slowly degradable nanoparticles may accumulate in organs or interact with biological processes in unforeseen ways. At present, knowledge of NP cytotoxicity remains incomplete and controversial [180, 181]. In this framework, ionic liquids (ILs) are increasingly used as functional components in ink formulations for 2D printing due to their ionic conductivity, thermal stability and tunable physicochemical properties. They are salts in liquid state at or near room temperature and exhibit low volatility, making them particularly suitable for digital printing techniques, where controlled viscosity and wetting behavior are essential. ILs can be used for multiple purposes and serve as conductive media,

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dispersants or even additives to tailor the printability, stability, and functional response of the inks [182]. Their intrinsic ionic conductivity makes them relevant in applications requiring ion transport, humidity responsiveness, or electrochemical activity, such as printed sensors, soft electronics, and energy storage devices [183]. They can also act as dopants for conductive polymers. Thus, neat PEDOT:PSS typically shows a conductivity of <1 mS cm^{-1}, whereas adding an IL can increase conductivity to 4100 S·cm^{-1} [184]. Moreover, the IL structure can be tuned to achieve specific rheological and intermolecular properties, enabling thus the formulation of inks compatible with a wide range of substrates and printing techniques [185].

However, several ILs contain imidazolium cation, which present toxicity. To tackle this problem, the structural modification of ILs with polar functional groups or using hydrophilic anions such as carboxylates or sulfonates can reduce the toxicity and improve the biodegradability of the IL. Another strategy is to substitute imidazolium with the bio-sourced choline cation (Choline = 2-hydroxyethyltrimethyl ammonium chloride). Choline-based ILs such as choline lactate ([Ch][Lactate]), choline glutarate ([Ch][Glu]) [186], or choline dihydrogen phosphate ([Ch][DHP]) [187] are generally regarded as nontoxic, biocompatible, biodegradable, and readily available.

Additionally, various active ILs are capable of producing an optical and/or electrical response and have been used in responsive and sensitive sensors, actuators, environmental remediation, batteries, fuel cells [183]. Specifically, ionic liquids based on platinum (II) cyclometalated complexes and polymeric IL based on poly(diallyldimethylammonium) present electroluminescent response [183]. Humidity sensors have been fully inkjet printed using choline chloride combined with melanin as active layer. It was demonstrated that the materials of this active layer could be recovered by dissolution in water and reprinted on the same electrode [188]. Another humidity sensor has been inkjet printed with an ink based on the IL bis(1-butyl-3-methylimidazolium) tetrachloronickelate ($[Bmim]_2[NiCl_4]$) mixed with hydroxypropyl cellulose, water and ethylene glycol in ratio 2:1 as solvent and tergitol as surfactant. As shown in Figure 3e $[Bmim]_2[NiCl_4]$ changed from transparent to blue color below a 30% of relative humidity [137]. In terms of processability, the liquid nature of ILs results excellent for printing techniques. On the one hand, they do not form aggregates or sediments, and their high boiling point slows the drying process, both of which help to reduce clogging during the printing process and therefore can extend the life of assemblies [188]. On the other hand, the combination of IL and polymers avoids annealing or energy-consuming postprocessing after printing.

The mentioned materials are used as the active part in form of functional fillers or additives in the ink. Another key component of most of solvent-based inks is the binder or matrix that holds the particles inside as in a single material and provides adhesion to the substrate. Polymeric matrices in nanocomposites can comprise the highest weight ratio of solid component of an ink. Therefore, the selection of sustainable materials has a great impact in the environmental footprint and the handling complexity in the end-of-life. Particularly, cellulose and its derivatives have been employed for the formulation of inks for digital printing techniques blended with carbon fibers [189], graphite [190], and nanocarbon from food waste [191] for developing conductive inks and thermoelectric touch sensors [192], or with the ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide [EMIM][TFSI] for developing ionogels with high capacitance (5 to 15 μF cm^{-2}) [193] to be applied in capacitors and gates for field effect transistors [194]. CNCs are not only used as active part, but also as binder to develop films and membranes. García-Castrillo et al., mixed them with spherical Au NPs to formulate an aqueous ink for photothermally active material [195].

3.3 | Ink Formulation Strategies

Having reviewed the individual conductive, semiconductive, and biopolymer building blocks above, the remaining question is how they are combined into a single printable system. Ink formulations generally consist of functional materials (conducting, semiconducting, or insulating components), binders, solvents, surfactants, and various additives that together determine printability, film quality, and device performance. While the use of renewable, biodegradable, and low-toxicity functional materials and binders, as discussed in the previous section, represents a crucial step toward eco-friendly electronics, their successful implementation depends on their integration into printable ink systems that preserves the sustainability in all the components. Therefore, "*green*" solvents, surfactants and additives should be selected while maintaining optimal printability, stability, and performance.

Formulation design must also be adapted to the specific requirements of each printing technique, since the same material system may be printable in one process but unsuitable for another. As mentioned earlier, for inkjet printing, formulations must be tuned to a narrow viscosity window (typically 2–15 mPa·s) and a controlled surface tension, since these two parameters jointly govern droplet ejection and stability through the dimensionless *Z*-number. Surfactants, dispersants, and cosolvents are added not simply to help printability, but to hit this specific window. For example, surfactants lower surface tension to permit reliable Rayleigh break-up of the jet into a single droplet, while cosolvents suppress nozzle-tip drying and satellite-droplet formation. Sustainable examples of this balance include activated-carbon inks stabilized with ethyl cellulose [136] and ionic-liquid/hydroxypropyl cellulose formulations for inkjet-printed humidity sensors [137], both of which pair a biodegradable binder with a printable rheology. In EHD printing, the formulation must additionally support stable Taylor cone formation and jetting under an electric field, making viscosity, viscoelasticity, surface tension, and electrical conductivity particularly important. Bio-based ionic conductors such as self-healing gelatin/tannic acid elastomers have been successfully patterned via EHD printing, illustrating how EHD's requirement for controlled viscoelasticity can be met with biodegradable materials [172]. In AJP, atomization and aerosol transport require inks with suitable viscosity, shear-thinning behavior, surface tension, and dispersion stability to avoid overspray, nozzle instability, or poor feature definition. A graphene ink dispersed with polypropylene carbonate (PPC), a biodegradable polymer dispersant, has been aerosol-jet printed into micro-supercapacitor electrodes, with the PPC thermally decomposing post-print to

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leave a high-conductivity graphene layer [129]. By contrast, DIW requires highly loaded inks or pastes with yield stress, shear-thinning behavior, and rapid structural recovery, allowing the ink to flow through the nozzle while preserving filament shape after deposition. Achieving these properties sustainably is challenging, rheology modifiers such as cellulose nanocrystals, lignin, xanthan gum, or biopolymer thickeners are increasingly used in recent studies for tuning into water-based DIW pastes without depending to petrochemical counterparts, and recent work on water-based conductive pastes for extrusion-printed electrodes and biodegradable supercapacitors demonstrates this feasibility [196–198]. Systematic frameworks for quantifying DIW printability, linking yield stress, storage/loss modulus ratios, and nozzle geometry to print fidelity, have also matured recently, providing formulators a more rigorous basis for reformulation than trial-and-error viscosity adjustment [199].

These technique-dependent requirements highlight that sustainable reformulation cannot simply rely on replacing a hazardous solvent or binder with a greener alternative, because each substitution can modify viscosity, surface tension, evaporation rate, particle dispersion, wetting, film morphology, conductivity, and mechanical integrity. One of the main challenges in eco-friendly ink formulations is to replace petrochemical-derived and toxic traditional solvents such as NMP, DMF and toluene with sustainable, nontoxic, and biodegradable alternatives. In this context, the best alternative is the use of water as solvent or mixed aqueous–organic systems such as water with ethyl lactate or other biomass-derived cosolvents used to minimize solvent derived VOCs emissions while maintaining particle stabilization and retard drying during printing, especially in inkjet and EHD systems [58, 69]. Cyrene (dihydrolevoglucosenone) has emerged as promising substitute to conventional solvents as it enables liquid-phase exfoliation of 2D nanomaterials (graphene or MoS_2) and produces stable inks without the toxicity associated with traditional high-boiling solvents [200]. Comparative studies highlight the good dispersion quality and device-level functionality using Cyrene-based graphene inks [201]. γ-valerolactone (GVL) is a nontoxic compound derived from lignocellulose that is substituting DMF and DMSO in inkjet printed photovoltaic devices [202]. Considering the complexity of solvent selection, Larsen et al. developed a green solvent selection tool for printed electronics, based on Hansen solubility parameters, ink-relevant properties, and sustainability descriptors, to identify functional and "green" replacement solvents [56].

In parallel, surfactants and dispersants are being reformulated toward biodegradable and low-toxicity species. Furthermore, they should present tunable interfacial properties and compatibility with polymer-nanoparticle systems which is useful for reducing surface tension, controlling droplet formation, and stabilizing colloids for inkjet deposition [58]. Biosurfactants such as rhamnolipids, sophorolipids, or saponins/glycolipids gather these requirements [203]. In addition, readily biodegradable non-ionic surfactants with petroleum-free chemistries (e.g., secondary alcohol ethoxylates) are being used. For instance, the addition of TERGITOL 15-S-9 reduced the surface tension of water-based PEDOT:PSS from ~70 to ~30 mN m^{-1} and enabled homogeneous inkjet-printed transparent electrodes (demonstrated in OLED stacks), while avoiding fluorinated/ionic additives whose end-of-life profiles are problematic [204].

Overall, eco-friendly ink formulations are advancing by integrating green solvents, biodegradable/bio-based surfactants, safer binders, and fillers/additives. However, their development requires careful optimization because the components that improve printability may also affect film formation, conductivity, adhesion, mechanical stability, recyclability, biodegradability, and end-of-life safety. Beyond their environmental benefits, the eco-friendly formulation of inks also ensures compatibility with a broad range of substrates and digital printing methods. When combined with optimized printing processes, these eco-friendly inks enable the fabrication of fully functional and high-performance electronic devices. In the following section, we highlight how these material strategies are being implemented in practical applications such as transistors, displays, sensors, actuators, and energy devices.

4 | Applications

The materials and digital printing techniques discussed in the previous sections have effectively contributed to the development of a new variety of more sustainable electronic devices. This section explores some of these applications, such as transistors, displays, sensors, actuators, energy harvesters, storage, transmission, and other electronic devices, where sustainability concepts have been utilized for their digital fabrication.

4.1 | Electronic Devices

4.1.1 | Transistors

Transistors are the building blocks of electronic circuits and have been used for applications ranging from simple switching to complex amplification and integrated circuits. The advancement in printing technologies has opened pathways for printing transistors with reduced material waste, lower processing temperatures, and compatibility with flexible form factors [205]. The development of eco-friendly transistors is a crucial first step toward achieving fully sustainable electronics. Digitally printed transistors have been demonstrated in multiple architectures and functional modalities, such as field-effect (FET), electrolyte-gated (EGT), and synaptic implementations, employing eco-friendly materials for substrates, dielectrics, electrodes, and active layers.

A FET, a three-terminal device, operates by using an electrically isolated gate electrode that modulates the conductivity of a semiconducting channel. The source and drain serve as the two ohmic contacts to the channel. When a voltage is applied to the gate, it creates an electric field that alters the distribution of charge carriers within the channel, thereby changing its conductivity, termed the "field effect". An inkjet-printed FET was demonstrated on a biodegradable Ormocere substrate, using β-carotene (p-type) and indigo (n-type) as the semi-conductive channels, and shellac as the dielectric [206]. The transistor exhibited mobilities ranging from 6×10^{-4} to 7×10^{-2} cm^2 V^{-1} s^{-1}, which should be improved to target real applications and reach performance comparable to conventional organic semiconducting materials [207, 208]. This demonstrates the feasibility of bio-sourced materials for enabling complementary metal–oxide–semiconductor (CMOS) circuits in the future. However, PEDOT:PSS used for electrodes has

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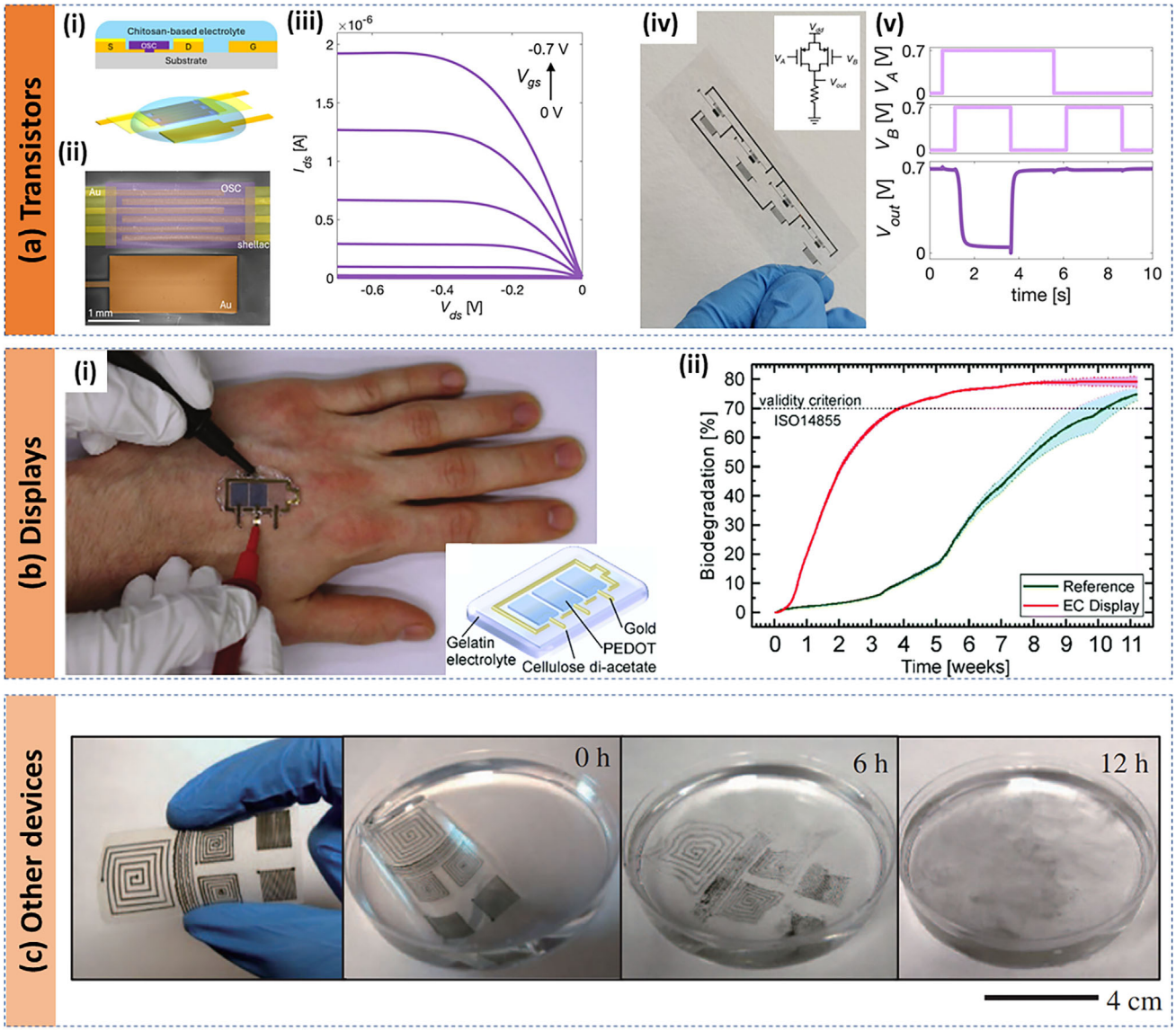


FIGURE 4 | Eco-friendly Devices. (a) Transistors: (i) schematic showing an eco-friendly EGT and its (ii) SEM photograph. (iii) The output IV characteristics showing the linear and saturation regimes. (iv) A logic NAND gate realized using the EGT (inset showing the circuit diagram) and (v) its *V*–*t* output characteristics for different combination of inputs V_A and V_B. Reproduced under the terms of the CC-BY license [210]. Copyright 2025, John Wiley and Sons. (b) Displays: (i) electrochromic display attached to skin with inset showing the device structure. (ii) Degradation of the device with time confirming compatibility with the ISO14855 standard. Reproduced under the terms of the CC-BY license [170]. Copyright 2020, Royal Society of Chemistry. (c) Passive devices: dissolution of printed Zn passive devices in water. Reproduced with permission [211]. Copyright 2018, Springer Nature.

exhibited degradation into microplastics [74, 209] and may need to be replaced with more eco-friendly alternatives or combined with biodegradable materials (as mentioned in Section 3) that improve its degradability. Semiconducting CNTs have also been used as the channel material in aerosol-jet printed FETs, paired with graphene source/drain electrodes and a nanocellulose dielectric on a paper substrate; the sorted, semiconducting-CNT network provides sufficient carrier mobility for transistor operation while the paper substrate allows straightforward material recovery and reuse at end-of-life [163].

An EGT is similar to a FET, except that the dielectric layer is replaced by an ionically conducting and electrically insulating electrolyte, which is in contact with both the gate and the channel. Figure 4a(i,ii) shows an EGT utilizing a chitosan-based electrolyte [210]. The device is fabricated by inkjet printing of Au electrodes on ethyl cellulose substrates, an organic polymer as the channel material, and shellac as an electrode passivation layer, and. Most of the inks were formulated using low-toxicity solvents such as ethanol and terpineol; however, water-based formulations would make it more eco-friendly, as well as the substitution of Au with a carbon-based conducting electrode. The typical *I*–*V* characteristics of the printed EGT are shown in Figure 4a(iii), demonstrating both linear and saturation regimes, at gate voltages below 0.7 V, comparable to conventional counterparts. The EGT can also be used to realize logic gates, including the universal logic gate such as NAND, as demonstrated in Figure 4a(iv,v). Compared with FETs, EGTs have larger propagation delays and is more

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challenging when limited to eco-friendly electrolytes, especially to realize electronic circuits [212]. However, EGTs are widely used in bioelectronics because they operate at low voltages, provide high transconductance, and interface effectively with ionic environments [213, 214].

An emerging category of transistors are synaptic transistors, which working principle is analogous to human neurons by concurrently storing and processing information. This is achieved by adjusting the internal electrical properties similar to how the synaptic weights are updated in our neural system. Inkjet-printed synaptic transistors have been reported. A cellulose nanofibril/PVA hybrid electrolyte-based synaptic transistor demonstrated mimicking synaptic plasticity by modulating Na+ ion conductivity [215]. Under weak electrical stimulation, the Na^+ ions temporarily shift but will recover due to electrostatic attraction when the gate potential is removed, enabling short-term plasticity. Under stronger stimulation, Na^+ ions penetrate and become stabilized within the PVA layer, leading to long-term plasticity. This ion-based, reversible and stable switching mechanism not only reduces the need for rare or toxic materials, but also supports low-power, biodegradable, and sustainable neuromorphic computing [215]. Similarly, an inkjet-printed reconfigurable memristor array (16 × 65 devices, >90% yield) using MoS_2 as the resistive switching layer on paper has been demonstrated, with device disassembly, material recovery, and re-fabrication shown explicitly, that offering a recyclable, end-of-life pathway for neuromorphic hardware, rather than intrinsic material renewability, since MoS_2 itself is a mined, nonrenewable resource [216].

Table 2 summarizes the electrical performance of various transistors demonstrating selected aspects of sustainability. Notably, most of these devices have been fabricated using inkjet printing. The compatibility of inkjet printing with low-viscosity inks allows the deposition of thin films with precise thickness and dimensional control. This is crucial for fine-tuning transistor performance. Compared with printed Si-based transistors [219–222], most of them have demonstrated comparable I_{ON}/I_{OFF} ratios; however, their charge carrier mobility is more than three orders of magnitude lower. This significant limitation restricts their use in high-frequency applications, underscoring the need to enhance mobility. Closing this gap will require developing new solution-processable eco-friendly semiconductors and advances in printing technology that enable smaller channel dimensions. Both are critical for achieving high-performance sustainable transistors, though, as discussed further in Section 6.3, the approaches summarized in Table 2 address only selected dimensions of sustainability rather than fully sustainable electronic systems.

4.1.2 | Displays

The eco-friendly material strategies developed for transistors carry over readily to other active devices—displays being a natural next example. Displays are an essential component in every modern electronic appliance, serving as an interface for human-machine interaction [223]. However, conventional manufacturing processes are highly resource-intensive and generate significant environmental burdens through the emission of non-eco-friendly chemicals and byproducts [6, 224]. Furthermore, advances in fabrication technologies have reduced production costs, inadvertently accelerating the rate of display deployment and, consequently, the volume of electronic waste. End-of-life management of displays remains a persistent global challenge, particularly in the context of hazardous material recovery and recycling [225, 226]. The development and adoption of eco-friendly printed display technologies offer a promising pathway to mitigate these issues by enabling sustainable manufacturing, reducing resource consumption, and facilitating environmentally benign disposal.

Electrochromism refers to a reversible, voltage-driven change in a material's optical absorption, typically via a redox reaction that alters the conjugated polymer's electronic structure and hence which wavelengths of visible light it absorbs [227]. An eco-friendly resource efficient, printed electrochromic (EC) display comprising PEDOT:PSS as the EC layer, a gelatin-based electrolyte, and Au electrodes on a cellulose diacetate substrate illustrates this approach [170]. Owing to its biocompatibility, such a display can be directly applied to human skin for biomedical applications (Figure 4b(i)). Furthermore, compliance with the International Standard ISO 14855 (Figure 4b(ii)) for biodegradability, over 79% degradation after 9 weeks, underscores its potential as a sustainable solution. The device demonstrated an EC contrast of 32%. Another inkjet-printed eco-friendly display was demonstrated, that can exhibit electrochemiluminescence (ECL) in addition to the EC based on alternating (AC) or direct current (DC) input mode of operation. A 45% EC contrast was achieved, primarily due to the PEDOT:PSS layer. In the dual-mode display, an eco-friendly ECL gel that composed of biodegradable poly(lactic-co-glycolic acid) dissolved in Dimethylsulfoxide (DMSO), an eco-friendly salt BMIMBOB as the ionic species, and the nonfluorinated emitter $Ru^{2+}(bpy)_3Cl_2$, exhibited a luminance of 52 mcd m^{-2} at an AC input of 4 V and 40 Hz [228]. However, the relatively long switching time of such displays warrants further optimization and currently limits its application to static displays that do not require fast refresh rates.

Quantum dots (QDs) are semiconductor nanocrystals whose emission color is set by particle size through quantum confinement, allowing narrow, highly saturated emission colors compared to conventional phosphors [229]. Modern display technologies, such as quantum dot light-emitting diodes (QLEDs), offer high-speed switching and exceptional color purity; however, many utilize Cd-based QDs, which are intrinsically toxic [230]. In recent developments, QDs, such as InP (red and green), AgCuInGaS (red), AgInGaS (green), and ZnTeSe (blue), have been employed to inkjet-print RGB QLED arrays [231, 232]. This work demonstrates the suitability of inkjet printing for large-area display fabrication. While the reported materials are less-toxic compared to their Cd-based counterparts, In and Te are rare elements, and appropriate end-of-life recycling protocols are necessary to improve sustainability. Alternatively, these elements could be replaced with earth-abundant materials such as Si [233]. Furthermore, as with many nanomaterials, the small size, high surface reactivity, and potential persistence of QDs raise considerations regarding their long-term environmental and biological interactions, which remain not yet fully understood. Particle-free electroluminescent ionic liquids should be explored as safer alternatives [234]. Nonetheless, the use

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TABLE 2 | Performance comparison of various transistors demonstrating selected aspects of sustainability.

		Materials									
Refs.	**Device**	**Active**	**Electrode**	**Dielectric/ Electrolyte**	**Substrate**	**Solvent**	**Sustainability aspect of device**	**Printing technique**	**μ ($cm^2\ V^{-1}\ s^{-1}$)**	**I_{ON}/I_{OFF}**	**VT (V)**
[135]	FET	P(NDI2OD-T2)*	PEDOT:PSS*/ polyethylenimine (ZnO)*	Parylene C	Polyhydroxybutyrate	Water, EG, IPA, Mesitylene	Biochemical oxygen demand (BOD) biodegradation analysis	Inkjet	0.07	10^3	4
[217]	FET	CNT*	Au*	water/Chitosan	Ethyl cellulose	Water, glycerol	Edible because each material quantity is less than acceptable daily intake values	Inkjet	$\mu s_{at}.C = 0.16\ \mu F\ V^{-1}.s^{-1}$	—	0.4
[218]	FET	P(NDI2OD-T2)*	PEDOT:PSS*/ polyethylenimine (ZnO)*	Parylene C	Mater-Bi	EG, IPA, water	BOD biodegradation analysis	Inkjet	0.01	10^5	2.7
[163]	FET	CNT*	Graphene*	Nanocellulose*	Paper	Water, toluene, phosphate-buffered saline (PBS)	Material recovery and reuse	AJP	—	$10^3 - 10^4$	—
[210]	EGT	P3HT*	Au*	Shellac*/ Chitosan	Ethyl cellulose	Glycerol, acetic acid, ethanol, terpineol, chlorobenzene, 1,2 dichlorobenzene	Edible because each material quantity is less than acceptable daily intake values	Inkjet	$\mu_{sat}C = 0.094\ \mu F\ V^{-1}\ s^{-1}$	10^3	−0.25
[206]	EGT	Carotine* or Indigo*	PEDOT:PSS*	Shellac*	Ormocere	Water, EG, ethanol, PGMEA, anisol, dimethyl sulfoxide	Accelerated degradation demonstrated in NaCl solution	Inkjet	$6\times10^{-4} - 7\times10^{-2}$	—	—
[215]	Synaptic	IGZO	Al, ITO	PVA (Cellulose nanofibril)*	Glass	EG, water	NA	Inkjet	10	5×10^4	—
[216]	Memristor	MoS_2*, Graphene*	Au, Ag*	—	Paper	IPA, 2-butanol	Material recovery and reuse	Inkjet	—	10^5	—

*Material layers that are printed.

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of solvents such as cyclohexylbenzene and octane in the ink formulations presents environmental concerns. Their replacement is not straightforward, since particle dispersion stability, ligand compatibility, drying kinetics, film morphology, and device performance are strongly governed by solvent-particle interactions. Therefore, greener solvent selection should be guided by solubility-parameter approaches, such as Hansen solubility parameters, together with relevant properties for inkjet printing including viscosity, surface tension, and volatility. In this context, lower-toxicity solvent systems or bio-derived nonpolar solvents may offer realistic alternatives [235].

4.1.3 | Passive Devices

Similar to transistor and display technologies, passive elements such as resistors, capacitors, inductors, and printed circuit boards (PCBs), play a crucial role in supporting the functionality of active devices. Recent work has demonstrated aerosol jet printing of polyvinylpyrrolidone (PVP)-stabilized Zn NPs on bioresorbable sodium carboxymethyl cellulose substrates to fabricate eco-friendly PCBs, resistors, and inductors [211]. Zinc is earth-abundant, low-cost, and biocompatible, offering a sustainable alternative to conventional conductive materials. Moreover, Zn-based printed electronics have been shown to dissolve completely in water within 12 h, marking a significant step toward transient and recyclable electronic systems (Figure 4c). Another potential route involves carbon-based materials; for example, a graphite/carbon-black composite ink using shellac as the binder has been extrusion-printed to fabricate resistors and capacitors [236]. Additionally, inkjet-printed bioresorbable capacitors have been developed using a PEDOT:PSS/shellac/PEDOT:PSS parallel plate structure on Ormocere substrates [206]. Printable bio-based dielectric such as shellac [206], cellulose [237, 238], and polylactic acid (PLA) [239] must be more popularized [240]. However, replacing PEDOT:PSS with metals such as Zn or Mg, or with carbon conductors could be considered based on the formulation and application requirements [241]. These alternatives may enhance conductivity while improving environmental compatibility, potentially accelerating the adoption of fully eco-friendly capacitors. Nonetheless, they also introduce challenges related to oxidation, dispersion stability, sintering or curing conditions, mechanical integrity, and long-term device reliability. Therefore, the development of fully eco-friendly capacitors requires the optimization of conductor, dielectric, substrate, and processing conditions simultaneously rather than the isolated substitution of a single material.

4.2 | Sensors and Actuators

4.2.1 | Sensors

Printed electronics are not limited to circuit components, however; they are also widely used to interface directly with the physical environment, which is the focus of this section. Sensors generally convert physical or chemical variables into electrical signals and are widely employed for many applications including medical diagnostics, manufacturing, home automation, automobiles, and robotics. One such application is in environmental monitoring, to collect data on parameters such as temperature, humidity, water and soil quality, light, and motion. Such monitoring is highly beneficial in agriculture, enabling long-term tracking of environmental trends to better understand and support crop productivity. At the same time, it is equally important to ensure that these sensors do not become an environmental burden at the end of their life cycle, potentially contributing to pollution or disrupting microorganisms that are essential for healthy crop development. Fabrication of sensors using bio-sourced materials together with inkjet printing is a promising combination in this context. In most printed resistive or capacitive sensors, the target stimulus alters the sensing material's electrical resistance or dielectric permittivity, for example, moisture absorbed by a hygroscopic material changes its ionic conductivity, and this change is transduced into a measurable electrical signal [242]. Inkjet printed melanin, a biopolymer extracted from the black soldier fly, has been successfully employed to demonstrate a humidity sensor for monitor soil moisture (Figure 5a(i)) [137]. The resistance of the melanin/ChCl composite was observed to be varying with humidity conditions (Figure 5a(ii)). Such a sensor fabricated on a cellulose diacetate substrate stands out not only for its performance but also for its potential biodegradability and compatibility with circular economy principles. Its active layer can be dissolved in water, enabling reuse of the substrate and electrodes, as shown in Figure 5a(iii,iv). This reusability reduces reliance on precious metals, while the biodegradable CA substrate minimizes environmental impact, highlighting the sensor's potential for sustainable agricultural monitoring. Another notable example is a temperature sensor with a temperature coefficient of resistance (TCR) of 1.21% K^{-1}, fabricated by inkjet printing using a graphene oxide ink formulated with the natural surfactant alkaline lignin [243].

Another important application is in healthcare, where the sensor can be employed for continuous monitoring of vital signs, early disease diagnosis, and therapeutic interventions. However, most wearable and point-of-care sensors are designed for single use, and their large-scale deployment risks creating significant environmental burdens. Therefore, the development of disposable yet biocompatible sensors is critical. For example, a fully inkjet printed glucose biosensor has been demonstrated using graphene ink for electrodes, Ag as reference, and Au as working electrodes [244]. The device exhibited a sensitivity of 140 $\mu A.cm^{-2}$ mM^{-1}, a linear response between 0.1 to 10 mM, and a limit of detection (LoD) of 0.09 mM, highlighting the strong potential of fully printed biosensing platforms. Additionally, an organic electrochemical transistor (OECT) glucose sensor, with an inkjet printed PEDOT:PSS channel and screen printed carbon electrodes, showed a linear sensitivity of −3.4%/dec across the range of 0.001 to 1 mM [214]. This device illustrates that transistors can serve as sensing elements as well as circuit components. As noted above for PEDOT:PSS's stability limitations, it's in vivo degradation pathways specifically remain insufficiently understood. Beyond glucose sensing, self-healable gelatin/tannic acid composites, capable of modulating ionic conductivity under strain, have been employed for monitoring joint movements (Figure 5a(v,vi)) [172]. The sensors exhibited a resistance variation of 35% under 100% strain and with a self-healing recovery efficiency of above 96%. Such bio-based and ionic conductors, which can be patterned via EHD printing, represent a viable route toward future healthcare applications.

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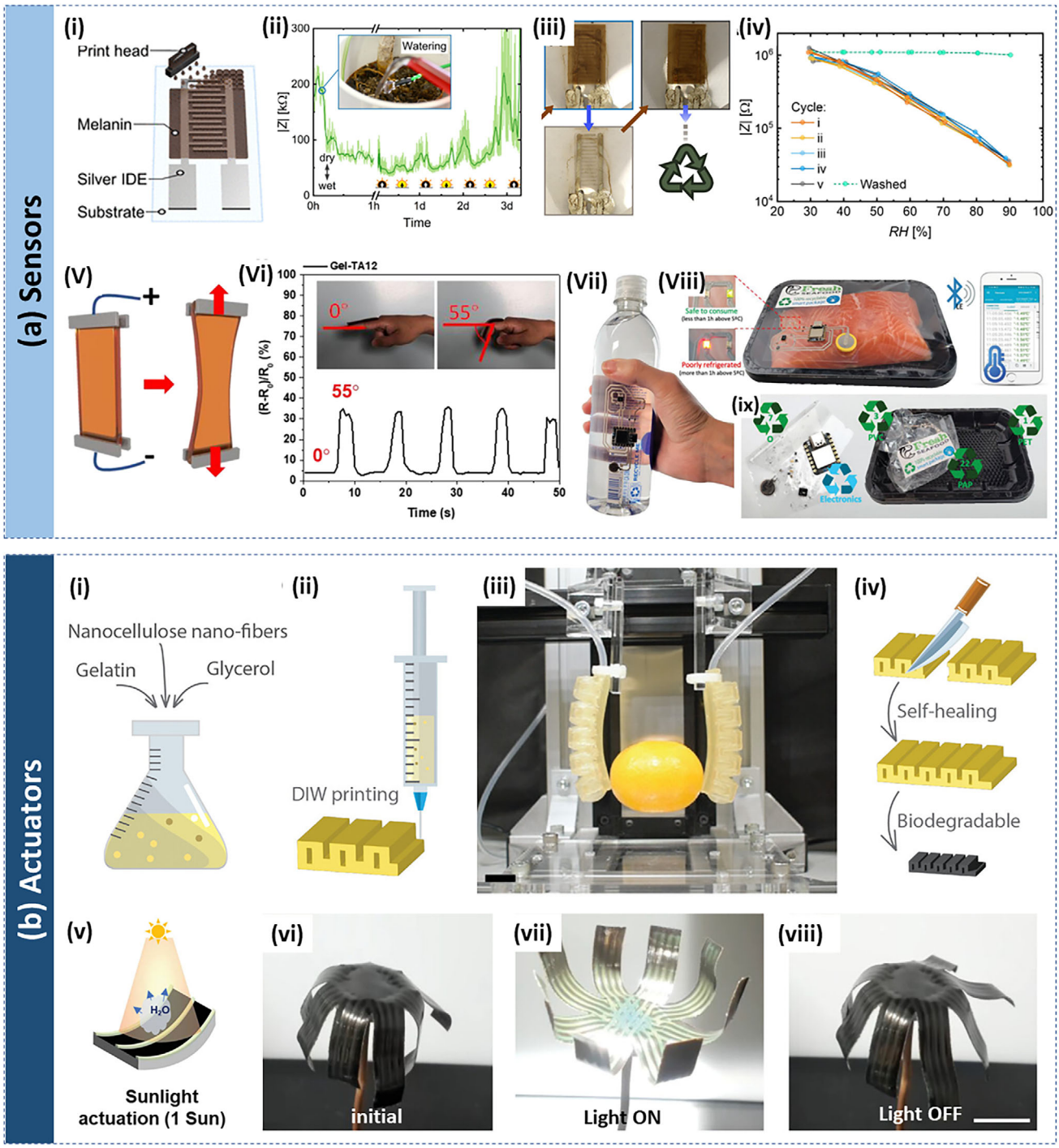


FIGURE 5 | Eco-friendly Sensors and Actuators. (a) Sensors: (i) schematic demonstration of the inkjet printing of melanin sensor. (ii) Recycling of the sensor by washing and reprinting of the melanin active layer and reusing the electrodes and (iii) the sensor demonstrated stable and repeatable characteristics after every reuse cycle. (iv) The deployment of the sensor for soil humidity monitoring of an indoor plant. Reproduced under the terms of the CC-BY license [137]. Copyright 2024, American Chemical Society. (v) Schematic demonstrating the working principle of printed gelatin/TA composite strain sensor. (vi) The change in resistance of the sensor attached to finger during the strain induced bending the finger at 55°. Reproduced with permission [172]. Copyright 2022, Elsevier. (vii) A recyclable smart package attached to a water bottle. (viii) A recyclable smart label with an LED indicator for quality monitoring. It can transfer the storage history and current temperature via Bluetooth to a smartphone. (ix) demonstration of separation and recycling of the smart labels. Reproduced under the terms of the CC-BY license [245]. Copyright 2023, John Wiley and Sons. (b) Actuators: (i) DIW printable ink formulation consisting eco-friendly material. (ii) DIW printing to fabricate the pneumatic actuators. (iii) Final gripper consisting of two pneumatic actuators and it exhibit (iv) self-healing and biodegradable properties. Reproduced under the terms of the CC-BY license [246]. Copyright 2025, John Wiley and Sons. (v) Working mechanism of photo-actuator consisting of photothermal NPs and hygroscopic actuators. The working of the actuators is demonstrated under (vi) normal, and (vii) sunlight conditions. (viii) The actuator returns to its initial state once the light is removed [247]. Reproduced under the terms of the CC-BY license. Copyright 2023, John Wiley and Sons.

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Sensors play a crucial role in ensuring food safety and quality by continuously tracking chemical, biological, and environmental indicators throughout the entire food supply chain. From harvesting to storage and consumption, real-time sensing helps detect spoilage, contamination, and degradation that may otherwise go unnoticed. Embedding eco-friendly sensors into food packaging provides a particularly powerful approach, enabling on-package monitoring of freshness while minimizing environmental impact. For example, integrating an ammonia sensor into packaging can offer early detection of food spoilage, supporting safer consumption and reducing unnecessary food waste [256]. One such sensor was fabricated using aerosol jet printing and flash photonic sintering to deposit carbon electrodes on cellulose substrates [248]. The device works by monitoring changes in ionic conductivity of absorbed water, since ammonia dissolves and dissociates in the water layer, increasing ion transport. The selective sensor demonstrated a 12% change in resistance at an ammonia concentration of 3 ppm. Rather than biodegradation, a fully recoverable and recyclable printed smart labels were demonstrated using combinations of water-based polyurethane (PU), Ag, and liquid metals (Figure 5a(vii–ix)) [245]. The approach could be beneficial where high performance electronics are necessary, however the materials are toxic, not intended for direct contact with food and uneconomical to discard after single use. Table 3 shows various digitally printed eco-friendly sensors that demonstrated some degree of sustainability together with its performance.

4.2.2 | Actuators

With modest reformulation, several of the materials introduced above for sensing can just as readily drive motion instead of detecting it. Actuators are devices that convert energy into mechanical displacement, torque, or force, and are widely used across diverse applications, including industrial automation, medical surgery, environmental monitoring, and robotics. They can be classified according to the type of energy that drives their motion, such as electric, pneumatic, or photo-responsive actuation [257]. However, end-of-life management for actuators has received limited attention; in most cases, they are simply discarded, contributing to environmental pollution. This underscores the need for eco-friendly actuator technologies, which are now a growing research focus.

Manufacturing actuators from bio-sourced materials offers significant promise for eco-friendly electronics. A notable example is a pneumatic actuator fabricated via DIW using gelatin and cellulose nanofibers dispersed in a glycerol–water solvent system [246]. Figure 5b(i) shows the formulation, (ii) DIW fabrication, and (iii) functioning of the pneumatic actuator. This approach employs nontoxic materials and solvents, and its mold-free fabrication minimizes material waste, enhancing environmental sustainability. The printed actuators are self-healable (Figure 5b(iv)), hence, could be repaired and reused over a longer period after the physical damage. Furthermore, their biodegradability allows safe disposal without environmental harm. Performance testing demonstrated that these eco-friendly actuators achieve up to 80° bending and generate a blocked force of 0.1 N at 15 kPa, comparable to conventional counterparts [246]. Similarly, a bio-sourced pneumatic actuator based on a calcium-alginate hydrogel has demonstrated tunable and long-term reliable performance. Such actuators hold strong promise for sustainable soft-robotic systems that can be safely deployed in aquatic environments, interact gently with delicate plants and animals, and ultimately biodegrade after mission completion, reducing ecological impact [174].

Actuators driven by environmental energy sources, such as light or humidity, are inspired by mechanisms found in plants and seeds, and hold significant potential for sustainable robotics and environmental monitoring. Such systems exemplify embodied intelligence, where sensing and actuation are inherently integrated within the material. For instance, photo-responsive liquid crystal elastomers combined with sustainable cellulose nanocrystals have been formulated into DIW printable inks for photo-actuator fabrication [258]. Similarly, composites of CMCand cellulose nanofibrils can respond to changes in hydration by swelling, functioning as hydration-responsive actuators [259]. A prototype valve utilizing this mechanism has demonstrated reversible opening and closing through hydration/dehydration cycles. Furthermore, by incorporating metallic nanoparticles as photothermal agents, the hygroscopic nature of CMC can be harnessed to develop photothermal actuators (Figure 5b(v–viii)) [247].

4.3 | Energy

4.3.1 | Generation

The demand for eco-friendly energy sources is increasing due to the urgent need to reduce environmental impact. Technologies such as photovoltaics (PV), piezoelectric (PENG), and triboelectric nanogenerators (TENG) are promising, yet their limited lifetimes and disposal challenges can create additional environmental concerns. In addition, the ecological impacts associated with raw material extraction and energy-intensive device fabrication remain insufficiently characterized. Therefore, a truly sustainable energy source must be environmentally responsible across its entire life cycle, from material synthesis and manufacturing to operation and end-of-life.

Among renewable energy technologies, PVs are central to sustainable energy production due to their ability to directly convert sunlight into electricity. Si is the most widely used semiconductor for PV and is the second most abundant element in the Earth's crust [260, 261]. However, despite its availability and nontoxicity, Si fabrication relies on energy-intensive, high-temperature processing, and hazardous chemical [262]. End-of-life disposal of Si PV modules further introduces pollution and recycling challenges [263, 264]. Printable PV technologies offer a scalable and low-cost route toward greener solar cells [265]. Yet, most printed PV materials, such as perovskites, organic semiconductors, and quantum dots, are still dependent on toxic elements, heavy metals, or nondegradable polymers. Although some progress has been made with eco-friendly ink formulations for digital printing, the active materials themselves remain environmentally problematic [202, 266, 267]. Recently, unconventional bio-PV technologies have emerged, in which photosynthetic microorganisms convert light energy into

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TABLE 3 | Various digital printed sensors demonstrating selected aspects of sustainability and its performance.

		Materials								
Refs.	**Sensing parameter**	**Active**	**Electrode**	**Substrate**	**Solvent(s)**	**Sustainability aspect of device**	**Fabrication technique**	**Sensitivity**	**Response/ Recovery time (s)**	**Detection range**
[248]	Ammonia	Carbon*	—	Cellulose paper	—	Eco-friendly materials	Aerosol-jet printing and flashlamp sintering	30.9%	—	0 to 12 ppm
[249]	Acetone	chitosan hydrogel	Ag* (Electrode/Antenna)	Bio-based PLA resin	—	Eco-friendly materials LCA [120]	Inkjet printing	63 Ω per ppm	—	0 to 31 ppm
[250]	NO2	Alkaline lignin-Edge oxidized graphene oxide*	—	Paper, polyster, PU, PVA, PI	DI water	Eco-friendly sensing material	DIW	1.70%/ppm LoD: 12.7 ppb	610 to 250s/-	1 to 100 ppm
[175]	Food freshness (pH, Ammonia)	Alginate-gelatin-nanocellulose hydrogel*	—	—	—	Eco-friendly materials	DIW	75	—	—
[251]	Pb	—	Ag*	Photopaper	DI water	Biodegradable device	Inkjet printing	LoD: 72.35 ppb	—	100 to 1000 ppb
[251]	Cd	—	Ag*	Photopaper	DI water	Biodegradable device	Inkjet printing	LoD: 111.89 ppb	—	100 to 1000 ppb
[214]	Glucose (OECT)	PEDOT:PSS*	Carbon, Zinc	PLA	Water, DMSO, (3-glycidyloxypropyl) trimethoxysilane	Bio-degradation in soil	Inkjet printing	−3.4 ± 0.6%/dec	—	1 μ to 1 mM
[244]	Glucose	PtAuNC@ consensus tetratricopeptide repeat (CTPR)-Graphene*	Ag*	Paper	Water, t-butanol, propylene glycol	Eco-friendly materials	Inkjet printing	1.67 μA cm^{-2} mM^{-1}	—	0.1–1 mM
[244]	H_2O_2	PtAuNC@CTPR-Graphene*	Ag*	Paper	Water, t-butanol, propylene glycol	Eco-friendly materials	Inkjet printing	32.30 μA cm^{-2} mM^{-1}		0.05–1 mM

(Continues)

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TABLE 3 | (Continued)

Refs.	Sensing parameter	Materials				Sustainability aspect of device	Fabrication technique	Sensitivity	Response/ Recovery time (s)	Detection range
		Active	Electrode	Substrate	Solvent(s)					
[136]	Humidity	Nanosized activated carbon*	Au*	Tattoo paper	Ethanol–terpineol	Fully based on renewable sources	Inkjet printing	9.2 Ω/%RH	297.6/146.6 s	40% to 90%
[252]	Humidity	PEDOT:PSS-(3-glycidyloxypropyl) trimethoxysilane (GOPS)*	PEDOT:PSS*	Cellulose	EG	Eco-friendly materials	Aerosol-jet printing	148.7%	—	20% to 90%
[188]	Humidity	BSF melanine(ChCl)*	Ag*	polyethylene naphthalate (PEN)	DI water, TGME	Reuse PEN and Ag electrodes	Inkjet printing	170	0.8/0.8	30% to 90%
[137]	Humidity	HPC($[Bmim]_2$ [NiCl4])*	Ag*	Cellulose acetate	Water, EG	Eco-friendly materials	Inkjet printing	163	0.8/7.2	30% to 90%
[253]	Strain	CNT*	—	PU	—	Water-based ink formulation	Inkjet printing	Gauge factor (GF):~3.3	—	0% to 20%
[254]	Strain	PDMS (Carbon black) *	—	Textiles	Water, DMSO, (3-glycidyloxypropyl) trimethoxysilane	NA	DIW	GF: 7 to 150	—	4% to 28%
[172]	Strain	Gelatin-Tannic acid-ChCl*	—	Ecoflex	Water, t-butanol, propylene glycol	Self-healable	EHD printing	GF: ~0.325	—	200%
[255]	Touch	—	PEDOT:PSS*	Nanopaper	Ethanol–terpineol	Material recovery and reuse	Inkjet printing	—	—	—
[252]	Temperature	PEDOT:PSS-GOPS*	PEDOT:PSS*	Cellulose	EG	Eco-friendly materials	Aerosol-jet printing	6.8%	—	20 to 50°C
[243]	Temperature	GO-lignin*	—	Paper	Water, t-butanol, propylene glycol	Eco-friendly materials and water-based ink	Inkjet printing	TCR: 1.21% K−1	—	25 to 52°C

*Material layers that are printed.

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electrical power, offering self-sustaining and self-maintaining operation [268–270]. One example is an inkjet-printed bio-PV device where conductive CNT inks were used to form anode and cathode electrodes on paper, followed by printing cyanobacterial cells on the anode and a hydrogel layer on top (Figure 6a(i,ii)) [268]. The microorganisms harvest light energy to oxidize water, releasing electrons and protons. The electrons travel through an external circuit to generate power, while the protons migrate through the nanoporous paper to the cathode, maintaining electroneutrality. Figure 6a(iii) shows the mechanism. Unlike conventional PVs, bio-PVs deliver a peak output power of 0.38 mW m^{-2} under illumination and can still generate 0.22 mW m^{-2} in the dark (Figure 6a(iv)), as the bacteria continue to release electrons during respiration from stored nutrients. However, the power output remains lower than that of conventional PVs, underscoring that the development of fully eco-friendly and reliable printed commercial PV devices, combining green materials, sustainable processing, and safe end-of-life strategies remains as a challenge and an important direction for future.

Mechanical energy harvesters, such as PENGs and TENGs, have emerged as promising candidates because they can utilize abundant mechanical energy sources from human motion, vibrations, or other ambient forces. Their intrinsic sensitivity to pressure, strain, and other mechanical motions allows them to operate simultaneously as self-powered sensing elements and hence ideal for wearable health monitoring, human–machine interfaces, and soft robotics. Electric dipoles are generated in materials with non-centrosymmetric crystal structures when subjected to mechanical stress, a phenomenon known as the piezoelectric effect, which can be exploited for energy harvesting. PENGs utilize this principle, and demonstrated using bio-derived piezoelectric materials. For example, a bio-ceramic extracted from chicken bone exhibited a piezoelectric coefficient of ~68.7 pC N^{-1} and was extrusion printed between Cu electrodes to fabricate a sustainable PENG [271]. Figure 6a demonstrated concept of (v) chicken bone extract-based PENG (vi) fabricated using DIW technology and (vii, viii) its piezoelectric performance. Cellulose nanocrystals, one of the most abundant biopolymers on Earth, were extrusion-printed between CNT electrodes and have been shown to exhibit a piezoelectric response as well as a pyroelectric effect, enabling energy harvesting from thermal fluctuations. With a mechanical sensitivity of 0.8 V kPa^{-1} and a pyroelectric coefficient of 0.58 μC m^{-2} K^{-1}, the device shows strong potential as a wearable energy source, capable of extracting energy from both human body temperature and mechanical motion, as well as serving as a self-powered sensor [272]. Similarly, TENGs, which operate via contact electrification and separation, have been fabricated using biopolymers extracted from rapeseed and soybean oil (Figure 6a(ix)) [273]. A 3D-printed TENG based on these biopolymers achieved a charge density of 0.211 nC cm^{-2}, a current of 70 nA, a peak voltage of 22 V under a 10 GΩ load, and a maximum power density of 340 μW m^{-2} at 100 MΩ. Importantly, this device demonstrated significant degradation within 90 days in 1 M NaOH solution, highlighting its degradability (Figure 6a(x, xi)). Extrusion printed nanocellulose was also used for TENG applications [274]. Although their performance still lags conventional devices, such developments demonstrate progress toward eco-friendly mechanical energy harvesting.

4.3.2 | Storage

On-site energy generation alone is often impractical, as the availability of sources such as solar, thermal, or mechanical energy is intermittent and cannot always meet demand. To ensure continuous operation of electronic systems, the generated energy must be stored and later delivered when required. Energy storage devices, particularly supercapacitors and batteries, play a crucial role by buffering the mismatch between energy supply and consumption. Supercapacitors store charge electrostatically at the electrode-electrolyte interface (electric double-layer capacitance) or via fast surface redox reactions (pseudo-capacitance), giving rapid charge/discharge at the cost of lower energy density, while the batteries instead store charge through bulk electrochemical redox reactions and ion intercalation, trading charge/discharge speed for higher energy density [275]. Their integration with eco-friendly energy harvesting technologies is therefore essential to move toward sustainable electronic systems. However, conventional storage devices often rely on heavy metals such as Co and Mn in electrodes and employ toxic organic solvents such as ethylene carbonate in liquid electrolytes [276–278]. The associated environmental and biological risks highlight the urgent need for safer alternatives and thorough investigation of their long-term impacts.

One promising direction toward safer energy storage is the development of biodegradable supercapacitors. For instance, a micro-supercapacitor fabricated using extrusion printing with activated carbon and graphene electrodes on a polycaprolactone/starch composite substrate, combined with a LiCl-based electrolytes, demonstrates an environmentally conscious design [279]. In such systems, renewable and degradable carboxymethyl cellulose has been employed as a binder for the conductive electrodes, while bentonite, a natural clay component, is combined with LiCl to form the electrolyte (Figure 6b(i)). The device exhibited a specific capacitance of 250 mF cm^{-2} with 67.1 μWh cm^{-2} energy density (Figure 6b(ii)) and can completely degrade in 90 days when buried in natural soil. Figure 6c(iii,iv) shows the natural degradation, indicated by the percentage reduction in mass over time.

Table 4 summarizes the properties of various supercapacitors, demonstrating selected aspects of sustainability. The majority of these devices utilize carbon-based electrodes, owing to their high electrical conductivity, tunable porous structures for large surface area, and excellent thermal and chemical stability. Most reported devices adopt a symmetric configuration, in which both anode and cathode electrodes are made from the same active material, as it simplifies fabrication, ensures balanced charge storage, and provides stable cycling performance. Inkjet and DIW are typically selected based on the film thickness and pattern resolution requirements. Notably, the energy density and capacitance retention of these eco-friendly devices are comparable to those of printed nonsustainable supercapacitors [281]. Furthermore, the use of biodegradable and renewable materials not only reduces production costs but also mitigates challenges related to end-of-life management. Importantly, the table also demonstrates that such eco-friendly supercapacitors retain more than 87% of their initial capacitance even after 10,000 charge–discharge cycles, significantly exceeding the durability requirements specified under EU eco-design regulations for energy storage systems, which

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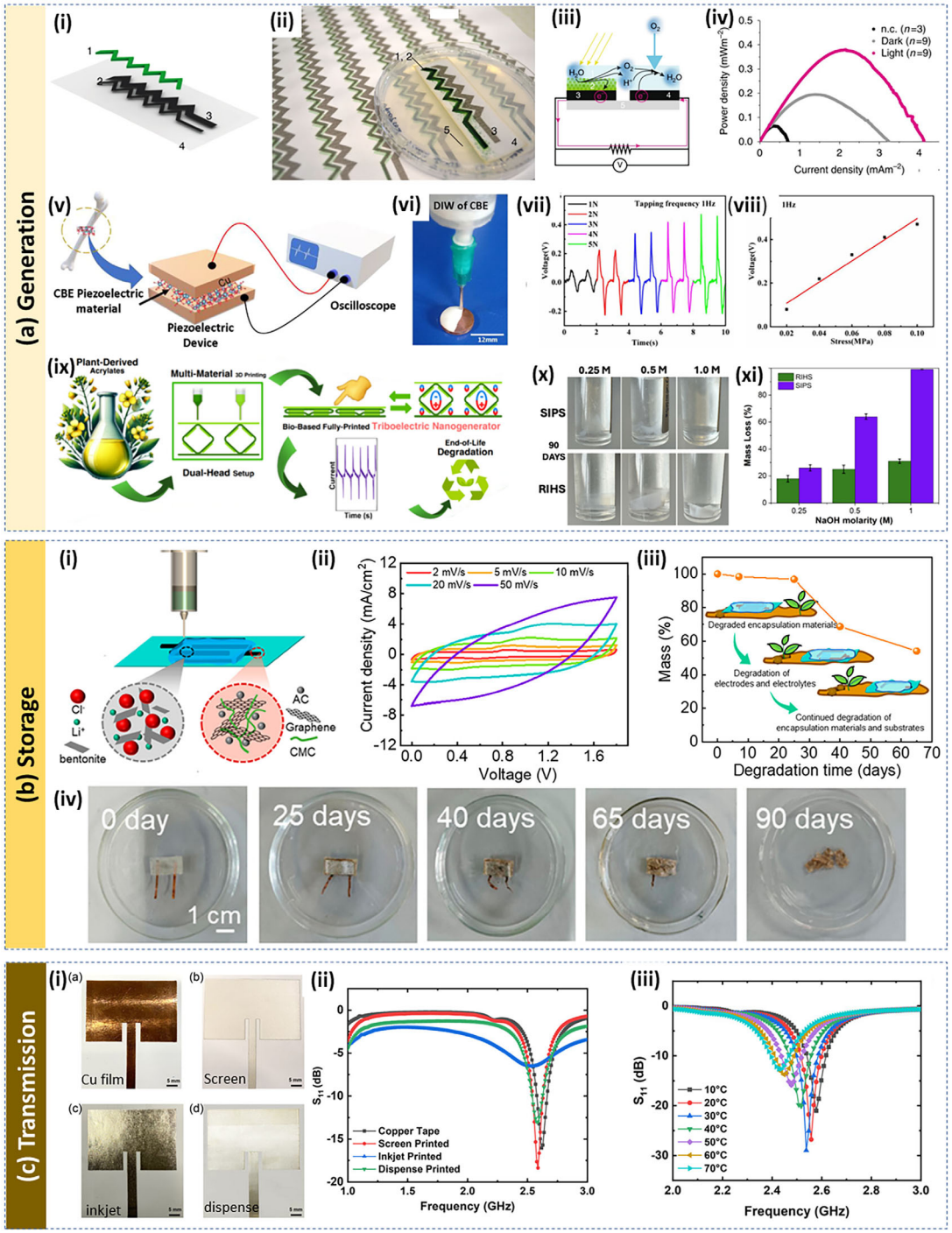


FIGURE 6 | Digital printed Eco-friendly Energy devices. (a)Energy Generation: (i) schematic design of fully printed bio-PV system and (ii) Photograph of the array (1: Photosynthetic micro-organisms; 2: CNT anode; 3: CNT cathode; 4: Paper substrate). (iii) Schematic showing the working of the bio-PV. (iv) Power density of the bio-PV without and with printed microorganism and under dark and light conditions. Reproduced under the terms of the CC-BY license [268]. Copyright 2017, Nature. (v)Schematic showing the concept of PENG produced from chicken bone extract. (vi) DIW fabrication of the PENG. (vii) Voltage generated under various tapping forces and (viii) the linear relation between the generated voltage and applied stress. Reproduced with permission [271]. Copyright 2023, American Chemical Society. (ix) Schematic showing the concept of biodegradable TENG produced from plant-derived acrylates. (x) Photographs showing the degradation of the TENG after 90 days and (xi) its mass loss at the end of experiment. Reproduced under the terms of the CC-BY license [273]. Copyright 2025, American Chemical Society. (b) Energy Storage: (i) schematic illustration of the DIW fabricated biodegradable micro-supercapacitor. (ii) The CV curve demonstrating the electrochemical performance at room temperature with various scan rates. Degradation behavior of the micro-supercapacitor in natural soil, (iii) change in mass (%) with different time and (iv) the photograph. Reproduced with permission [279]. Copyright 2023, American Chemical Society. (c) Energy transmission. (i) Photographs of antennae fabricated using (a) conventional Cu tape, (b) screen printed Ag, (c) inkjet printed Ag, and (d) extrusion-printed Ag. (ii) The reflection coefficient of the various antennas. (iii) the effect of temperature on antenna performance at a constant relative humidity of 30%. Reproduced under the terms of the CC-BY license [280]. Copyright 2023, Wiley.

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TABLE 4 | Properties of digital printed supercapacitors demonstrating selected aspects of sustainability.

	Materials										
Refs.	**Current collector**	**Electrode**	**Electrolyte**	**Separator**	**Substrate**	**Solvent**	**Sustainability aspect of device**	**Printing technique**	**Specific capacitance**	**Energy density**	**Capacitance retention**
[279]	—	Carboxymethyl cellulose (Activated carbon -graphene)*	LiCl -bentonite*	—	Polycaprolactone/ starch	Glycerol, EG, DI water	Biodegradation demonstrated in natural soil	DIW	251 mF cm^{-2}	67.1 μWh cm^{-2}	—
[283]	Graphite*	Dopamine grafted bare activated carbon*	ChCl -urea*	—	PLA	DMF, DI water	Eco-friendly materials	DIW	96 F g^{-1}	22 Wh kg^{-1}	87% after 10,000 cycles
[284]	Graphite*	Catechin modified bare activated carbon *	ChCl -urea*	Cellulose*	PLA	Water, acetic acid, dimethylacetamide	Eco-friendly materials	Inkjet	75 F g^{-1}	20.6 Wh kg^{-1}	90.2% after 10,000 cycles
[196]	—	Activated carbon- super P conductive carbon- Na CMC *	Na_2SO_4 - Na CMC*	—	Photopaper	DI water	Water-based ink formulation	DIW	127.8 mF cm^{-2}	4.95 mWh cm^{-2}	88.6% after 10,000 cycles
[285]	Ag*	MXene–PH1000*	($LiCl/SiO_2$) gel*	—	Photopaper	Water, glycol	Water-based ink formulation	Inkjet	22.6 mF cm^{-2}	0.56 μWh cm^{-2}	87.4% after 5,000 cycles

*Material layers that are printed.

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mandate at least 800 cycles with 80% capacity retention [282]. This highlights their potential not only as sustainable alternatives but also as highly durable energy storage solutions aligned with emerging regulatory standards.

When comes to batteries, an inkjet-printed, environmentally benign Zn-ion battery (ZIB) has been demonstrated on a paper substrate [286]. The device features a planar architecture, with Zn as the anode and MnO_2 as the cathode, integrated using PEDOT:PSS as a binder. rGO serves as the current collector, while a drop-cast gel electrolyte composed of $ZnSO_4$, $MnSO_4$, and PVA provides ionic conductivity. The fabricated ZIB delivers a high discharge capacity of 300.14 mAh g^{-1} at a current density of 200 mA g^{-1}. Moreover, the device exhibits mechanical robustness, retaining nearly 100% of its capacity under repeated bending and folding conditions. These attributes underscore its strong potential as a sustainable power source for emerging flexible and wearable electronics, including applications in healthcare monitoring, soft robotics, and distributed IoT platforms.

4.3.3 | Transmission

Alongside energy generation and storage, transmission plays an equally relevant role in electronic systems and is typically implemented via transmission lines and antennas. Efficient transmission requires minimizing resistive losses and impedance mismatches, for which metals are generally preferred because of their high conductivity, ease of design, and structural uniformity. Printed metallic NP inks on eco-friendly substrates represents an approach toward eco-friendly transmission devices [287, 288]. For example, a passive radio frequency identification (RFID) antenna fabricated by inkjet printing Ag NP ink onto uncoated paper and operating at 1 MHz demonstrated wireless sensor readout up to 3 mm via inductive coupling. In this system, an alternating magnetic field generated by the reader induces a voltage in the sensor's resonant circuit, and changes caused by sensing events are reflected back to the reader signal, enabling data extraction [289]. In another example, a printed antenna array (15 × 5) composed of Ag NP conductive elements and PEDOT:PSS-based resistive transmission lines on paper substrates was used for microwave nondestructive evaluation of thin dielectric composites [290]. Beyond noble metals, additive-free titanium carbide MXene inks processed by extrusion printing have been demonstrated for near-field communication (NFC) antennas and RFID sensors, offering a metal-free alternative with high conductivity [291]. Reactive inkjet-printed rGO dipole antennas operating at 2.4 GHz have also shown promise for WiFi applications, achieving data transmission speeds of up to 4.6 Mbps, comparable to commercial counterparts [292]. Such demonstrations highlight the potential of printed antennas for sustainable smart packaging, wireless monitoring, and energy harvesting.

The choice of printing technique plays a decisive role in determining antenna performance, as it directly influences film morphology, electrical conductivity, and radiation efficiency. Parameters such as film thickness, surface roughness, ink spreading, and edge definition vary across printing methods, thereby influencing electrical resistance, design fidelity, and overall efficiency. Figure 6c(i) shows the comparative studies of Ag antennas operating at 2.6 GHz fabricated via screen, inkjet, and extrusion printing [280]. The study reveals that screen and extrusion printing yield thicker films with lower sheet resistance, whereas inkjet printing, limited by low ink viscosity and thin film formation, typically results in higher resistance. As expected, increased film thickness correlates with reduced resistance and improved antenna performance, a trend reflected across multiple printed antenna designs (Figure 6c(ii)). However, at higher operating frequencies, surface smoothness becomes increasingly important due to skin-effect losses, where inkjet printing offers a relative advantage by producing smoother conductive features compared to thicker but rougher printed films.

Despite such advances, further progress requires the development of sustainable conductive materials beyond metallic nanoparticles. Moreover, eco-friendly substrates such as paper introduce additional challenges, including high dielectric loss tangent and dielectric instability under varying temperature and humidity, both of which compromise radiation, or transmission efficiency [280]. For instance, Figure 6c(iii) demonstrates the performance variation of a paper-based antenna with temperature at constant humidity, highlighting this instability. A practical short-term pathway may involve the selective use and recovery of non-biodegradable substrates and conductive materials to balance sustainability with performance until fully biodegradable high-performance alternatives are developed [293, 294].

5 | End-of-Life Management

As discussed in previous sections, the growing demand for low-cost electronic devices has accelerated resource depletion and generated large quantities of e-waste. E-waste contains valuable and critical materials that are frequently discarded rather than recovered, despite their significance [295–297]. Furthermore, unmanaged e-waste can release hazardous substances that pose significant environmental and ecological risks. Consequently, robust end-of-life management is essential. A circular electronics framework that targets near-complete recovery and reuse of materials and energy offers a viable path toward sustainability. Recent work on digitally printed electronics identifies three primary end-of-life management strategies: (i) extending device lifetime through repair and reuse, (ii) recycling and recovering materials for refabrication, thereby delaying the return of materials to the environment, and (iii) environmentally responsible degradation of nonrecoverable components. Therefore, the end-of life of electronics depends not only on materials selection but also on fabrication methods, interfacial adhesion, and layer architecture and compatibility [298].

Repairable electronics are particularly promising, as they allow damaged components to be restored with minimal material and energy input. Self-healing materials are especially advantageous for this purpose. These materials can autonomously recover from mechanical damage, enabling long-term device operation (Figure 7a). Digitally printed self-healing gelatin-based sensors and actuators have been demonstrated [172, 246, 299]. For example, an EHD-printed gelatin/tannic acid elastomer has been shown to recover more than 95% of its mechanical and electrical performance after being cut into two pieces and allowed to

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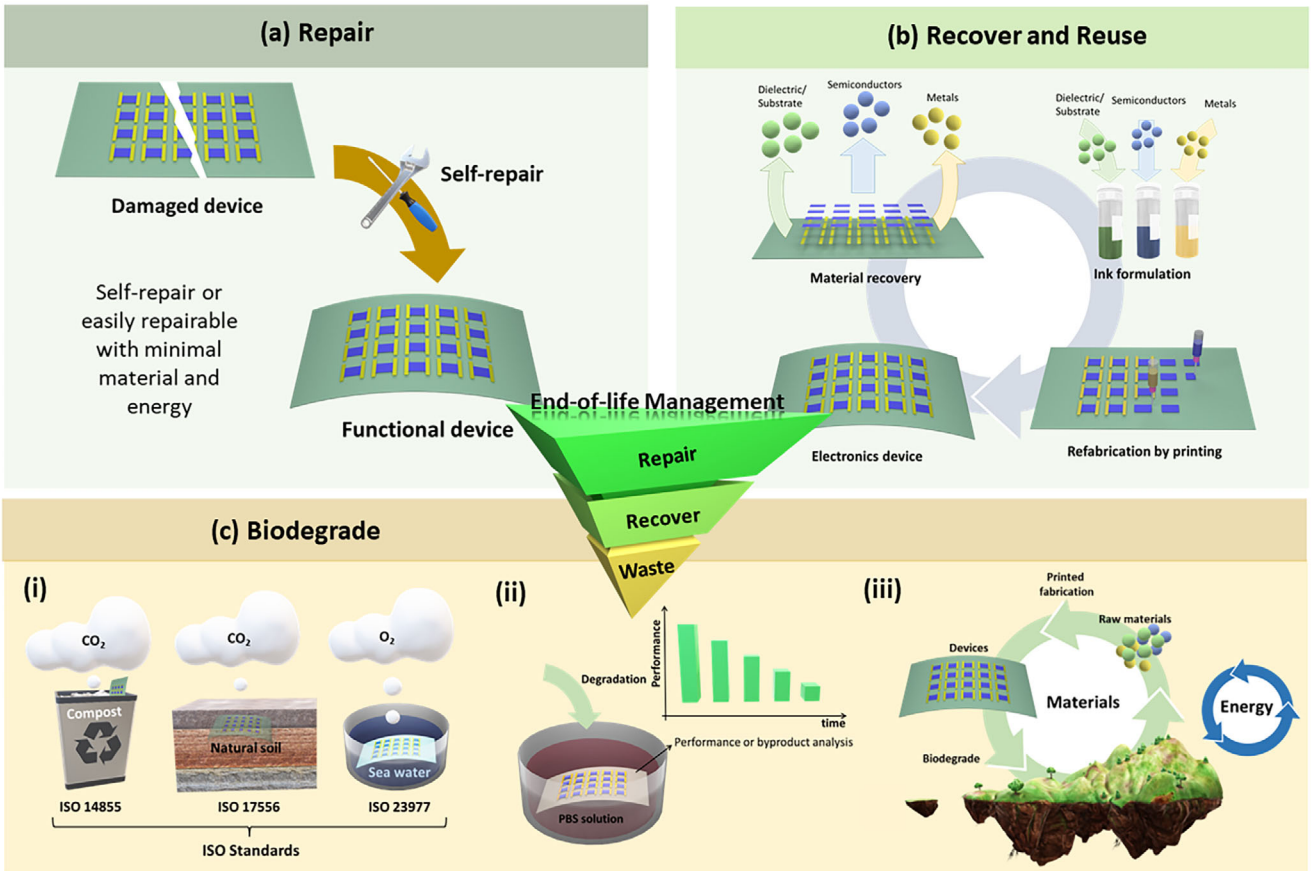


FIGURE 7 | End-of-life management strategies for digitally printed electronics. (a) Self-repairable or easily repairable devices. (b) Recycling and recovery of materials for refabricating new devices. (c) Methods for assessing device degradability: (i) ISO standards for evaluating material biodegradation, (ii) accelerated degradation testing in aqueous NaCl or PBS solutions, and (iii) life-cycle assessment of electronics considering materials and energy.

self-heal for 48 h without any external treatment [172]. This self-healing behavior arises from dynamic hydrogen bonding between the gelatin chains and tannic acid molecules. Because these bonds can break and reform dynamically, the network reconnects after damaged.

If devices are not easily repairable, an alternative strategy is to recover and reuse functional materials and components in subsequent fabrication cycles (Figure 7b) [300, 301]. Recovery of Ag and MoS_2 from inkjet-printed memristors has been demonstrated, enabling reformulation of functional inks and refabrication of devices [216]. In this approach, printed devices on paper were first bath-sonicated in triethylene glycol monomethyl ether (TGME) to detach the Ag electrodes. The resulting Ag–TGME dispersion was collected by vacuum filtration and further dispersed by sonication. The dried paper substrate was then sonicated in IPA to remove the inkjet-printed MoS_2, while an additional IPA wash of the initial filter paper recovered any MoS_2 flakes retained during Ag extraction. The combined MoS_2 dispersions underwent solvent exchange via high-speed centrifugation, followed by a second vacuum filtration to isolate the flakes. Both the recovered Ag nanoparticles and MoS_2 flakes were subsequently re-dispersed to formulate fresh inks for printing new devices [216]. Reusability is particularly important for precious metals and non-eco-friendly polymers, while recovery of biodegradable components may be unwarranted if the process requires excessive energy. One such example is the reuse of inkjet-printed Ag electrodes and PEN substrates in humidity sensors, where both components have been redeployed for subsequent fabrication cycles whereas melanin-based active materials were discarded without recovery [188]. Similar benefits have been reported for conventional PCB components, which are typically produced with high material and energy demands but can be reused without compromising reliability [245, 302]. Additionally, directly printed Ag conductive traces and soldering pastes have been recovered and reused in ink formulations [245]. These reuse-oriented approaches are aligned with emerging European policies such as the "Right to Repair/Return", which emphasize prolonged device lifetimes, material recovery, and the systematic reintegration of functional components into new electronic products [303]. Such frameworks reinforce the relevance of designing printed electronics with recoverable materials and architectures that support multiple reuse cycles.

The third end-of-life strategy focuses on employing biodegradable materials, which can naturally decompose into environmentally benign constituents that reintegrate into ecosystems. Over longer timescales, these materials may be recovered as secondary raw materials for future electronics fabrication [74]. To support this approach, the International Organization for Standardization (ISO) has established several experimental standards to quantitatively assess the biodegradability of materials under

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various environmental conditions (Figure 7c(i)) [304]. ISO 14855 evaluates ultimate aerobic biodegradability under controlled composting conditions by measuring CO_2 evolution, with ISO 14852 and ISO 17556 offering comparable procedures for aqueous media and natural soil environments, respectively [305]. Testing is typically conducted at thermophilic temperatures (~58°C) with controlled aeration and humidity, where samples are mixed with an inoculum and composted for up to 6 months. Biodegradation is quantified by comparing the measured CO_2 with the theoretical amount expected from complete mineralization. A material is generally classified as biodegradable when it achieves a defined level of degradation, such as 90% within 180 days. The biodegradation rate is usually benchmarked against a reference biodegradable material to enable quantitative comparison. A notable example is the ISO 14855 biodegradability assessment of an inkjet-printed electrochromic display [170]. Here, the display and a micro-cellulose reference were composted under controlled conditions. The display exhibited a significantly faster degradation rate and reached a higher final biodegradation level (~79% in 9 weeks) compared to the reference, which required two additional weeks to meet the ISO validation threshold. ISO 23977 provides a complementary method for assessing aerobic biodegradation in natural seawater using a closed respirometer, in which oxygen consumption by native marine microorganisms is monitored while CO_2 is absorbed by an alkali trap, allowing direct measurement of biochemical oxygen demand (BOD). Examples include inkjet-printed polymer FETs demonstrating clear degradation behavior under ISO 23977 seawater conditions [135, 218]. Although these standards were originally developed for plastics and do not capture all biodegradation aspects relevant to electronic devices, their applicability to printed electronics remain largely unexplored and represents a viable direction for future research [306, 307].

Standardized ISO protocols are not the only way to assess biodegradation: in vivo soil testing offers a complementary, more realistic route, where devices or materials are buried in natural soil and monitored over time [214, 279]. This method provides a more realistic assessment of environmental degradation, as it exposes the material to natural microorganisms, moisture levels, temperature fluctuations, and soil chemistry. Performance degradation of the device is periodically measured to determine the rate and mechanism of breakdown under real-world conditions. Complementary to this, ex vivo testing is often performed using controlled corrosive buffer solutions, such as aqueous NaCl, PBS, or other ionic media (Figure 7c(ii)) [206]. These solutions accelerate degradation by offering controlled chemical environments that replicate key features of natural exposure, thereby enabling precise analysis of degradation kinetics.

By combining eco-friendly materials with digitally printed fabrication, the field is steadily progressing toward electronics that are both high-performance and environmentally responsible. However, a full LCA must look beyond material reuse alone [308]. Ideally, printed devices should biodegrade safely in soil, allowing their constituent materials to be naturally recovered over time and ultimately reused as raw materials for manufacturing new devices (Figure 7c(iii)) [309].

However, quantitative LCAs have demonstrated that the sustainability of an end-of-life route directly depends on the complexity of the individual components. For instance, hybrid printed systems that integrate printed traces with conventional integrated circuits (ICs) can reduce the global warming footprint around 50% through recycling ICs [310]. On the other hand, simpler fully printed devices achieve the highest reduction in the environmental impact. In addition, the choice between material degradation or recycling is influenced by the use of renewable and abundant materials. Currently, recycling printed electronics is often highly favorable in order to recover scarce materials such as silver [311].

Nevertheless, even biodegradable materials require proper disposal. For instance, if printed electronics based on substrates of polylactic acid (PLA) or thermoplastic starch (TPS) end up in anaerobic landfills instead of industrial composting facilities, they produce methane. Hence the global warming potential generated is higher than that of recycling or incinerating fossil-based plastic substrates with energy recovery [312].

When evaluating the sustainability of digitally printed electronics, material-specific life-cycle impacts must also be considered, as materials that are often presented as eco-friendly alternatives may still involve energy-intensive synthesis routes. Reported LCA data show that carbon-based fillers exhibit different environmental footprints depending on their structure and production method. CNTs generally show the highest impact, followed by graphene, whereas carbon black exhibits substantially lower reported energy demand and global warming potential (GWP) values [313]. Similarly, bio-derived nanomaterials such as CNCs and CNFs can reduce reliance on fossil-based resources, but their environmental benefit strongly depends on the extraction route, chemical treatment, feedstock and process scale. For instance, acid hydrolysis, TEMPO oxidation, or poor reagent recovery can substantially increase the GWP of cellulose-based nanomaterials, whereas enzymatic routes, waste-derived feedstocks, and large-scale processing can reduce their impact.

These examples highlight that sustainability cannot be considered only from the renewable origin, biodegradability or carbon-based nature of a material. Instead, quantitative indicators such as synthesis energy demand, GWP, solvent use, reagent recovery and end-of-life assumptions should be reported whenever possible. Table 5 summarizes representative LCA-related values for common materials and processes used in printed electronics, illustrating the wide variability in environmental impact and the need for standardized reporting.

Circularity should also include energy considerations, since recovery processes must not consume more resources than they save [29, 314, 315]. For example, an end-of-life strategy cannot be considered sustainable if the energy required to separate, recover, or degrade a small electronic component exceeds the environmental benefit gained from material recovery. Therefore, LCA should be integrated early in the design of printed electronic devices to identify environmental hotspots across raw material extraction, ink formulation, printing, post-processing, use, and end-of-life stages [316]. This approach can guide the selection of materials, fabrication routes, and device architectures toward lower overall ecological footprints [317]. However, as summarized in Table 5, reported LCA values remain highly dependent on assumptions, system boundaries, feedstocks, processing routes,

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TABLE 5 | Representative LCA-related indicators reported for materials and processes relevant to digitally printed electronics. The values illustrate the strong dependence of environmental impact on material type, synthesis route, feedstock, process scale, and device architecture. Energy demand is expressed as MJ kg^{-1} and GWP as kg CO_2-eq kg^{-1} when available.

Material/process	Reported energy demand (MJ kg^{-1})	Reported GWP	Main sustainability consideration
CNTs [318, 319]	Up to 4.09×10^5	2.85×10^4 – 1.53×10^5 kg CO_2-eq kg^{-1}	Highest reported footprint among the compared carbon fillers; impact strongly depends on synthesis route.
Graphene [319]	300–1879	3.4–146 kg CO_2-eq kg^{-1}	Lower impact than CNTs but still highly method-dependent; renewable precursors may reduce the footprint.
Carbon black [319]	88–89	2.36 kg CO_2-eq kg^{-1}	Lowest reported energy demand and GWP among the listed carbon fillers, although feedstock origin remains important.
CNCs, pilot scale [319]	—	21–30 kg CO_2-eq kg^{-1}	Impact depends on feedstock, acid hydrolysis route, acid recovery, and processing scale.
CNCs, large scale [320]	—	7.5–12 kg CO_2-eq kg^{-1}	Scale-up can reduce GWP compared with pilot-scale production.
CNFs by TEMPO oxidation [320]	—	190–1160 kg CO_2-eq kg^{-1}	Chemical oxidation can strongly increase environmental impact.
CNFs from enzymatic/waste-derived routes [311]	—	Down to 4.41 CO_2-eq kg^{-1}	Enzymatic processing and waste feedstocks can substantially lower GWP.
CNC coating for paper-based electronics [311]	—	CNCs contribute >99% of total environmental impact	Bio-derived components can dominate the total impact if their extraction route is intensive.
CNC-reinforced eco-friendly resin [321]	—	~3× lower GWP than commercial resin	CNC reinforcement can reduce impact when combined with suitable resin systems.
Printed PEDOT:PSS vs. lithography [322]	—	Up to 99% GWP reduction	Avoids highly fluorinated solvents used in photoresists and developers.
Printed PEDOT:PSS replacing Ag paste/ITO [322]	—	10–85% lower GHG emissions	Can reduce device-level emissions by replacing higher-impact conductive materials.

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and reporting practices, highlighting the need for more standardized sustainability metrics in printed electronics.

6 | Challenges and Future Perspective

This section discusses the major challenges in realizing eco-friendly electronics, focusing on the limitations of sustainable materials, fabrication techniques, and transient devices, while also highlighting future research perspectives. Figure 8 highlights the various challenges and opportunities.

6.1 | Materials

In terms of sustainability, the emerging materials discussed here, processed by digital printing, align with the principles of *green* chemistry [323] and *green* engineering [324]. Despite rapid progress in eco-friendly printed electronics, several material-related challenges still limit their large-scale adoption.

A first issue is the scalability problems that obstruct their fabrication beyond the laboratory. Three main factors hinder this development: (i) lack of mass production of raw materials and their immature supply chains that increase costs, (ii) the bio-based inks and additives often show poor stability over time, and (iii) the complex ink development to adapt inks to high-speed printing or roll-to-roll manufacturing. However, the availability of the raw materials and the accessibility to printing techniques can decentralized and distributed the production, which can partially mitigate these challenges. Moreover, the widespread availability of renewable feedstocks such as cellulose, chitin, or lignin enables local or regional production of functional inks, helping to shorten supply chains, reduce transport-related emissions, and lower dependency on centralized raw material manufacturing. In addition, bio-derived inks are typically processed without high-temperature curing, requiring only mild drying steps, which reduces post-processing energy consumption and relaxes compatibility constraints with large-scale industrial equipment. Finally, small- to medium-scale production facilities can more easily adapt ink formulations to specific printing techniques, alleviating some of the formulation complexity associated with high-throughput manufacturing. Together, these aspects contribute to material- and energy-efficient production routes, while promoting regional resilience and reduced environmental impact.

A further challenge is the reduced performance of eco-friendly materials, which show lower electrical conductivity, lower thermal or chemical resistance, and shorter device lifetime that the conventional counterparts. Particularly, naturally sourced materials show greater variability, making it hard to guarantee uniform mechanical or electrical properties from one batch to another. Yet, their inherent instability can be exploited advantageously in transient electronics, devices intentionally designed to disintegrate in certain conditions after use. This property is particularly attractive for short-lived applications such as biomedical sensors, environmental monitoring, or smart packaging, where controlled degradation can prevent e-waste accumulation. The key opportunity lies in developing predictable, complete, and nontoxic degradation pathways, ensuring environmental safety and effective resource circularity.

Biopolymer-based inks represent a promising route toward more sustainable extrusion-printable materials. In this regard, polysaccharides and protein-based polymers such as alginate, gelatin, and chitosan are particularly attractive because of their aqueous processability, gel-forming ability, biocompatibility, and potential biodegradability. As discussed in Section 3.2., alginate- and gelatin-based formulations have already been explored in printed electronics [175]. However, chitosan has also been investigated in electronic devices as insulator [249] and as electrolyte [210, 217] although its use in DIW-printed electronic components remains less developed. By contrast, chitosan-based formulations have been printed by DIW for biomedical applications, demonstrating their suitability for extrusion-based fabrication of 3D structures. Therefore, materials based on printable polysaccharides, which have already demonstrated their usefulness in electronics, could be reproducibly manufactured with precise control of the structure and geometry.

Another key issue is the recovery and reuse of multimaterial structures. Printed devices typically require multilayer stacks of conductive, dielectric, protective, and/or sensing materials and their composites. This increased complexity reduces the possibility of material recovering for its recycling or reuse at the end-of-life stage. Nevertheless, this challenge is currently being tackled by design-for-recycling principles, where printed layers and polymer substrates can be separated and reused by the selective dissolution of specific layers, extending the life cycle of resources and reducing e-waste generation [325]. The coatings can be based on unstable covalent bonds (Diels Alder or disulfides), noncovalent interactions (hydrogen bonds and ionic interactions), and modifying the chain structure or lateral functionalities by inserting highly soluble segments [326]. In these cases, complete biodegradability of all electronic components is not desirable, because long-term applications could benefit more from the recovery of targeted material. Hybrid approaches, combining recyclable conductors with biodegradable substrates and protective layers, can offer a balanced compromise between stability, functionality, and end-of-life management, contributing to the development of long-lasting and circular printed electronics.

Finally, there still is a lack of clear sustainability metrics and standardized LCA methods for printed electronics. Although some screening tools, exists most studies use different assumptions and indicators, making it difficult to compare environmental impact of different printed materials or establish reliable benchmarks for industry [327, 328]. In many cases, LCAs do not explicitly quantify or report the exact material composition and mass deposited in each functional layer of printed devices, despite this information being critical for assessing resource efficiency, recyclability, and end-of-life scenarios. Providing quantitative indicators would allow to define global sustainable benchmarks to guide future material selection and accelerate the adoption of most efficient technologies.

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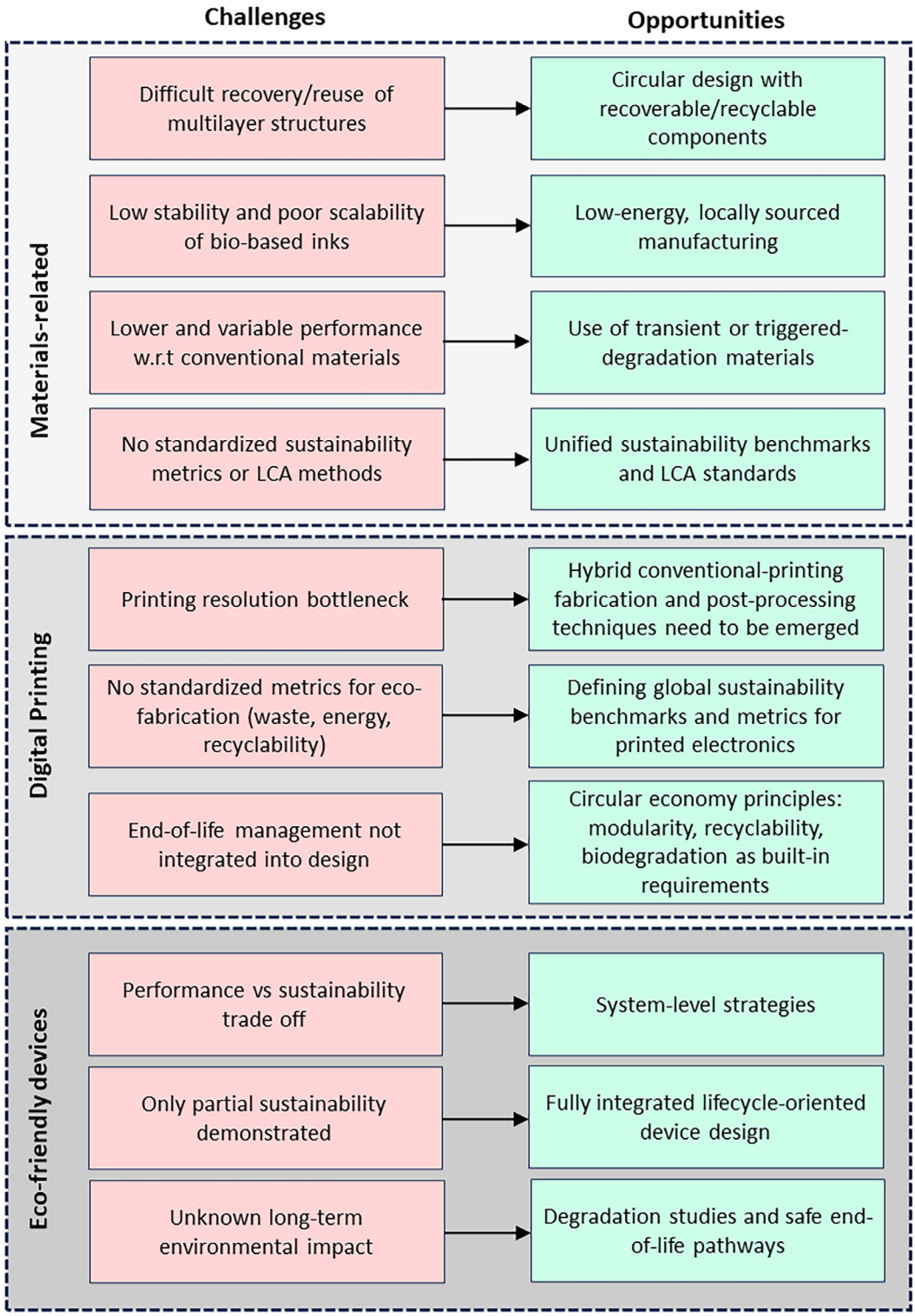


FIGURE 8 | Challenges and Opportunities in digitally printed eco-friendly electronics. The schematic showing the major challenges described in this section together with the potential opportunities in the future to overcome those challenges.

6.2 | Advancing the Printing Techniques

As discussed earlier, eco-friendly printed electronics face not only material-related performance bottlenecks but also significant fabrication challenges. Current digital printing techniques, while offering scalability and material efficiency, are still limited in terms of resolution, performance, uniformity, and throughput. To achieve similar performance as that with conventional fabrication, new printing strategies must evolve on multiple fronts.

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In this regard, multimodal digital printing offers a near-term pathway. Rather than relying solely on one type of printing, hybrid schemes combining multiple printing techniques or with laser patterning could deliver finer features and improved device yield. Such hybridization can help bridge the gap between sustainability and performance until fully green, higher-resolution printing methods are developed. Critically, hybrid digital printing strategies that enable precise control over processing temperature, energy input, and exposure duration are required. For instance, printing techniques that integrating low-temperature photonic curing, microwave processing, or plasma treatments into digital printing workflows offers promising pathways to enhance performance by improving crystallinity and interfacial quality without resorting to energy-intensive annealing or toxic solvents and maintaining eco-compatibility.

Along with the technical advancement, the standardization gap noted for materials (Section 6.1) extends to the fabrication stage as well. Beyond conventional performance indicators such as device efficiency or fabrication yield, printing processes must also be assessed in terms of material utilization and wastage, energy consumption, solvent toxicity, and water wastage/pollution. This perspective shifts sustainability from being an "add-on" to becoming a built-in requirement of the fabrication process itself.

6.3 | Device Performance versus Sustainability

Beyond the material-level performance gap discussed above, the main challenge facing eco-friendly printed devices is the trade-off between sustainability and device performance. While bio-derived materials and environmentally benign fabrication routes reduce environmental impact, they often impose constraints on charge transport, device stability, and operational lifetime. Similarly, sustainable processing conditions typically avoid the high temperatures, aggressive solvents, and energy-intensive treatments that are commonly used to maximize device performance in conventional electronics. Furthermore, the long-term and cumulative environmental impacts of many so-called eco-friendly devices remain insufficiently understood.

These performance constraints extend beyond transistors. In eco-friendly displays, illumination efficiency, refreshing speed, and device lifetime are still far inferior compared to commercial OLED and QLED technologies. In printed sensors, limited sensitivity, poor reproducibility, and long-term instability restrict their use in high-density integration. Printed photovoltaics, though promising in concept, remain heavily reliant on toxic elements and heavy metals for efficiency gains. Mechanical energy harvesters such as PENG and TENG offer theoretical potential but cannot yet meet realistic energy demands. There is limited research on fully eco-friendly printed battery, and the current supercapacitors lack sufficient power density for practical applications. Furthermore, encapsulation remains a critical hurdle, as biodegradable barrier layers generally fail to provide adequate protection against oxygen and moisture, severely limiting device lifetime.

Additionally, it must be emphasized that the majority of reported devices in the literature do not represent fully sustainable end to end solutions, but rather demonstrate only isolated or partial aspects of sustainability. For example, some studies employ bio-derived or biodegradable materials but rely on toxic or nongreen solvents during fabrication; others use environmentally benign inks but are fabricated on nondegradable plastic substrates; and in several cases, devices incorporate inorganic or scarce materials that are not biodegradable. In certain works, sustainability claims are instead linked to recyclability or material recovery, rather than intrinsic environmental benignity. Thus, current eco-friendly printed electronics should be interpreted as incremental progress addressing specific sustainability metrics, rather than complete end-to-end sustainable systems. Furthermore, the long-term and cumulative environmental impacts of many proposed materials, including those considered edible or low toxicity, remain largely unknown.

Bridging the gap between performance and sustainability requires a prioritized research roadmap that addresses material, fabrication, and circularity challenges in a sequential manner. In the short term, the primary focus should be the development of scalable green inks and bio-derived functional materials with improved electrical performance, storage stability, and compatibility with industrial printing processes. In parallel, standardized protocols for material characterization, sustainability assessment, and life-cycle analysis should be established to enable meaningful benchmarking across studies. In the medium term, efforts should concentrate on advancing high-resolution, low-energy printing technologies and post-processing approaches, including photonic curing, plasma treatments, and bio-compatible sintering strategies, to improve device performance without compromising sustainability. Finally, long-term research should focus on circular manufacturing pathways, including recyclable device architectures, design-for-disassembly principles, material recovery strategies, and safe end-of-life degradation mechanisms.

Ultimately, the eco-friendly printed electronics should progress through a coordinated roadmap centered on (i) scalable green inks and bio-derived materials, (ii) high-performance and resource-efficient manufacturing technologies, (iii) circular device design and end-of-life management, and (iv) standardized sustainability assessment frameworks. Addressing these priorities in a structured manner will accelerate the transition of eco-friendly printed electronics from proof-of-concept demonstrations toward industrially relevant and environmentally responsible technologies.

7 | Conclusions

This review provides a comprehensive overview of recent progress in eco-friendly electronics enabled by digital printing, addressing sustainability challenges throughout the entire device lifecycle. We first examined major digital printing techniques, such as inkjet, electrohydrodynamic-jet, aerosol-jet, and extrusion-based printing, highlighting their inherent sustainability advantages, technical limitations, and suitability for environmentally responsible fabrication. Eco-friendly material systems and ink formulations were then reviewed, encompassing green solvents, sustainable conductive, semiconductive, and dielectric materials, as well as environmentally benign additives. Building on these foundations, the review surveyed a broad

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range of applications enabled by digitally printed eco-friendly materials, including active and passive electronic components, sensors, actuators, energy harvesting, and storage devices. End-of-life management strategies, including device repair and reuse, material recycling, and biodegradability, were further discussed as essential elements for reducing the long-term environmental impact of electronic systems.

Despite the rapid progress outlined in this review, several critical challenges remain that must be addressed to enable the widespread adoption of eco-friendly digitally printed electronics. A fundamental obstacle is the persistent trade-off between sustainability and device performance. Limitations in printing resolution, film uniformity, and multilayer integration further restrict device scalability and long-term reliability. In addition, the recovery and reuse of complex multimaterial printed architectures remain challenging, underscoring the need for design-for-recycling strategies and improved interfacial engineering. Looking ahead, future research must focus on the discovery of high-performance sustainable materials, the development of hybrid fabrication approaches that integrate printing with complementary patterning and post-processing techniques, and the establishment of standardized sustainability metrics and life-cycle assessment frameworks. Strategies such as triggered degradation, recyclable material systems, and low-energy post-processing offer promising pathways to reconcile performance with environmental responsibility. Ultimately, advancing eco-friendly printed electronics will require coordinated progress across materials science, manufacturing technology, and system-level design to realize truly sustainable electronic platforms.

Author Contributions

Nitheesh M. Nair: conceptualization, writing – original draft, methodology, investigation. **Mikel Rincón-iglesias**: writing – original draft, conceptualization, methodology, investigation. **Cristian Mendes Felipe**: conceptualization, methodology, investigation, writing – original draft. **Fatemeh Etehadi**: writing – original draft, conceptualization, methodology, investigation. **Senentxu Lanceros-méndez**: writing – review and editing, supervision, funding acquisition. **Gerardo Hernandez-sosa**: funding acquisition, writing – review and editing, supervision, conceptualization, writing – original draft, methodology, investigation.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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