

Review

# Polyaniline-Based Ink for Inkjet Printing for Supercapacitors, Sensors, and Electrochromic Devices

Ekta Kundra Arora <sup>1</sup>, Vibha Sharma <sup>1</sup> , Aravind Ravi <sup>1</sup>, Akanksha Shahi <sup>1</sup>, Shweta Jagtap <sup>2</sup>, Arindam Adhikari <sup>3</sup>, Jatis Kumar Dash <sup>4,\*</sup> , Pawan Kumar <sup>5,\*</sup>  and Rajkumar Patel <sup>6,\*</sup> 

- <sup>1</sup> Chemistry Department, St. Stephen's College, University of Delhi, Delhi 110007, India; ekta.kundra@ststephens.edu (E.K.A.); vibha.sharma@ststephens.edu (V.S.); aravindravi037@gmail.com (A.R.); akanksha.shahi0211@gmail.com (A.S.)
- <sup>2</sup> Department of Electronic and Instrumentation Science, Savitribai Phule Pune University, Pune 411007, India; shweta.jagtap@gmail.com
- <sup>3</sup> Aadarsh Innovations, Balewadi, Pune 411045, India; arindam.adhikari@gmail.com
- <sup>4</sup> Department of Physics, SRM University-AP, Amaravati 522502, India
- <sup>5</sup> Department of Chemistry and Biochemistry, University of Windsor, 401 Sunset Ave, Windsor, ON N9B 3P4, Canada
- <sup>6</sup> Energy and Environmental Science and Engineering (E2SE), Integrated Science and Engineering Division, Underwood International College, Yonsei University, Incheon 21983, Republic of Korea
- \* Correspondence: author: jatis.d@srmmap.edu.in (J.K.D.); pawankumar.nst@gmail.com (P.K.); rajkumar@yonsei.ac.kr (R.P.)

**Abstract:** In recent years, there has been a huge surge in interest in improving the efficiency of smart electronic and optoelectronic devices via the development of novel materials and printing technologies. Inkjet printing, known to deposit 'ink on demand', helps to reduce the consumption of materials. Printing inks on various substrates like paper, glass, and fabric is possible, generating flexible devices that include supercapacitors, sensors, and electrochromic devices. Newer inks being tested and used include formulations of carbon nanoparticles, photochromic dyes, conducting polymers, etc. Among the conducting polymers, PANI has been well researched. It can be synthesized and doped easily and allows for the easy formation of composite conductive inks. Doping and the addition of additives like metal salts, oxidants, and halide ions tune its electrical properties. PANI has a large specific capacitance and has been researched for its applications in supercapacitors. It has been used as a sensor for pH and humidity as well as a biosensor for sweat, blood, etc. The response is generated by a change in its electrical conductivity. This review paper presents an overview of the investigations on the formulation of the inks based on conductive polymers, mainly centered around PANI, and inkjet printing of its formulations for a variety of devices, including supercapacitors, sensors, electrochromic devices, and patterning on flexible substrates. It covers their performance characteristics and also presents a future perspective on inkjet printing technology for advanced electronic, optoelectronic, and other conductive-polymer-based devices. We believe this review provides a new direction for next-generation conductive-polymer-based devices for various applications.

**Keywords:** conducting polymers; inkjet printing; polyaniline; PEDOT; PSS; supercapacitors; sensors; electrochromic devices; patterning of conductive polymers



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## 1. Introduction

Advances in materials science, material engineering, and printing technologies for smart electronic, optoelectronic, and other devices are paving the way for the development of extremely efficient devices that are improving quality of life rapidly [1–11]. Materials and processes that improve the quality and enhance the performance of the devices while reducing the cost are most desirable. Inkjet printing, one such technology, will continue to expand, driven by the demand for smart devices, lightweight flexible materials, and environmentally

sustainable technologies. Over the past years, this technology has advanced from developing simple printable electronic elements like pressure and temperature sensors to state-of-the-art applications including medical diagnostics, analytical applications, and the development of optoelectronic systems. Inks used for printing have a wide array of properties ranging from electrically conducting and semi-conducting properties to thermal conductivity, electroluminescence, etc. Functionality combined with easy processability are necessary to fabricate tailor-made devices. Novel functional inks include but are not limited to carbon nanoparticles [12], redox-responsive organometallic polymers like poly(ferrocenylsilane) (PFS) [13], photochromic dyes based on organic spiro compounds [14], polysiloxane-based water-repellent microemulsions [15], conductive polymer polyaniline (PANI) and its composite-based inks [16], *p*-doped poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) [17,18], and *n*-type poly(benzimidazobenzophenanthroline):poly(ethyleneimine) (BBL:PEI) electrically conductive polymers [19].

Inkjet printing offers rapid, reproducible, and low-cost ways to pattern conductive polymers on substrates and is an excellent tool for printing conductive-polymer-based completely functional products, which include supercapacitors, sensors for different compounds, and energy-storing electrochromic devices on different substrates. Its advantages over other enabling technologies include scalability (small as well as large surface areas), high precision, simultaneous deposition of multicomponent layers [20,21], printing of delicate membranes without fear of contamination (non-contact printing where the substrate and the print head never come in contact) [22], and printing from aqueous solutions, making it well suited for biological species [23]. An inkjet printer has an ink chamber to store ink and a channel that connects this chamber to the nozzle. Two types of inkjet printers based on thermal and piezoelectric effects dominate drop-on-demand technologies. Piezoelectric print heads are advantageous, as the risk of thermal degradation of the ink is minimized [24].

Since their accidental discovery by H. Shirakawa et al., conjugated conductive polymers have been well researched due to their distinctive electronic properties [25,26]. These conductive polymers with C=C conjugated double bonds forming the backbone display interesting electrical properties, which include low-energy optical transitions, low ionization potentials, and high electron affinities. These polymers can be oxidized and reduced, thereby converting them into *p*- and *n*-doped materials with increased conductivity [27]. Among the conjugated conductive polymers, polyaniline, poly(phenylenevinylene), polypyrrole, and PSS have attracted much interest [28]. Conductive polymers fall in the class of smart polymer materials with dynamic electroactive structures and have been explored for use in electrochromic devices, electrophotocatalytic materials, sensors for pH and humidity, smart clothes and coatings, soft matter applications, and solar cells, to name a few [29–33]. Electrically conductive membranes are being studied as alternatives to conventional membranes with enhanced performance for water filtration and wastewater treatment [34]. Conductive-polymer-based thermoelectric devices exhibit flexibility, are inexpensive, and are potential candidates to convert waste heat to power [35]. Conductive polymers are also being researched as electromagnetic radiation shields. Electromagnetic radiation is not only a health hazard but also interferes with the functioning of electronic equipment. Nanohybrids prepared by graft polymerization of aniline on graphene oxide substrate [36] and 3D epoxy nanocomposites based on 3D P-PANI@f-Cu show promise as electromagnetic interference (EMI) shielders [37]. Similarly, epoxy-graphene hybrid nanocomposites show an increase in EMI shielding as the amount of PANI increases [38], while polyaniline and vanadium pentoxide nanocomposites not only exhibit good electromagnetic interference shielding but are also great corrosion inhibitors [39]. Various nanomaterials in the PVA-PEDOT matrix have microwave-absorbing properties [40], and the polyamide 6/carbon fiber (PA6/CF) composite also exhibits good EMI shielding properties [41]. An interesting application of conductive polymers is their use in the preparation of Janus fibers, which are multifunctional. Janus fibers with PANI as a component have been proposed to be useful for drug delivery and electromagnetic shielding [42].

Microfabrication, which is now necessary for several applications like microelectronics [43], microanalytical systems [44,45], micro-optics, potable biomedical devices [46,47], and microfluidics [48,49], is facilitated by inkjet printing. As discussed above, due to the excellent electronic and processability features of conductive polymers, growing efforts are devoted to ink formulation and fabrication of devices with conductive polymers via inkjet printing technologies.

One of the most useful conductive polymers is PANI. Electrochemical polymerization, chemical polymerization, vapor-phase polymerization (VPP), photochemically initiated polymerization, enzyme-catalyzed polymerization, and polymerization employing electron acceptors are used for synthesizing PANI. The electrical conductivity of PANI can be modified easily [50]. Three structures are well established for PANI based on the oxidation states. They include the leucoemeraldine base (LEB) form, which is the completely reduced state and appears white in color; the emeraldine base (EB) form, which is the semi-oxidized state and appears blue in color; the salt form appears green; and the pernigraniline base (PAB), the totally oxidized form, appears blue to violet in color [50,51]. The emeraldine state can be converted to the conductive polymer form by either acid or oxidative doping, as shown in Figure 1.

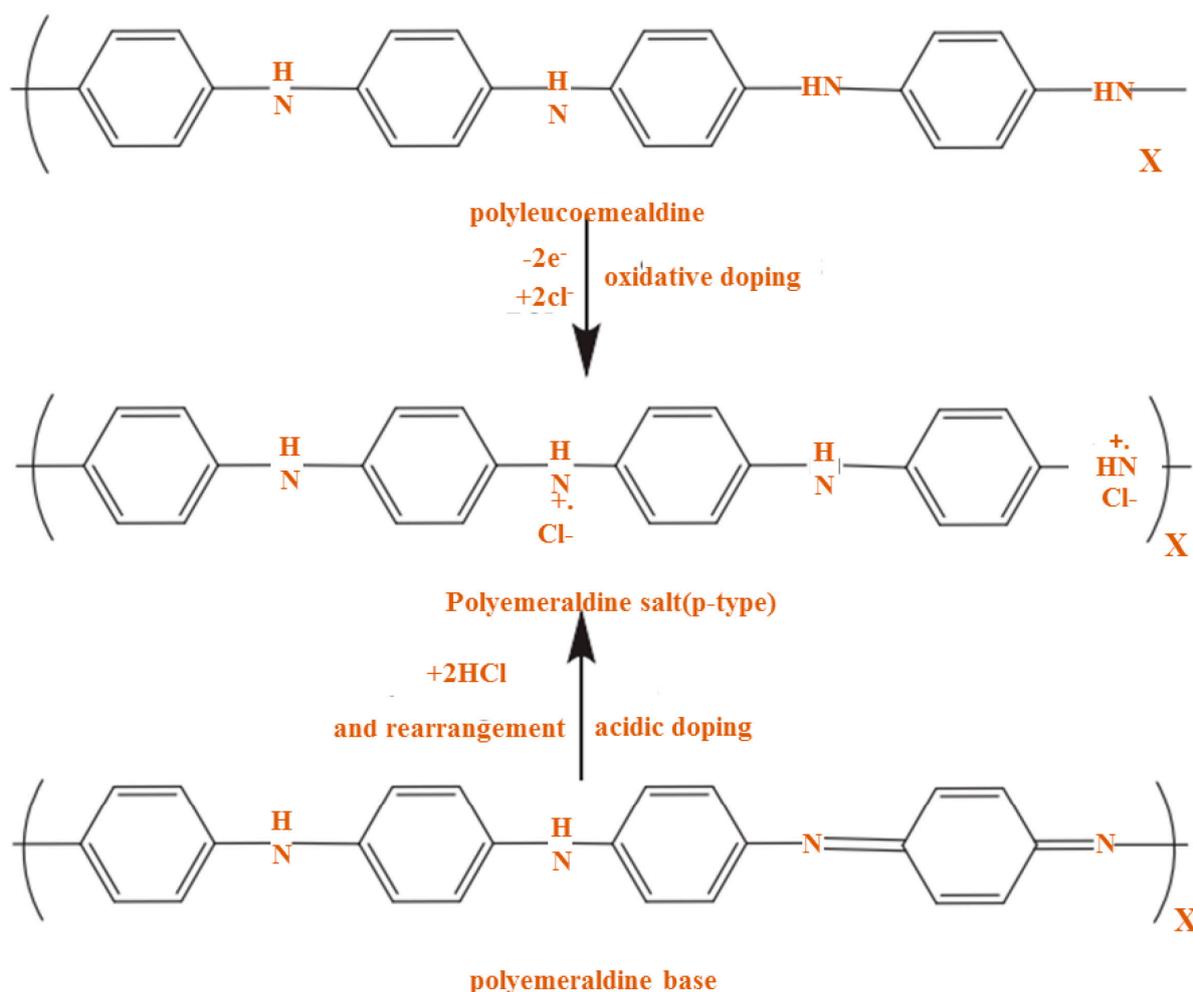


Figure 1. Oxidative and acid doping of PANI [51].

In this review, we focus on the formulation of the conductive polymer PANI-based inks, their inkjet printing onto substrates, and the fabrication of devices, with special reference to tailor-made supercapacitors, sensors, electrochromic devices, and the patterning of conductive polymers on flexible substrates. Many of the devices fabricated until recently were rigid with a fixed architecture, but inkjet printing, falling under additive manufac-

turing technologies, allows deposition on flexible substrates. For proper deposition of the conductive material, the inks should be non-viscous with a viscosity lower than 100 mPa. The limited solubility of some conducting polymers in solvent- or water-based formulations may pose a hindrance [24]. Sequential deposition and centrifugation techniques allow for control of viscosity.

## 2. Applications of Inkjet Printer Conducting Polymers

Conductive polymers are explored for various applications, including supercapacitors, sensors, and electrochromic devices, owing to their excellent mechanical and electrical properties, flexibility, and low cost of production [33,52–55]. DuPont, Sigma-Aldrich, and Pedotinks are some of the leading manufacturers of conductive-polymer-based inks. Companies like Xaar, FUJIFILM Dimatix, and Global Inkjet Systems Ltd. provide functional material printing for various applications. This section reviews conducting polymers for supercapacitors, sensors, and electrochromic devices, including the formulation of the ink, the inkjet printing process, and the performance highlights. In addition, the patterning of conductive polymers on flexible substrates is reviewed in this section.

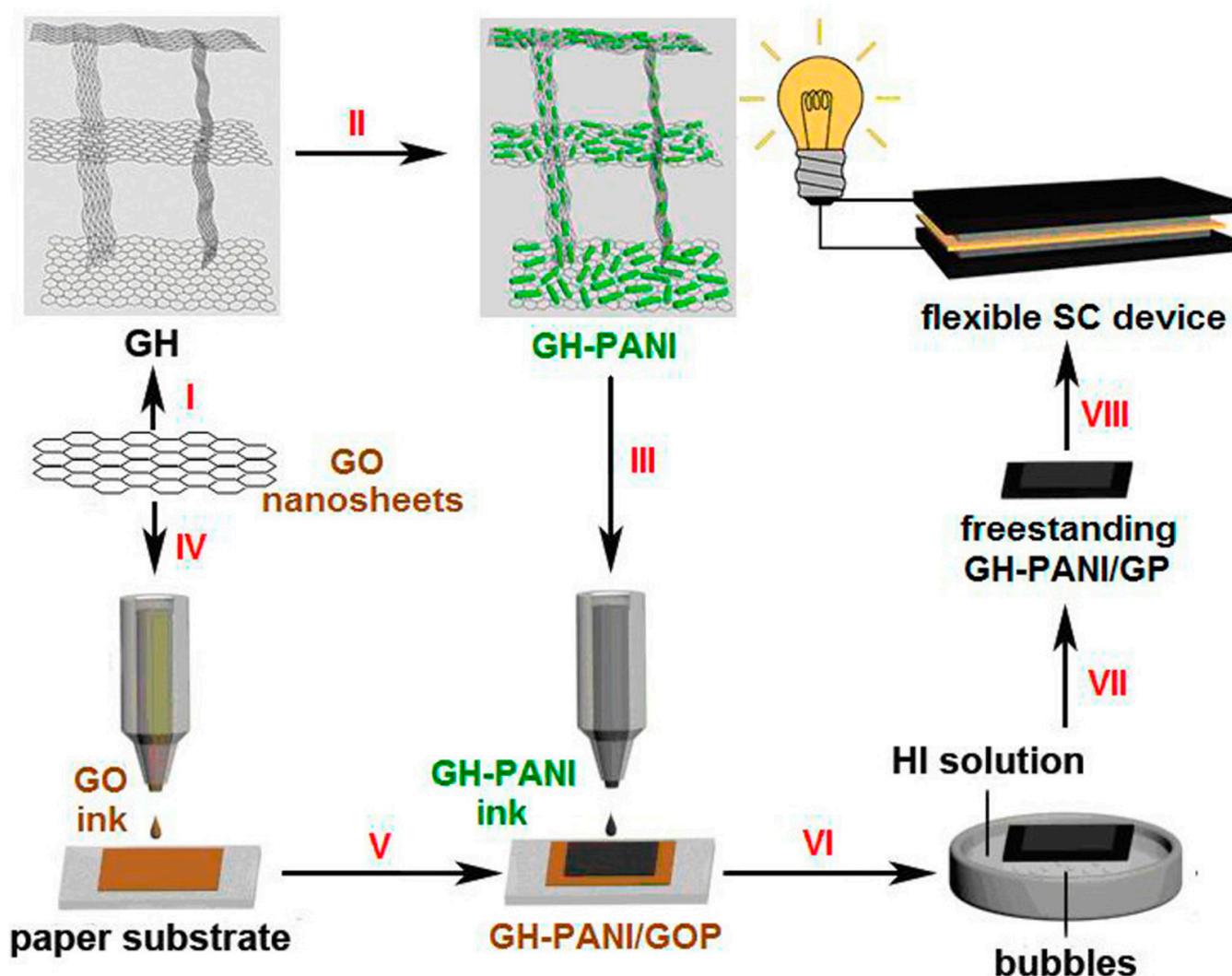
### 2.1. Conductive Polymer for Supercapacitors

An excellent choice as a sustainable device for energy storage, the supercapacitor has two electrodes, an electrolyte, and a separator, with the electrochemical material being of utmost importance. Supercapacitors are classified as electrochemical double-layer capacitors (EDLCs), pseudocapacitors, or hybrids formed by a combination of both types [56]. The EDLCs based on large-surface-area carbon materials like carbon nanotubes, activated carbon, etc., are non-Faradaic in nature, storing electrical energy via nanoscale charge separation at the electrode–electrolyte interface and forming electrical double layers. The pseudocapacitors based on metal oxide, metal-doped carbon, and conductive polymers are Faradaic in nature, based on a reversible and quick surface redox reaction between the electrode and electrolyte. They provide a much higher energy density in comparison to EDLCs. Polyaniline (PANI), polypyrrole (PPy), polythiophene (PTh), and poly 3,4-ethylenedioxythiophene (PEDOT) are being extensively researched for pseudocapacitive applications because of their large specific capacitance.

Inkjet printing, known to deposit ink on demand, helps greatly economize the consumption of material while also allowing printing of the material on various substrates like paper, glass, and fabric of different designs and patterns, helping make flexible supercapacitors. Thin paper-like electrode materials as well as free-standing electrode materials, which have the advantage of being free of insulating binders, conductive agents, current collectors, and solvents, are best suited for flexible and lightweight devices of the future. The development of a variety of methods by which polyaniline (PANI) can be synthesized has led to it being extensively researched for inkjet-printed supercapacitors. A nanohybrid freestanding electrode based on GP (graphene paper) and GH-PANI (graphene hydrogel-PANI)  $\pi$ – $\pi$  stacking interactions has been developed via inkjet printing. The steps involved in the fabrication of GP (graphene paper)- and GH-PANI (graphene hydrogel-PANI)-based supercapacitors are illustrated in Figure 2. The asymmetric supercapacitor device demonstrated good mechanical, electrochemical, and capacitive properties, with a tolerable energy density of 24.02 Wh kg<sup>−1</sup> at a power density of 400.33 W kg<sup>−1</sup> [57].

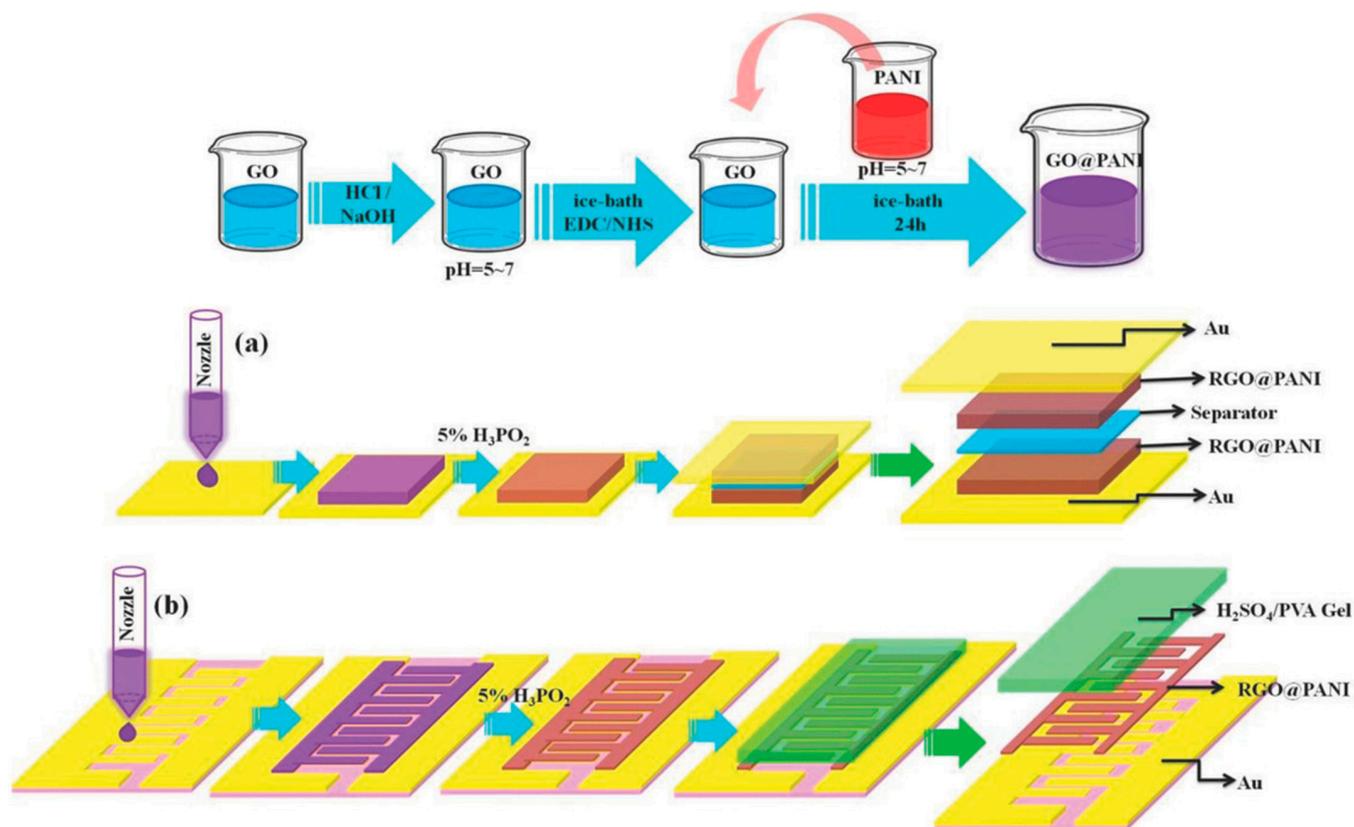
PANI production has traditionally been achieved by polymerizing the monomer aniline. A benign method where the aniline dimer (DANI) in an organic solvent is added to the oxidative solution of ammonium persulfate in an aqueous medium has been used to prepare the emeraldine PANI (PANI-EB) as a fast reaction, eliminating hazardous and toxic aniline monomer usage directly. Inks with extremely good jettability and geometric realization have been obtained by dedoping this salt doped with chloride ions to obtain PANI doped with trifluorosulphonic acid and camporsulphonic acid as counterions, thereby increasing the conductivity of the resultant inks for producing inkjet-printed tracks of diverse geometry [58]. These inks are also of interest as they demonstrate

that counterion displacement induced by the measurement signal leads to negative resistance/capacitance and supercapacitance ( $-2.3 \text{ mF @ } 30 \text{ Hz}$ , which corresponds to a mass capacity of  $-799 \text{ F g}^{-1}$ ), ideal for application in low-frequency-range devices. An ink based on PANI nanoparticles covalently attached to the graphene oxide, i.e., (GO) @ PANI composites, was synthesized by first synthesizing GO and PANI separately, followed by mixing EDC/NHS (1-(3-dimethylaminopropyl)-3-ethylcarbodiimide hydrochloride (EDC) and N-hydroxysulfosuccinimide (NHS)) activated GO and PANI in the ratio 9:1 to prepare the composite, which was inkjet printed on a gold-coated polymer substrate (PET) and reduced by hypophosphorous acid to fabricate the desired electrodes [59]. Sandwiched and interdigitated supercapacitors were assembled (Figure 3). Volumetric supercapacitance for both the sandwiched ( $258.5 \text{ F cm}^{-3}$  at  $1 \text{ mV s}^{-1}$ ) and interdigitated supercapacitors ( $554 \text{ F cm}^{-3}$  at  $1 \text{ mV s}^{-1}$ ) was high, with an excellent cycling retention of over 90% after 2000 cycles of charging and discharging.

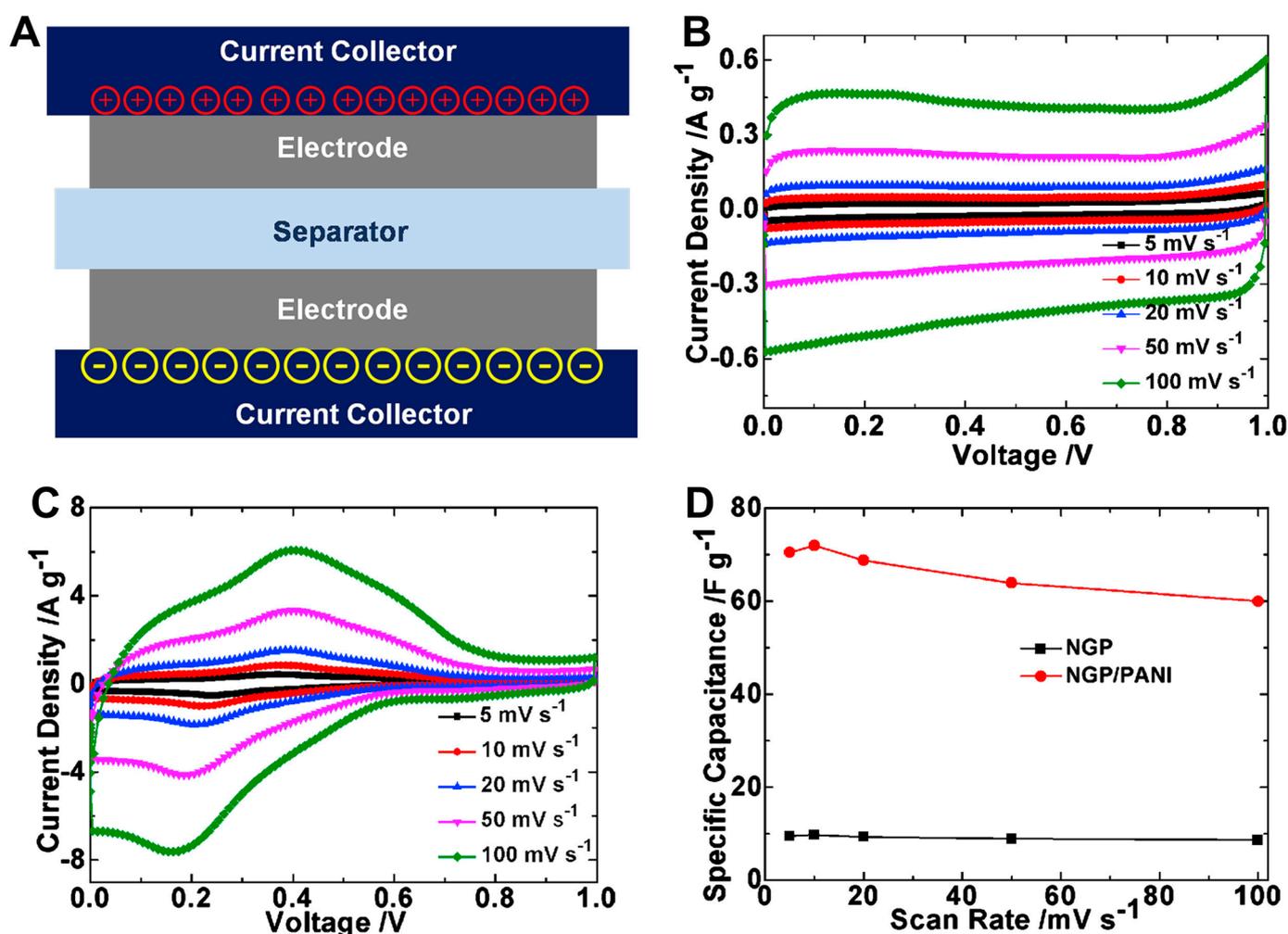


**Figure 2.** (Steps I–V): The fabrication of the GH-PANI supercapacitor involves graphene oxide (GO) self-assembly into 3D graphene hydrogels (3D GHs), followed by in situ polymerization of PANI on GH. The GH-PANI ink is obtained via ball milling and ultrasonic treatment of the 3D GH-PANI composite formed in the previous step. The ink is printed on the paper substrate, giving GOP, followed by overprinting as represented in step VI. Soaking the fabricated GH-PANI/GOP in the HI solution results in reduction by HI, and removal by peeling from the substrate results in freestanding GH-PANI/GP. Fabrication the occurs of a flexible SC device based on the GH-PANI/GP electrode and gel electrolyte. Reproduced with permission from [57].

Xu and co-workers formulated nanographene/polyaniline (NPG/PANI) inks of appropriate viscosity, surface tension, and conductivity. These inks can be deposited as thin films on substrates using inkjet printing (Figure 4) [60]. Two NGP/PANI ink-printed electrodes with carbon fabric as the substrate were sandwiched together using a separator, forming an electrochemical capacitor, and studied using a 1 M H<sub>2</sub>SO<sub>4</sub> electrolyte. They show a maximum specific capacitance of 82 F g<sup>-1</sup>, a power density of 124 kW kg<sup>-1</sup>, a cycle life of 1000 cycles, and an energy density of 2.4 Wh kg<sup>-1</sup> with a scan rate of 20 mVs<sup>-1</sup> (Figure 4). On macro-porous graphene foam (ultrathin graphene sheets), a thin film of PANI was deposited via electrochemical deposition (polyaniline-coated graphene). The pores were filled with aniline, phytic acid, and ammonium persulfate by inkjet printing to allow oxidative polymerization. This resulted in PANI-filled graphene foams. The PANI-filled macro-porous graphene used as an electrode for supercapacitors exhibited a high areal capacitance of over 1700 mF cm<sup>-2</sup>, more than two times the capacitance achievable with pure graphene, metal oxide, or conductive polymer thin-layer coated graphene [61]. Various conducting polymers, such as PANI, PPy, and PTh, have been widely used for the fabrication of supercapacitors via inkjet printing. Some reported supercapacitors based on conductive polymers are tabulated in Table 1. In the case of supercapacitors, improvement in mechanical robustness, structural complexity, convenient fabrication, and device configuration is still a critical task. For example, although conductive polymers have an almost 10-fold stretchability capacity, final devices do not have this ability due to the presence of other device components that are not stretchable.



**Figure 3.** (a) Sandwich-structured supercapacitors. (b) Interdigitated supercapacitors on flexible gold films. Reproduced with permission from [59].



**Figure 4.** Representative supercapacitor device structure is depicted in (A). (B,C) Comparisons of current voltage (CV) curves of nanographene (NGP) and NGP/PANI electrodes, measured at different scan rates, in 1 M  $\text{H}_2\text{SO}_4$  electrolyte for a two-electrode system. (D) Comparison of the specific capacitance vs. scan rate plots of nanographene (NGP) and NGP/PANI electrodes in two-electrode system, with 1 M  $\text{H}_2\text{SO}_4$  aqueous electrolyte. Specific capacitance as a function of scan rate in two-electrode system, with 1 M  $\text{H}_2\text{SO}_4$  aqueous electrolyte. Reproduced with permission from [60].

**Table 1.** Conductive-polymer supercapacitor inks for inkjet printing.

Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	Reduction of GH-PANI/GOP by hydroiodic acid and simultaneously peeling off from the commercial paper substrate to give the freestanding electrode (GH-PANI/GP electrode).	Fabrication of graphene-based nanohybrid materials for use in many electronic systems.	A maximum energy density of $24.02 \text{ Wh kg}^{-1}$ at a power density of $400.33 \text{ W kg}^{-1}$ and a power density of $3202.4 \text{ W kg}^{-1}$ at an energy density of $13.29 \text{ Wh kg}^{-1}$ are achievable at an operating voltage of 0.8 V.	[57]

Table 1. Cont.

Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	Two distinct doped PANI inks were prepared by dissolving the emeraldine salt of PANI in dimethylsulphoxide, followed by the addition of trifluorosulphonic acid and camporsulphonic acid.	Materials with negative capacitance in the low-frequency range can be used in devices working in nominal grid conditions (50–60 Hz) and up to short-wave radio frequencies.	Negative capacitance is reported. The highest negative supercapacitance achieved is $-2.3 \text{ mF}$ at 30 Hz, corresponding to a specific mass capacity of $-799 \text{ F g}^{-1}$ .	[58]
Polyaniline	Electrodes were fabricated by printing GO @ PANI composites on gold-coated polymer substrates and then reducing them. Sandwiched and interdigitated supercapacitors were developed.	This method allows the end users to precisely deposit active materials according to their designs for miniature and wearable electronics.	Devices fabricated have high volumetric capacities of $258.5 \text{ F cm}^{-3}$ at $1 \text{ mV s}^{-1}$ for sandwich structures and $554 \text{ F cm}^{-3}$ at $1 \text{ mV s}^{-1}$ for interdigitated ones. Even after 2000 cycles of charging and discharging, over 90% capacitance retention could be achieved.	[59]
Polyaniline	Graphene polyaniline (NGP/PANI) inks of appropriate surface tension and viscosity were formulated and then inkjet printed to produce thin-film supercapacitor electrodes.	This preparation method allowed good control over pattern geometry and location in thin films. A major application is manufacturing energy storage devices in printable electronics.	In $1 \text{ M H}_2\text{SO}_4$ solution as the electrolyte, a maximum specific capacitance of $82 \text{ F g}^{-1}$ , a power density of $124 \text{ kW kg}^{-1}$ , and an energy density of $2.4 \text{ Wh kg}^{-1}$ were observed when a scan rate of $20 \text{ mV s}^{-1}$ was applied. A long life cycle of over 1000 cycles.	[60]
Polyaniline	Graphene foam is made up of a few layers of graphene electrodeposited with a thin layer of PANI, with subsequent filling of the submillimeter-sized pores with PANI by using inkjet printing.	Good option for high-performance supercapacitors.	The synergistic effect of graphene and PANI provides a large areal capacitance of over $1700 \text{ mF cm}^{-2}$ .	[61]

## 2.2. Conductive Polymers for Sensors

Nanostructured conductive polymers are good candidates for sensor and biosensor design. A response is generated when the electrical conductivity is modified via redox reactions or protonation. The polymers can be deposited on various surfaces with good thermal and chemical stability at a low cost, making them ideal for many applications. In addition, the conductivity can be tuned by doping or combining the conductive polymer with various additives, including oxidants like ammonium persulfate and ammonium peroxy disulfate, aqueous copper (II) and iron (III) salts, and halogen electron acceptors like bromine and iodine. Inkjet printing a solution or suspension containing the conductive polymer offers a simple and cost-effective method for preparing varied sensors for environmental sensing, food testing, and clinical applications. Among various sensors, pH sensors have received much attention in the food and healthcare industries, especially for foodborne pathogen detection and as personal wearable biochemical sensors to monitor sweat, blood, and tumors as a complement to lab-based equipment.

A paper-based colorimetric sensor using Polyaniline-Pectin nanoparticles (PANI-PEC NPs) printed on Whatman filter paper grade 4 has been used for sensing the presence of *E. coli*, both qualitatively and quantitatively in milk and milk products. The acid metabolites produced by the bacterium (succinates, lactates, acetates, malates, etc.) convert the blue emeraldine base form of PANI to a green-colored emeraldine salt form with increased conductivity. With a sensitivity of  $0.52 \pm 0.17 \log \text{CFU/mL}$  *E. coli* within 10: 21 h, it represents a practical system with good applicability and efficiency [62].

Aqueous-based PANI ink formulations with acrylic resin additives as binders were used to print pH-sensitive electrodes by Bilbao and coworkers. The working electrode is a screen-printed carbon ink layer on Valox<sup>®</sup> (polybutylene terephthalate), modified by inkjet-printed PANI ink. Its performance was compared with the electrodes prepared by electropolymerization of PANI on carbon paste electrodes and found to be better. The response of these electrodes fabricated by inkjet printing PANI inks and acrylic resins onto screen-printed carbon electrodes for synthetic sweat showed good sensitivity of up to 69.1 mV/pH [63].

PANI has long been used as a sensor for toxic ammonia gas. It deprotonates the emeraldine salt and converts it to the emeraldine base form, with a corresponding drop in conductivity. Toxic ammonia gas can be sensed by piezoelectric inkjet printing of polyaniline nanoparticle films on interdigitized silver electrode arrays (Ag IDAs) on a PET substrate. The sensor is further attached to polyimide-based flexible heating foils to facilitate operation at different temperatures. It has a stable logarithmic response of 1–100 ppm to ammonia with a  $t_{50}$  value of 15 s at room temperature, with better performance at 80 °C [64]. The response was also found to be largely independent of the relative humidity range of 35 to 98%.

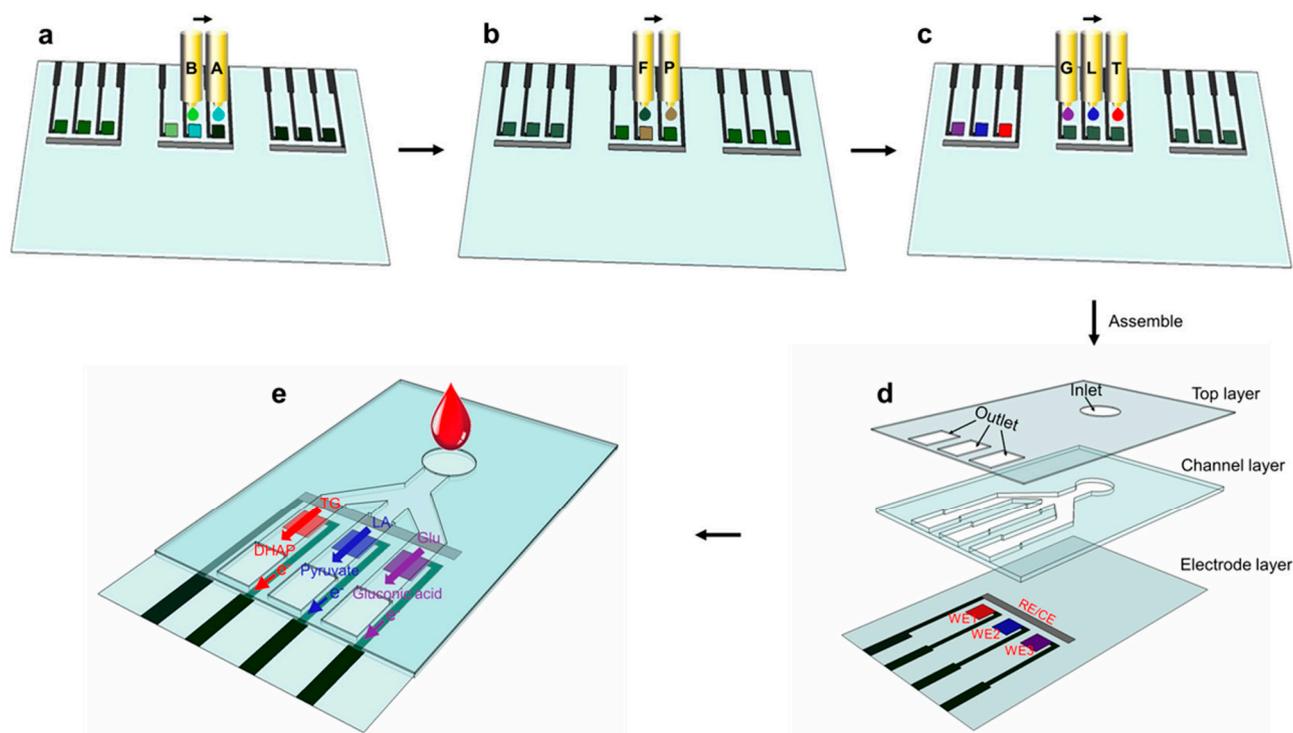
Sensors for gaseous ammonia developed by Duy Dam Le and co-workers gave promising results. Silver inkjet printed on Si/SiO<sub>2</sub> substrates was drop-coated with a blend film of the emeraldine salt form of PANI and ethylene glycol (EG) to sense NH<sub>3</sub>. It showed good sensitivity as well as a linear response to ammonia concentration at a relative humidity of 3–70% [65]. The sensor could be regenerated for use by heat treatment at 60 °C in about 15 min. Inkjet-printed layers (Dimatix material printer) of polyaniline and copper (II) chloride on a screen-printed silver interdigitated electrode (IDE) on a flexible PET substrate function as a chemiresistor for hydrogen sulfide with a sensitivity of 2.5 ppm by volume [66]. A linear relationship between measured current and concentration is observed in the 10–100 ppmv region. The variation in current in the circuit is due to the protonation of PANI to the emeraldine salt by H<sub>2</sub>S, as opposed to the protonation of ammonia by PANI in ammonia sensors. A class of CuCl<sub>2</sub>/PANI chemiresistor sensors with inkjet-printed silver electrodes on kaolin-coated paper substrates has also been studied for sensing H<sub>2</sub>S. Exposure to H<sub>2</sub>S resulted in the formation of copper sulfide and the acid HCl, which protonates the EB PANI, resulting in an increase in conductivity. Concentrations as low as 10 ppm H<sub>2</sub>S could be sensed [67].

Inkjet printing provides a low-consumption, cost-effective method of preparing disposable diagnostic tools for medical applications. A disposable paper-based electrochemical sensor for the detection of ascorbic acid has a good sensitivity of 17.7 μA/mM, with the limit of detection for ascorbic acid being  $30 \pm 3 \mu\text{M}$  in a concentration range of 30–270 μM. The assembly consists of screen-printed conductive carbon graphite paste on Whatman Grade No. 1 filter paper as the reference electrode and counter electrode, and five-layer PANI inkjet printed onto the screen-printed carbon electrodes as the working electrode in acetate buffer at a pH of 5 [68].

The conducting emeraldine salt phase of PANI ink prepared by ammonium persulfate oxidation used for the printing of interdigitated patterns on the flexible untreated polymer substrate using an HP inkjet printer exhibited a change in resistance with a change in relative humidity. The sensor exhibited a short response time of 5 s under low humidity conditions (6% RH); on exposure for 50 s, the electrical resistance exhibited an apparent increase; and when the sensor was exposed to high humidity conditions (97.3% RH), the

resistance started decreasing [69]. The sensor has applications as low-cost RFID tags, polymer photovoltaic cells, and printed flexible electronics devices.

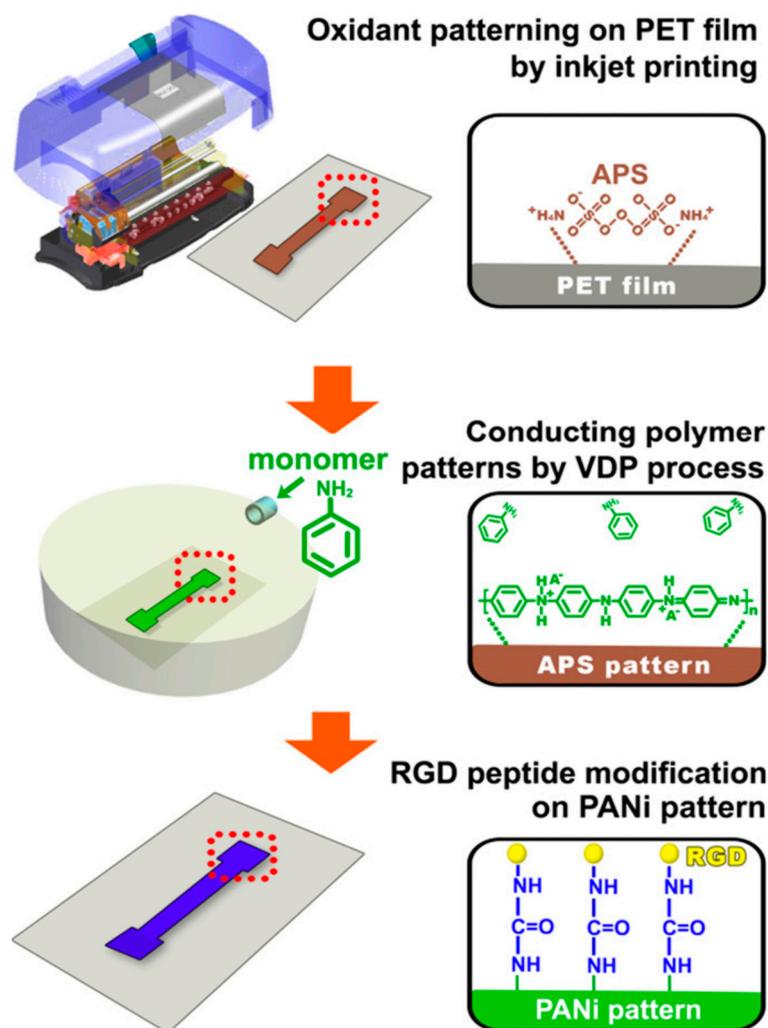
Multiplexed detection of various metabolites is useful for early and easy detection of diseases. It is a challenge to deposit electroactive materials and enzymes on selective electrodes in a precise fashion. By using a ‘drop on demand’ selective inkjet printing process, an assembly of a working electrode layer specific to each metabolite, a microfluidic channel layer, and a top cover has been fabricated (Figure 5). It has been tested for analyzing biomolecules like triglycerides, glucose, and lactates in phosphate-buffered saline as well as human serum samples [70]. The sensitivity with respect to triglycerides was found to be  $7.49 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$  between 0.1 mM and 6 mM, while that of lactate was  $3.94 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$  between 0.08 mM and 5 mM. The glucose sensitivity exhibited by the sensor was  $5.03 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$  between 1 mM and 25 mM.



**Figure 5.** Inkjet-printed multiplexed biosensor based on PANI conductive hydrogels: (a) Precursor solutions A and B were printed on the predefined areas to form a PANI hydrogel on the working electrode (WE). (b) Subsequent printing of chloroplatinic acid (P) and formic acid (F) solutions generated platinum nanoparticles (PtNPs) on the PANI hydrogel film. (c) This was followed by sequential printing of the enzyme solutions of glucose oxidase, lactic oxidase, and lipase/glycerol kinase/L- $\alpha$ -glycerophosphate oxidase on separate electrodes. (d) The biosensor has three layers: the top layer, the microfluidic channel layer in the middle, and the electrode layer at the bottom. (e) Detection of multiple metabolites in human blood. Reproduced with permission from [70].

Urea has long been known to function as an indicator of human kidney health. With the aim of developing a cheap disposable biosensor for urea, inkjet-printed assemblies have been tried. Carbon paste electrodes were screen printed onto pre-shrunk PET plates. This was followed by sequential inkjet printing of PANI nanoparticles, urease enzyme solution containing glycerol (0.1%, *v/v*), and then Triton X-100 (0.01%, *v/v*) in a phosphate buffer of pH 7.12 (0.1 M) on the working electrodes. The decomposition of urea was measured chronocoulometrically via the doping of ammonium at the polyaniline-modified, i.e., urease/nanoPANI biosensor electrode surface at  $-0.3 \text{ V}$  vs. Ag/AgCl [71]. The sensor successfully measured ammonia in the 0.1–100 mM range and urea in the 2–12 mM range ( $r^2 = 0.98$ ). Inkjet printing was used to fabricate a three-electrode configuration on a coated paper substrate. An assembly of inkjet-printed gold nanoparticle-based working

and counter electrodes, along with a quasi-reference inkjet-printed silver nanoparticle electrode with an electrochemically deposited layer of Ag/AgCl, was devised and studied as sensors after various modifications. The formation of self-assembled monolayers (SAMs) of alkanethiols on the gold electrode surface was studied as a possible sensor for materials where terminally substituted thio groups are used. Electropolymerization of the working electrode with polyaniline (PANI) conductive polymer for pH-sensing applications, and glucose oxidase (Gox)-entrapped poly-3,4-ethylenedioxythiophene (PEDOT) films (PEDOT-Gox) with the possibility of forming amperometric glucose sensors, were also explored [72]. Morrin and coworkers investigated PANI nanoparticles in an enzyme biosensing application. Aqueous nanoparticles of PANI along with horseradish peroxidase (HRP) enzyme were cast simultaneously using a drop coating method onto the surface of the screen-printed carbon working electrode, the disadvantage being the inherent thickness of the film, which affected its potential [73]. Oxidant patterning by inkjet printing on polyethylene terephthalate films, followed by deposition of the emeraldine salt form of PANI patterns and immobilization of the RGD peptide over it via covalent linkages, were used for the detection of biomolecules from live cells (Figure 6). Rat pheochromocytoma PC12 cells were cultured on the RGD-immobilized PANI pattern, and real-time electrical signal detection to track biomolecular release was studied [74].



**Figure 6.** Patterning of the oxidant on the PET substrate via inkjet printing. Formation of PANI conductive polymer patterns via VDP process and subsequent modification of PANI patterns via RGD peptide. Reproduced with permission from [74].

To study inkjet printing for printable chemical sensors, low-sheet-resistance electrodes of multiwalled carbon nanotubes were inkjet printed on a transparent film. This was followed by the printing of randomly oriented PANI nanowires dispersed in an aqueous medium, giving a fully printed sensor assembly. The sensor was capable of detecting changes in pH as PANI changed the resistivity with pH; furthermore, a  $\text{H}_2\text{O}_2$  sensor was fabricated by depositing silver nanoparticles (which catalyze the decomposition of  $\text{H}_2\text{O}_2$  to produce  $\text{OH}^-$ ) over the PANI layer [75]. To explore the superior sensing of nanotubes and nanoparticles, carbon nanotubes as electrodes and chemiresistive polyaniline nanowire sensors were inkjet printed sequentially on the PET film to prepare a chemiresistive glucose sensor. The glucose oxidase and platinum nanoparticle catalysts were placed between the CNT layers. The glucose oxidase enzyme catalyzes glucose oxidation to produce gluconolactone and hydrogen peroxide;  $\text{H}_2\text{O}_2$  is further catalyzed by platinum nanoparticles to produce  $\text{OH}^-$ ; the local change in pH is sensed by the PANI layer, which changes resistivity [76]. A paper-based sensor was fabricated as a DNA biosensor and used for the determination of high-risk HPV type 16 by inkjet printing graphene polyaniline (G-PANI) conductive ink onto the screen-printed carbon ink working electrode. The AQ-PNA (anthraquinone-labeled pyrrolidiny peptide nucleic acid (acpcPNA)) probe was immobilized on this electrode and modified to introduce negative charges, which facilitated coulombic immobilization on the positively charged G-PANI electrode. It showed a linear response range of 10–200 nM, and the HPV type 16 DNA detection limit of 2.3 nM was reported. The sensor was successfully tested for its ability to detect the PCR-amplified DNA from HPV type 16 positive SiHa cells, successfully demonstrating that it functions as a highly sensitive ePAD DNA biosensor for the diagnostic screening and detection of cervical cancer [77].

Soft photomasks were printed on different substrates. Desired design patterns were inkjet printed onto a transparent hybrid composite inorganic oxide film (biaxially oriented polypropylene coated with silica oxide (BOPP-SiO<sub>x</sub>)) [78]. The inkjet-printed soft photomasks were further used for depositing organic polymers (PANI) and inorganic materials on polymer films and photografting organic polymer materials like PAA onto polymer films and non-planar substrates. Inkjet printing was used by Zea et al. to create a printed pH sensor made of polyaniline, polypyrrole, and polyelectrolyte poly (sodium 4-styrenesulphonate) (PSS) based on PANI: PSS/PPy: PSS inks, which were deposited on a gold microelectrode printed on a flexible substrate (Figure 7). The gold ink functionalized with phthalocyanin as the substrate enhanced the conductivity by improving the electron mobility between the conductive polymer chain and gold nanoparticles. A linear super-Nernstian response (81.2 0.5 mV/pH unit) over a wide pH range (pH 3–10) makes it a promising sensor for various applications [79]. Furthermore, various inkjet-printed conducting polymers used in sensing applications are summarized in Table 2. In spite of the excellent potential of inkjet-printable conductive polymers, PANI conductive-polymer-based sensors have some issues such as cross-selectivity, short sensor lifetime, speed, and instability, which is a major drawback for conductive-polymer-based sensor technology. The durability and sensitivity of sensors are highly dependent upon conducting polymer nanocomposites and the interaction between organic and inorganic materials.

**Table 2.** Conductive-polymer sensor inks for inkjet printing.

Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	PANI-PEC dispersion in ethanol was printed on Whatman filter paper, followed by UV sterilization	Qualitative and quantitative detection of <i>E. coli</i> in each solution	Incredible sensitivity of $0.52 \pm 0.17 \log \text{CFU/mL}$ Simple and cost-effective	[62]

Table 2. Cont.

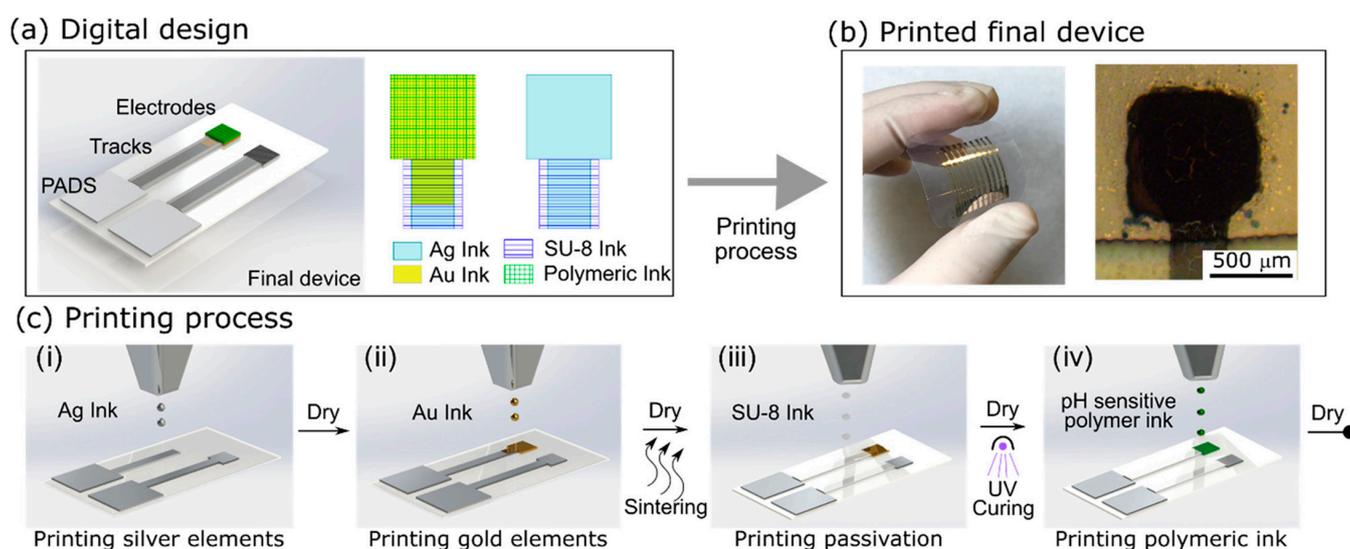
Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	Acrylic resin-based inks were applied to SDS stabilized PANI lattices while maintaining a PANI: resin weight ratio of 1:1 (dry basis)	Compatible for analysis in wearable systems	Improved viscosity as well as wear resistance Stable electrodes with reproducible pH-sensing ability Great pH sensitivity (up to 69.1 mV/pH)	[63]
Polyaniline	Silver and carbon IDA's were screen printed while nano-PANI suspension was inkjet printed	Detection of ammonia in air	Thermally stable sensor with high sensitivity to gaseous ammonia Unaffected by moisture and volatile organic compounds Can be used at elevated temperatures Very responsive in the analytically important (1–100 ppm) range	[64]
Polyaniline	Inkjet-printed silver electrodes on Si/SiO <sub>2</sub> were further drop-coated with a blend film of the emeraldine salt form of PANI and ethylene glycol (EG)	Detection of ammonia	Reliable output in the range of 0 to 100 ppm of ammonia gas Fast recovery time of about 15 min	[65]
Polyaniline	Alternate PANI and CuCl <sub>2</sub> layers were inkjet printed on silver and carbon interdigitated electrodes	Hydrogen-sulfide sensors for short-term analyses	High sensitivity to hydrogen sulfide of up to 2.5 ppmv (parts per million by volume)	[66]
Polyaniline	Inkjet-printed silver electrodes were drop-coated with dispersions of PANI/CuCl <sub>2</sub> to form films	Food quality monitoring	Appreciable sensitivity of up to 10 ppm due to protonation of PANI by H <sub>2</sub> S Low-cost H <sub>2</sub> S gas detector and can be used for food quality monitoring The relatively low absolute resistance values allow for the switching on of an LED using a low-voltage battery in a simple sensor circuit	[67]
Polyaniline	Screen-printed carbon electrode (SPCE) was subjected to inkjet printing with PANI to form a working electrode	Efficient sensor for ascorbic acid	Good sensitivity of 17.7 $\mu$ A/mM for ascorbic acid Low-cost, disposable, and point-of-care sensor	[68]
Polyaniline	The PANI ink was synthesized via oxidative polymerization	Low-cost RFID tags, polymer-based photovoltaic cells and in printed flexible electronic devices	Efficient at room temperature	[69]

Table 2. Cont.

Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	The substrate contains three carbon working electrodes and an Ag/AgCl shared counter and reference electrode. PANI hydrogel was synthesized from phytic acid, ammonium persulfate, and aniline.	The biosensor is efficient and multipurpose with capability of detecting glucose, lactates, and triglycerides with high accuracy. Integrated multiplexed biosensors for monitoring of parameters in humans can be mass produced.	Easy multiple-analyte detection by multiplexing multiple sensors on a chip. The sensitivity with respect to triglycerides was found to be $7.49 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$ between 0.1 mM and 6 mM, while that of lactate was $3.94 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$ between 0.08 mM and 5 mM. The glucose sensitivity exhibited by the sensor was $5.03 \mu\text{A}/\text{mM}^{-1} \text{cm}^{-2}$ between 1 mM and 25 mM.	[70]
Polyaniline	Screen printing of carbon electrodes onto PET plates, followed by inkjet printing of PANI NPs, and urease enzyme solution on the working electrodes.	Efficient urea detector in human serum samples.	An efficient sensor to measure ammonia in the 0.1–100 mM range and urea in the 2–12 mM range ( $r^2 = 0.98$ ).	[71]
PEDOT, polyaniline	Inkjet-printed gold NPs function as the working and counter electrodes. Inkjet-printed silver nanoparticle electrode functions as the reference electrode.	Good pH and glucose sensors.	Reusable after rinsing the aqueous samples. Sensitive to pH even after five weeks of storage. Paper chip allows for fast and on-site analysis. Cost-effective and easy. Functions with low sample volumes.	[72]
Polyaniline	PANI was used as NP, and the enzymes were drop-coated in parallel on the electrodes.	Enzyme biosensing.	Mass production is possible as no electrochemical processes are involved in fabrication.	[73]
Polyaniline	Inkjet printing on PET films by deposition of PANI patterns and immobilization of RGD peptide over it by covalent linkages.	Sensing biomolecules in live cells. Neurotransmitter detection from live cells, tracking biomolecular release, and detection of exocytosed biomolecules.	Good ability to translate and amplify exocytosis molecules into a detectable signals.	[74]
Polyaniline	Inkjet printing of multiwalled carbon nanotube electrodes, followed by printing of randomly oriented PANI nanowires dispersed in an aqueous medium.	pH and $\text{H}_2\text{O}_2$ sensor.	200-micrometer minimum printing resolution. Point-of-care diagnostics.	[75]

Table 2. Cont.

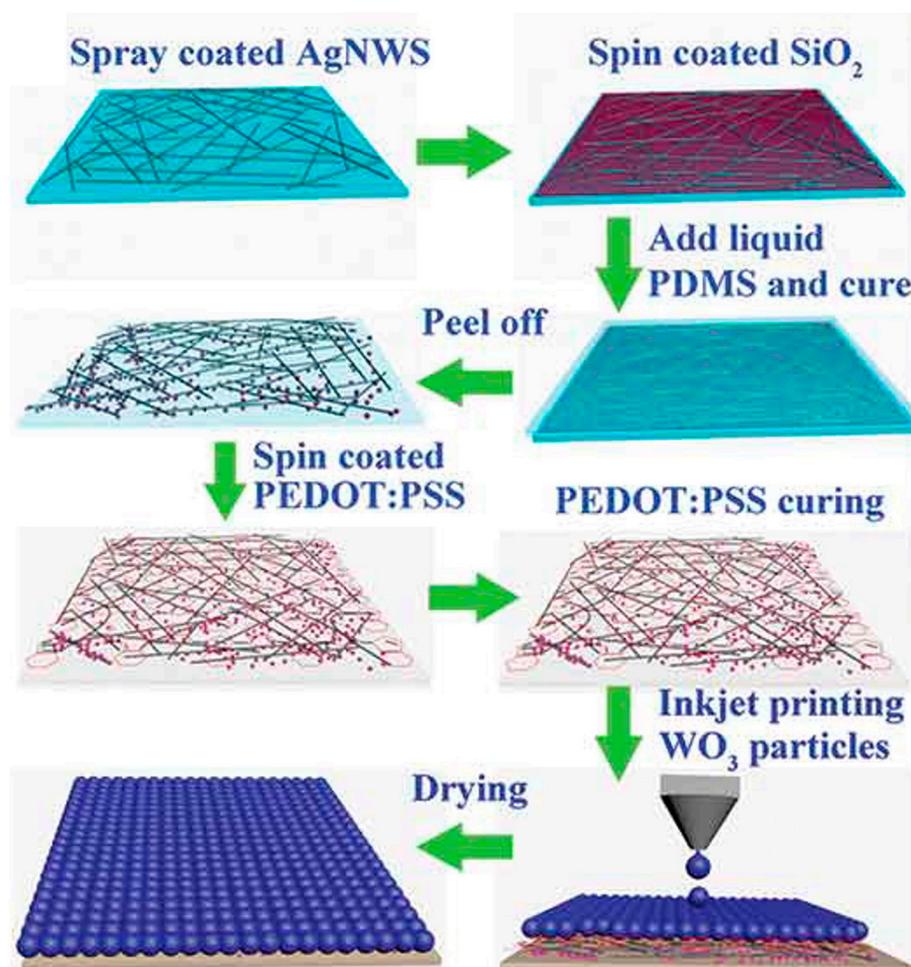
Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	Sequential inkjet printing of carbon nanotubes and polyaniline nanowires along with the glucose oxidase and platinum nanoparticle layers between the CNT layers	Excellent glucose sensor that has the potential to be an on-demand printable point-of-care diagnostic kit for glucose measurement	Quick and disposable Linear relationship between current measured and glucose concentration with a detection limit of 2 mM of glucose	[76]
Polyaniline	The AQ-PNA probe was immobilized on the working electrode, i.e., inkjet-printed G-PANI conductive ink onto the screen-printed carbon ink	A cost-effective sensor that can be incinerated for screening and monitoring of the amount of HPV-DNA type 16 to diagnose cervical cancer	A linear response range of 10–200 nM was obtained The detection limit of HPV type 16 DNA was found to be 2.3 nM Highly sensitive ePAD DNA biosensor	[77]
Biaxially oriented polypropylene covered with silica oxide (BOPP-SiO <sub>4</sub> )	The inkjet-printed soft photomasks were used for depositing organic polymers and inorganic materials on polymer films	Depositing organic polymers and inorganic material on polymer films Photografting organic polymers onto a polymer film Patterning on non-planar substrates	Utilized for making intricate patterns on non-planar substrates, microsensors, optical structures, and other devices that do not need to be extremely durable or dimensionally stable	[78]
Polyaniline, polypyrrole, and PSS	Inkjet printing of polyaniline, polypyrrole, and poly(sodium 4-styrenesulphonate) (PSS)-based inks deposited on gold microelectrode	Sensitive pH sensor that is stable over a wide pH range	Low-cost and disposable A linear super-Nernstian response (81.2 ± 0.5 mV/pH unit) over a wide pH range (pH 3–10) is obtained	[79]



**Figure 7.** Schematic depicting the (a) desired design patterns for a pH sensor, (b) microscopic image of the printed electrodes, and (c) the inkjet printing process: (i) printing of Ag RE, tracks, and pads, followed by a drying step; (ii) printing of the Au IE, followed by a drying step and thermal sintering of both metallic Au and Ag layers; (iii) printing of the dielectric SU-8, drying, and UV cross-linking; and (iv) printing of CP inks, followed by a drying step. Reproduced with permission from [79].

### 2.3. Conductive Polymers for Electrochromic Devices

Electrochromic devices can change their color reversibly and find use in display systems, smart windows, and camouflage garments [80–82]. Deformable or flexible electrochromic devices without any deterioration in performance are capable of indicating stored energy levels by visual color variation. Inkjet-printed stretchable transmissive electrochromic energy storage with  $\text{WO}_3$  nanoparticles on a transparent electrode was fabricated [83]. The silver nanowires were spray coated on a glass slide, and  $\text{SiO}_2$  nanoparticles were spin coated on the AgNW/glass slide surface. PDMS (polydimethylsiloxane) was poured over the transparent conductive electrode, cured, and the composite film was removed from the glass slide. The PEDOT:PSS buffer layer was spin coated on to the ST AgNWs/PDMS, followed by inkjet printing of the  $\text{WO}_3$  nanoparticles with a few-nanometer thickness (Figure 8). The electrode demonstrated large optical modulation of 40%, fast switching speed ( $<4.5$  s), high coloration efficiency ( $75.5 \text{ cm}^2 \text{ C}^{-1}$ ), good stability, and high specific capacity ( $32.3 \text{ mAh g}^{-1}$  and  $44.8 \text{ mAh cm}^{-3}$ ). The electrode demonstrated good functionality when stretched up to 50%, and electrochromic performance was maintained even when stretched further to 80% strain. Devices assembled with  $\text{WO}_3$ /PEDOT:PSS/AgNWs/PDMS negative hybrid electrodes, PANI/MWCNT composite positive electrodes,  $\text{H}_2\text{SO}_4$ /PVA gel electrolyte, and 3 M VHB tape as a separator showed good performance, making them well suited for wearable devices.



**Figure 8.** Schematic illustration of fabrication procedure of the inkjet-printed stretchable  $\text{WO}_3$  transparent electrode. Reproduced with permission from [83].

Small et al. reported the synthesis and inkjet processing of polyaniline/MWCN water dispersible composite inks. These films allowed for the switching between yellow, green,

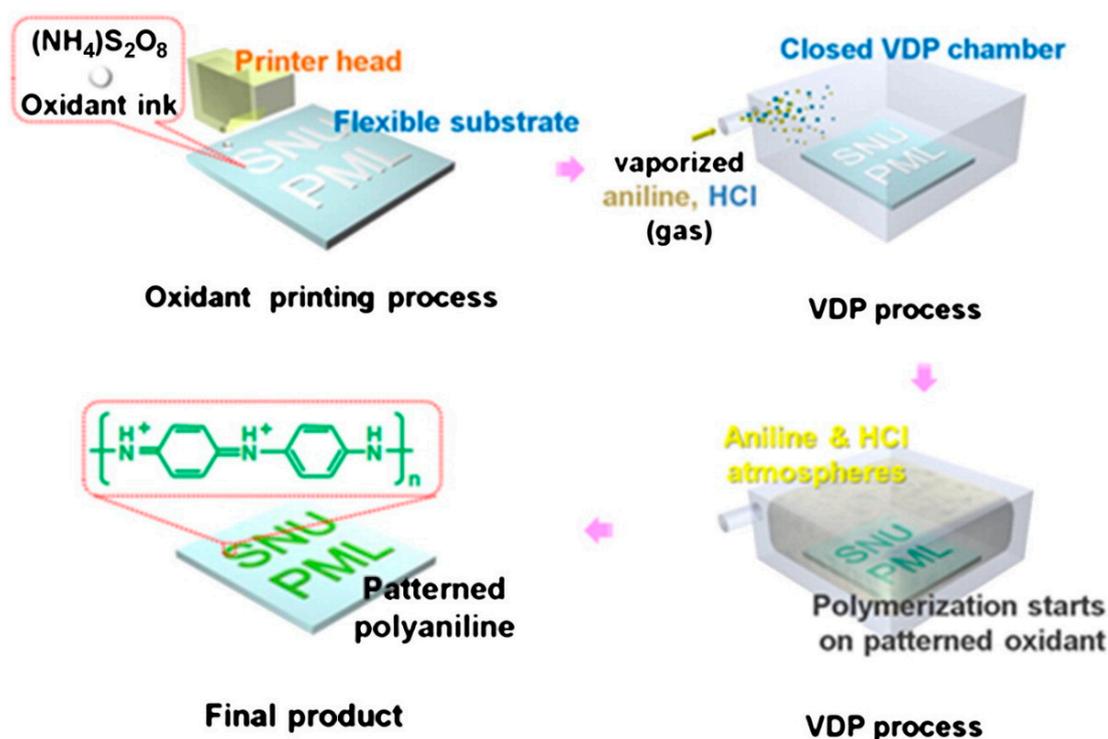
and blue when printed onto photopaper, PET, Pt-ITO, and Au-PVDF substrates and exhibited excellent optical transparency, sheet resistance, and electrochromic activity. A change in sheet resistance (1000–5000 ohm sq<sup>-1</sup>) and optical transmittance (30–70%) was reported with a change in the nanotube percentage [84]. PANI–silica and PEDOT–silica composites formed by oxidative polymerization of the monomers aniline and ethylenedioxythiophene (EDOT) on 20 nm sized silica sol particles were converted via solvent exchange to intrinsically conductive-polymer inks, which were inkjet printed on indium-tin-oxide-coated poly (ethylene terephthalate) films. PANI–silica or PEDOT–silica/ITO-PET as well as a blend of the two composites were studied as active layers in electrochromic devices fabricated using polyethylene glycol methacrylate (PEGMA)-based polymeric electrolytes. The color of the devices changed with the change in potential. The color could also be tuned by inkjet-printed PANI–silica and PEDOT–silica blended particles as an electrochromic layer [85]. The various electronic and optoelectronics devices fabricated via inkjet printing of conductive polymers are summarized in Table 3. The stability and conductivity of conductive polymers are major factors that determine the performance of electrochromic devices. Although conductive-polymer-based electrochromic devices are promising candidates for commercial-scale use, different challenges need to be addressed, such as non-dispersion and uniform morphology and size as comparable to metal oxide electrochromic devices.

**Table 3.** Conductive-polymer electrochromic inks for inkjet printing.

Conductive Polymer	Ink Composition and Device Fabrication	Applications	Highlights	Reference
WO <sub>3</sub> nanoparticles	PEDOT:PSS buffer layer was spin coated onto the ST AgNW/PDMS followed by inkjet printing of the WO <sub>3</sub> nanoparticle layer	Deformable and wearable electronics STEESDs with novel features	Large optical modulation of 40%, fast switching speed (<4.5 s), high coloration efficiency (75.5 cm <sup>2</sup> C <sup>-1</sup> ), and good stability and high specific capacity (32.3 mAh g <sup>-1</sup> and 44.8 mAh cm <sup>-3</sup> ) observed Good functionality and maintenance of electrochromic performance even when stretched up to 50–80% strain	[83]
Polyaniline	Water-soluble polyaniline composite materials with MWNT were dispersed in water and deposited via inkjet printing, yielding transparent conductive electroactive films	Development of inkjet printing as a viable tool for the fabrication of transparent conductive electroactive materials	These films allowed for the switching between yellow, green, and blue when printed onto photopaper, PET, Pt-ITO, and Au-PVDF substrates. A change in sheet resistance (1000–5000 ohm sq <sup>-1</sup> ) and optical transmittance (30–70%) was reported with a change in the nanotube percentage	[84]
PANI–silica and PEDOT–silica	PANI–silica and PEDOT–silica composites converted via solvent exchange to intrinsically conductive-polymer inks, which were inkjet printed on indium-tin-oxide-coated poly (ethylene terephthalate) films	Electrochromic display device fabrication with various intrinsically conductive-polymer colloidal solutions	Color of the devices changed with change in potential. The color could also be tuned by inkjet-printed PANI–silica and PEDOT–silica blended particles as an electrochromic	[85]

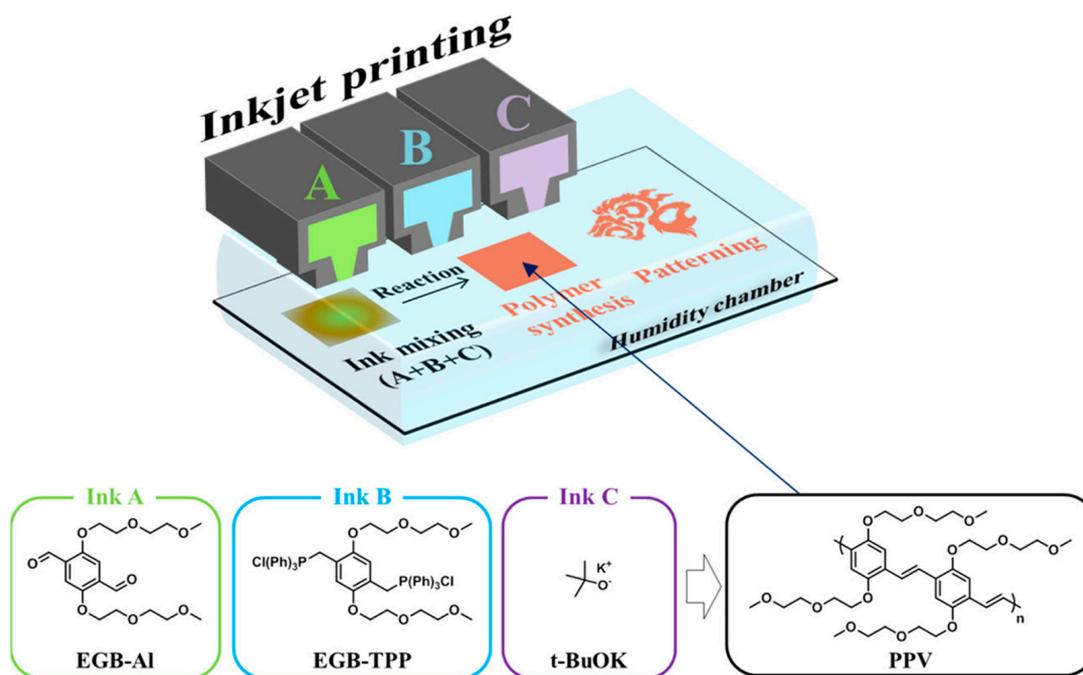
#### 2.4. Patterning with Conductive Polymers

Conductive polymers have been patterned using various techniques like template-assisted synthesis, photolithography, softlithography, electrochemical deposition and dip-pen nanolithography [86]. Conductive polymers have also been inkjet printed directly onto substrates, but the method has certain limitations. Conductive polymers like polypyrrole, polyaniline (PANI), and polythiophene and their derivatives are insoluble in common polar solvents, and inks are usually prepared using stabilizers and surfactants to achieve the desired viscosity and surface tension. Vapor deposition polymerization (VDP)-mediated inkjet printing (VDP-IJP) can overcome some of the processability issues. Jang and coworkers suggested a simple method to pattern conductive polymers using vapor deposition polymerization (VDP)-mediated inkjet printing (VDP-IJP), wherein uniform, well-defined patterns of PANI were formed by chemical oxidation polymerization of aniline vapor on oxidant patterns that were previously inkjet printed in the VDP chamber (Figure 9). This process resulted in the formation of densely connected polyaniline nanofibers without the use of surfactants and stabilizers, resulting in patterned PANI being electrically active with low sheet resistance in the range of  $10^3 \Omega \text{sq}^{-1}$  on the polymeric substrate [87].



**Figure 9.** Scheme for patterning conducting polymer using vapor deposition polymerization (VDP)-mediated inkjet printing (VDP-IJP). Reproduced with permission from [87].

Reactive inkjet printing can be used to pattern functional organic materials on a solid substrate with a small amount of reagent. A new strategy for the formation of poly(phenylenevinylene) (PPV) patterns on paper by using the reactive inkjet printing (RIJ) method was reported by Jeon and coworkers (Figure 10). Synthesized hydrophilic terephthaldehyde (ink A), bis(triphenylphosphonium salt) (ink B), and potassium t-butoxide (ink C) were printed using an inkjet printer under high humidity conditions in order (A, B, and C) at the same location (overprinting), and an in situ Wittig reaction ensued, resulting in the formation of PPV patterns [88]. The unreacted reagents and byproducts could be efficiently removed by washing with water and chloroform, as the prepared PPV was found to be insoluble in these solvents.



**Figure 10.** Formation of poly(phenylenevinylene) (PPV) patterns on paper by using the reactive inkjet printing (RIJ) method. Inks A, B, and C are inkjet printed at the same location. They react to produce the PPV pattern [88].

Inkjet printing of conductive polymer patterns on the surface of electrospun scaffolds using cell stimulation has been explored. As a part of the investigations, gelatin (Gel) and polycaprolactone (PCL) were modified with calcium phosphate nanoparticles (SG5) and Osteogenon (Osteo) drugs, respectively, and further utilized for the preparation of a bilayer scaffold by accumulating electrospun PCL/Osteo and Gel/SG5 fibers in a common collector [89]. Inkjet printing was then used to deposit thin PANI patterns on the surface of the hybrid bilayer scaffold. A bioactive hybrid scaffold system that can provide an electrically conductive environment for cells by bringing together conductive PANI, bioactive particles, and drugs in an electrospun scaffold was prepared. A combination of nanotemplating and inkjet printing was used to prepare nanopatterned thin polymer films. Nanoperforated  $\text{TiO}_2$  (NP- $\text{TiO}_2$ ) functionalized with hydrophobic perfluorinated phosphate Zonyl FSE (ZFNP- $\text{TiO}_2$ ) fabricated on top of silicon wafers were used as substrates. Biotinylated polythiophene derivative (PBTL) ink, a biosensor material, and dinonylnaphthalene-sulfonic-acid-doped polyaniline (PANI-DNNSA) ink, a pH- and gas-sensing material, were inkjet printed on NP- $\text{TiO}_2$  and ZFNP- $\text{TiO}_2$  substrates [90]. The non-functionalized polymer had higher surface energy and allowed for the deposition of uniform thin films. They have uses in biosensing and electrical devices. Aniline dimer DANI was used to produce PANI via oxidative polymerization in an aqueous medium using polystyrene sulphonate (PSS) as an emulsifying and doping agent. Controlling the amount of the oxidant resulted in mixed leucoemeraldine and emeraldine oxidation states. Solubilized in DMSO, PANI/PSS was suitable as an ink for inkjet printing on flexible substrates and gave interesting negative capacitance effects [16]. Inkjet-printed conductive polymers for nanopatterning are summarized in Table 4.

**Table 4.** Conductive-polymer pattern inks for inkjet printing.

Conductive Polymer	Ink Formulation and Device Fabrication	Applications	Highlights	Reference
Polyaniline	Vapor deposition polymerization (VDP)-mediated inkjet printing (VDP-IJP) The substrate undergoes chemical oxidation polymerization at elevated temperatures that form emeraldine salt PANI patterns	Micro-range accuracy Efficient PANI synthesis	Minimum width of patterned line = 80 $\mu\text{m}$ Average sheet resistance = $3.8 \times 10^3 \Omega\text{sq}^{-1}$ Does not require surfactants or stabilizers	[87]
Poly(phenylenevinylene) (PPV)	Reactive inkjet printing An in situ Wittig reaction results in the formation of PPV patterns	Readily generated PPV microarrays Patterning of functional organic materials on a solid substrate	High processability Easy removal of unreacted reagents and byproducts by dissolution in organic media	[88]
Polyaniline	Electrospinning to obtain a bilayer biodegradable scaffold for PANI printing	Bone tissue engineering	Stimulation of cellular functions like attachment, proliferation, migration, and differentiation	[89]
Polyaniline	Combination of nanotemplating and inkjet printing PBTL, and PANI-DNNSA inks were inkjet printed onto NP-TiO <sub>2</sub> and ZFNP-TiO <sub>2</sub> substrates	Biosensing and electronic devices	Low-cost and efficient	[90]
Polyaniline	Oxidative polymerization in an aqueous medium using polystyrene sulfonate (PSS) as an emulsifying or doping agent Controlling the amount of the oxidant resulted in mixed leucoemeraldine or emeraldine oxidation states	Ideal for devices where positive parasitic capacitances have to be compensated	Synthesis from the dimer DANI Negative capacitance Cost-effective and simple	[16]

### 3. Conclusions and Future Perspectives

In the past years, various inkjet printable conductive polymers such as PANI, PPV, polyaniline, PEDOT, etc., and their composites have been explored for various applications, with a significant improvement in device performance as the conductive and mechanical properties of conductive polymers are tunable. We have primarily reviewed conducting polymer PANI-based inks, and device fabrication using this conductive polymer ink via inkjet printing for a variety of applications such as supercapacitors, sensors, and electrochromic devices. The ink formulations for inkjet printing and the key improvements in devices were summarized in the tables in this article. The inkjet printing of conductive polymer PANI for nanopatterning directly on substrates was also briefly discussed in this review. The properties of conductive polymers need to be further tuned using optimized device configurations for better device performance.

Inkjet printing has proven to be a simple, cost-effective, and green method that saves material and deposits the ink on large areas uniformly, as well as allows accurate patterning. With more research into tuning the properties of conductive polymers like PANI

and PANI-based inks, and also with strides in the inkjet printing process, scalable flexible device fabrication with optimum performance is achievable. Improving conducting polymer nanocomposites via modifying the surface and functionalization with inorganic particles and substrates, using inorganic particles with a high aspect ratio, etc., are possible approaches that could be adopted for improving conductive-polymer-based sensors [91]. We believe this review provides an overview of recent developments in inkjet-printable conductive polymers for various applications and a new direction for next-generation conductive-polymer-based devices for various applications. The inkjet printing ability of conductive polymers provides an endless number of prospects for designing highly efficient devices with high reproducibility at a low cost, which can be easily scaled up at the commercial level to make technologies commercially viable. With improvements in inks and fabrication technology, scalable, low-cost, single-use, disposable glucose, urea, pH, gas, and enzyme sensors, along with better-performing supercapacitors and electrochromic devices, are achievable.

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## References

1. Oliveira, J.; Correia, V.; Castro, H.; Martins, P.; Lanceros-Mendez, S. Polymer-Based Smart Materials by Printing Technologies: Improving Application and Integration. *Addit. Manuf.* **2018**, *21*, 269–283. [[CrossRef](#)]
2. Wu, S.; Zeng, T.; Liu, Z.; Ma, G.; Xiong, Z.; Zuo, L.; Zhou, Z. 3D Printing Technology for Smart Clothing: A Topic Review. *Materials* **2022**, *15*, 7391. [[CrossRef](#)] [[PubMed](#)]
3. Mendes-Felipe, C.; Oliveira, J.; Etxebarria, I.; Vilas-Vilela, J.L.; Lanceros-Mendez, S. State-of-the-Art and Future Challenges of UV Curable Polymer-Based Smart Materials for Printing Technologies. *Adv. Mater. Technol.* **2019**, *4*, 1800618. [[CrossRef](#)]
4. Borghetti, M.; Cantù, E.; Sardini, E.; Serpelloni, M. Future Sensors for Smart Objects by Printing Technologies in Industry 4.0 Scenario. *Energies* **2020**, *13*, 5916. [[CrossRef](#)]
5. Kim, A.; Oh, S.H.; Adhikari, A.; Sathe, B.R.; Kumar, S.; Patel, R. Recent Advances in Modified Commercial Separators for Lithium–Sulfur Batteries. *J. Mater. Chem. A* **2023**, *11*, 7833–7866. [[CrossRef](#)]
6. Patel, M.; Patel, R.; Park, C.; Cho, K.; Kumar, P.; Park, C.; Koh, W.-G. Water-Stable, Biocompatible, and Highly Luminescent Perovskite Nanocrystals-Embedded Fiber-Based Paper for Anti-Counterfeiting Applications. *Nano Converg.* **2023**, *10*, 21. [[CrossRef](#)]
7. Kim, A.; Wert, N.A.; Gowd, E.B.; Patel, R. Recent Progress in PEG-Based Composite Phase Change Materials. *Polym. Rev.* **2023**, 1–52. [[CrossRef](#)]
8. Kim, A.; Dash, J.K.; Kumar, P.; Patel, R. Carbon-Based Quantum Dots for Photovoltaic Devices: A Review. *ACS Appl. Electron. Mater.* **2022**, *4*, 27–58. [[CrossRef](#)]
9. Patel, M.; Meenu, M.; Pandey, J.K.; Kumar, P.; Patel, R. Recent Development in Upconversion Nanoparticles and Their Application in Optogenetics: A Review. *J. Rare Earths* **2022**, *40*, 847–861. [[CrossRef](#)]
10. Gupta, B.K.; Kumar, P.; Kedawat, G.; Kanika; Vithayathil, S.A.; Gangwar, A.K.; Singh, S.; Kashyap, P.K.; Lahon, R.; Singh, V.N.; et al. Tunable Luminescence from Two Dimensional BCNO Nanophosphor for High-Contrast Cellular Imaging. *RSC Adv.* **2017**, *7*, 41486–41494. [[CrossRef](#)]
11. Gupta, B.K.; Kedawat, G.; Kumar, P.; Singh, S.; Suryawanshi, S.R.; Agrawal (Garg), N.; Gupta, G.; Kim, A.R.; Gupta, R.K.; More, M.A.; et al. Field Emission Properties of Highly Ordered Low-Aspect Ratio Carbon Nanocup Arrays. *RSC Adv.* **2016**, *6*, 9932–9939. [[CrossRef](#)]
12. Hrytsenko, O.; Hrytsenko, D.; Shvalagin, V.; Grodziuk, G.; Kompanets, M. The Use of Carbon Nanoparticles for Inkjet-Printed Functional Labels for Smart Packaging. *J. Nanomater.* **2018**, *2018*, 6485654. [[CrossRef](#)]
13. Cirelli, M.; Hao, J.; Bor, T.C.; DuVigneau, J.; Benson, N.; Akkerman, R.; Hempenius, M.A.; Vancso, G.J. Printing “Smart” Inks of Redox-Responsive Organometallic Polymers on Microelectrode Arrays for Molecular Sensing. *ACS Appl. Mater. Interfaces* **2019**, *11*, 37060–37068. [[CrossRef](#)] [[PubMed](#)]
14. Seipel, S.; Yu, J.; Periyasamy, A.P.; Víková, M.; Vik, M.; Nierstrasz, V.A. Inkjet Printing and UV-LED Curing of Photochromic Dyes for Functional and Smart Textile Applications. *RSC Adv.* **2018**, *8*, 28395–28404. [[CrossRef](#)] [[PubMed](#)]

15. Yu, J.; Seipel, S.; Nierstrasz, V.A. Digital Inkjet Functionalization of Water-Repellent Textile for Smart Textile Application. *J. Mater. Sci.* **2018**, *53*, 13216–13229. [[CrossRef](#)]
16. Bocchini, S.; Chiolerio, A.; Porro, S.; Accardo, D.; Garino, N.; Bejtka, K.; Perrone, D.; Pirri, C.F. Synthesis of Polyaniline-Based Inks, Doping Thereof and Test Device Printing towards Electronic Applications. *J. Mater. Chem. C* **2013**, *1*, 5101–5109. [[CrossRef](#)]
17. Nair, N.M.; Pakkathillam, J.K.; Kumar, K.; Arunachalam, K.; Ray, D.; Swaminathan, P. Printable Silver Nanowire and PEDOT:PSS Nanocomposite Ink for Flexible Transparent Conducting Applications. *ACS Appl. Electron. Mater.* **2020**, *2*, 1000–1010. [[CrossRef](#)]
18. Singh, A.; Katiyar, M.; Garg, A. Understanding the Formation of PEDOT:PSS Films by Ink-Jet Printing for Organic Solar Cell Applications. *RSC Adv.* **2015**, *5*, 78677–78685. [[CrossRef](#)]
19. Yang, C.-Y.; Stoeckel, M.-A.; Ruoko, T.-P.; Wu, H.-Y.; Liu, X.; Kolhe, N.B.; Wu, Z.; Puttison, Y.; Musumeci, C.; Massetti, M.; et al. A High-Conductivity n-Type Polymeric Ink for Printed Electronics. *Nat. Commun.* **2021**, *12*, 2354. [[CrossRef](#)]
20. Liu, X.; Shen, Y.; Yang, R.; Zou, S.; Ji, X.; Shi, L.; Zhang, Y.; Liu, D.; Xiao, L.; Zheng, X.; et al. Inkjet Printing Assisted Synthesis of Multicomponent Mesoporous Metal Oxides for Ultrafast Catalyst Exploration. *Nano Lett.* **2012**, *12*, 5733–5739. [[CrossRef](#)]
21. Negro, A.; Cherbuin, T.; Lutolf, M.P. 3D Inkjet Printing of Complex, Cell-Laden Hydrogel Structures. *Sci. Rep.* **2018**, *8*, 17099. [[CrossRef](#)]
22. Willert, A.; Tabary, F.Z.; Zubkova, T.; Santangelo, P.E.; Romagnoli, M.; Baumann, R.R. Multilayer Additive Manufacturing of Catalyst-Coated Membranes for Polymer Electrolyte Membrane Fuel Cells by Inkjet Printing. *Int. J. Hydrogen Energy* **2022**, *47*, 20973–20986. [[CrossRef](#)]
23. Hussain, A.; Abbas, N.; Ali, A. Inkjet Printing: A Viable Technology for Biosensor Fabrication. *Chemosensors* **2022**, *10*, 103. [[CrossRef](#)]
24. Criado-Gonzalez, M.; Dominguez-Alfaro, A.; Lopez-Larrea, N.; Alegret, N.; Mecerreyes, D. Additive Manufacturing of Conducting Polymers: Recent Advances, Challenges, and Opportunities. *ACS Appl. Polym. Mater.* **2021**, *3*, 2865–2883. [[CrossRef](#)]
25. Shirakawa, H.; Louis, E.J.; MacDiarmid, A.G.; Chiang, C.K.; Heeger, A.J. Synthesis of Electrically Conducting Organic Polymers: Halogen Derivatives of Polyacetylene, (CH). *J. Chem. Soc. Chem. Commun.* **1977**, *16*, 578–580. [[CrossRef](#)]
26. Shirakawa, H.; MacDiarmid, A.; Heeger, A. Twenty-Five Years of Conducting Polymers. *Chem. Commun.* **2003**, *1*, 1–4. [[CrossRef](#)]
27. Guo, X. Conducting Polymers Forward. *Nat. Mater.* **2020**, *19*, 921. [[CrossRef](#)]
28. El-Bery, H.M.; Salah, M.R.; Ahmed, S.M.; Soliman, S.A. Efficient Non-Metal Based Conducting Polymers for Photocatalytic Hydrogen Production: Comparative Study between Polyaniline, Polypyrrole and PEDOT. *RSC Adv.* **2021**, *11*, 13229–13244. [[CrossRef](#)]
29. Tajik, S.; Beitollahi, H.; Nejad, F.G.; Shoaie, I.S.; Khalilzadeh, M.A.; Asl, M.S.; Van Le, Q.; Zhang, K.; Jang, H.W.; Shokouhimehr, M. Recent Developments in Conducting Polymers: Applications for Electrochemistry. *RSC Adv.* **2020**, *10*, 37834–37856. [[CrossRef](#)]
30. Namsheer, K.; Rout, C.S. Conducting Polymers: A Comprehensive Review on Recent Advances in Synthesis, Properties and Applications. *RSC Adv.* **2021**, *11*, 5659–5697. [[CrossRef](#)]
31. Benny Mattam, L.; Bijoy, A.; Abraham Thadathil, D.; George, L.; Varghese, A. Conducting Polymers: A Versatile Material for Biomedical Applications. *ChemistrySelect* **2022**, *7*, e202201765. [[CrossRef](#)]
32. Atkare, S.; Hambir, S.; Jagtap, S.; Adhikari, A.; Singh, S.K.; Patel, R. Role of Polyaniline/Molybdenum Trioxide Nanocomposites in Tuning the Characteristics of Humidity Sensors. *Polym. Adv. Technol.* **2023**, *34*, 2585–2596. [[CrossRef](#)]
33. Kumaravel, S.; Kim, E.; Kale, B.B.; Adhikari, A.; Patel, R.; Kundu, S. Recent Developments in Conductive Polymer-Based Electro-/Photoelectrocatalytic Materials for Effective Hydrogen/Oxygen Evolution Reactions: A Review. *ChemElectroChem* **2022**, *9*, e202200724. [[CrossRef](#)]
34. Park, S.; Patel, R. Recent Progress in Conductive Polymer-Based Membranes. *Membr. J.* **2021**, *31*, 101–119. [[CrossRef](#)]
35. Li, J.; Huckleby, A.B.; Zhang, M. Polymer-Based Thermoelectric Materials: A Review of Power Factor Improving Strategies. *J. Mater.* **2022**, *8*, 204–220. [[CrossRef](#)]
36. Nasir, A.; Raza, A.; Tahir, M.; Yasin, T.; Nadeem, M.; Ahmad, B. Synthesis and Study of Polyaniline Grafted Graphene Oxide Nanohybrids. *Mater. Res. Bull.* **2023**, *157*, 112006. [[CrossRef](#)]
37. Yao, F.; Xie, W.; Ma, C.; Wang, D.; El-Bahy, Z.M.; Helal, M.H.; Liu, H.; Du, A.; Guo, Z.; Gu, H. Superb Electromagnetic Shielding Polymer Nanocomposites Filled with 3-Dimensional p-Phenylenediamine/Aniline Copolymer Nanofibers@copper Foam Hybrid Nanofillers. *Compos. Part B Eng.* **2022**, *245*, 110236. [[CrossRef](#)]
38. Dandapani; Devendra, K.; Revannasiddappa; Vishnu, K.R. Thermal Stability and Electromagnetic Interference of Epoxy-Graphene/Hybrid Composite Materials. *Mater. Today Proc.* **2022**, *66*, 1664–1670. [[CrossRef](#)]
39. Thakur, Y.S.; Acharya, A.D.; Sharma, S. Bhawna Reinforcement of V<sub>2</sub>O<sub>5</sub> Nanoparticle in Polyaniline to Improve the Optical and UV- Shielding Properties. *Results Opt.* **2023**, *11*, 100400. [[CrossRef](#)]
40. Ray, B.; Parmar, S.; Date, K.; Datar, S. Optically Transparent Polymer Composites: A Study on the Influence of Filler/Dopant on Electromagnetic Interference Shielding Mechanism. *J. Appl. Polym. Sci.* **2021**, *138*, 50255. [[CrossRef](#)]
41. Zhu, N.; Jiang, T.; Zeng, X.; Li, S.; Shen, C.; Zhang, C.; Gong, W.; He, L. High Strength and Light Weight Polyamide 6/Carbon Fiber Composite Foams for Electromagnetic Interference Shielding. *J. Appl. Polym. Sci.* **2023**, *140*, e53818. [[CrossRef](#)]
42. Qi, H.; Wang, G.; Ma, Q.; Li, D.; Dong, X.; Yu, W.; Wang, J.; Liu, G.; Zhang, X. Conjugative Electrospinning towards Janus-Type Nanofibers Array Membrane Concurrently Displaying Dual-Functionality of Improved Red Luminescence and Tuneable Superparamagnetism. *J. Mater. Sci. Mater. Electron.* **2022**, *33*, 4438–4449. [[CrossRef](#)]

43. Guo, X.; Xue, Z.; Zhang, Y. Manufacturing of 3D Multifunctional Microelectronic Devices: Challenges and Opportunities. *NPG Asia Mater.* **2019**, *11*, 29. [[CrossRef](#)]
44. Satoh, Y.; Ding, H.; Yang, H.; Deng, Y.; Hsueh, A.-J.; Shimizu, T.; Qiao, M.; Ma, C.; Kariya, K.; Kurihara, T.; et al. Wired Microfabricated Electrochemical Systems. *Anal. Chem.* **2021**, *93*, 12655–12663. [[CrossRef](#)] [[PubMed](#)]
45. He, B.; Tan, L.; Regnier, F. Microfabricated Filters for Microfluidic Analytical Systems. *Anal. Chem.* **1999**, *71*, 1464–1468. [[CrossRef](#)]
46. Greci, G.; Bertocchi, C.; Ravasio, A. Integrating Microfabrication into Biological Investigations: The Benefits of Interdisciplinarity. *Micromachines* **2019**, *10*, 252. [[CrossRef](#)] [[PubMed](#)]
47. Wang, J.; Chatrathi, M.P.; Tian, B.; Polsky, R. Microfabricated Electrophoresis Chips for Simultaneous Bioassays of Glucose, Uric Acid, Ascorbic Acid, and Acetaminophen. *Anal. Chem.* **2000**, *72*, 2514–2518. [[CrossRef](#)]
48. Tiwari, S.K.; Bhat, S.; Mahato, K.K. Design and Fabrication of Low-Cost Microfluidic Channel for Biomedical Application. *Sci. Rep.* **2020**, *10*, 9215. [[CrossRef](#)]
49. Su, W.; Cook, B.S.; Fang, Y.; Tentzeris, M.M. Fully Inkjet-Printed Microfluidics: A Solution to Low-Cost Rapid Three-Dimensional Microfluidics Fabrication with Numerous Electrical and Sensing Applications. *Sci. Rep.* **2016**, *6*, 35111. [[CrossRef](#)] [[PubMed](#)]
50. Majeed, A.H.; Mohammed, L.A.; Hammoodi, O.G.; Sehgal, S.; Alheety, M.A.; Saxena, K.K.; Dadoosh, S.A.; Mohammed, I.K.; Jasim, M.M.; Salmaan, N.U. A Review on Polyaniline: Synthesis, Properties, Nanocomposites, and Electrochemical Applications. *Int. J. Polym. Sci.* **2022**, *2022*, 9047554. [[CrossRef](#)]
51. Beygisangchin, M.; Abdul Rashid, S.; Shafie, S.; Sadrolhosseini, A.R.; Lim, H.N. Preparations, Properties, and Applications of Polyaniline and Polyaniline Thin Films—A Review. *Polymers* **2021**, *13*, 2003. [[CrossRef](#)] [[PubMed](#)]
52. Park, J.T.; Patel, R.; Jeon, H.; Kim, D.J.; Shin, J.-S.; Hak Kim, J. Facile Fabrication of Vertically Aligned TiO<sub>2</sub> Nanorods with High Density and Rutile/Anatase Phases on Transparent Conducting Glasses: High Efficiency Dye-Sensitized Solar Cells. *J. Mater. Chem.* **2012**, *22*, 6131–6138. [[CrossRef](#)]
53. Ramanavicius, S.; Ramanavicius, A. Development of Molecularly Imprinted Polymer Based Phase Boundaries for Sensors Design (Review). *Adv. Colloid Interface Sci.* **2022**, *305*, 102693. [[CrossRef](#)] [[PubMed](#)]
54. Gerard, M.; Chaubey, A.; Malhotra, B.D. Application of Conducting Polymers to Biosensors. *Biosens. Bioelectron.* **2002**, *17*, 345–359. [[CrossRef](#)] [[PubMed](#)]
55. Cho, S.I.; Lee, S.B. Fast Electrochemistry of Conductive Polymer Nanotubes: Synthesis, Mechanism, and Application. *Acc. Chem. Res.* **2008**, *41*, 699–707. [[CrossRef](#)]
56. Ahmad, K.; Raza, W. *Current State and Prospective of Supercapacitors BT-Handbook of Nanomaterials and Nanocomposites for Energy and Environmental Applications*; Kharisova, O.V., Torres-Martinez, L.M., Kharisov, B.I., Eds.; Springer International Publishing: Cham, Switzerland, 2021; pp. 1835–1853, ISBN 978-3-030-36268-3.
57. Chi, K.; Zhang, Z.; Xi, J.; Huang, Y.; Xiao, F.; Wang, S.; Liu, Y. Freestanding Graphene Paper Supported Three-Dimensional Porous Graphene-Polyaniline Nanocomposite Synthesized by Inkjet Printing and in Flexible All-Solid-State Supercapacitor. *ACS Appl. Mater. Interfaces* **2014**, *6*, 16312–16319. [[CrossRef](#)]
58. Chiolerio, A.; Bocchini, S.; Porro, S. Inkjet Printed Negative Supercapacitors: Synthesis of Polyaniline-Based Inks, Doping Agent Effect, and Advanced Electronic Devices Applications. *Adv. Funct. Mater.* **2014**, *24*, 3375–3383. [[CrossRef](#)]
59. Diao, J.; Yuan, J.; Ding, A.; Zheng, J.; Lu, Z. Flexible Supercapacitor Based on Inkjet-Printed Graphene@Polyaniline Nanocomposites with Ultrahigh Capacitance. *Macromol. Mater. Eng.* **2018**, *303*, 1800092. [[CrossRef](#)]
60. Xu, Y.; Hennig, I.; Freyberg, D.; James Strudwick, A.; Georg Schwab, M.; Weitz, T.; Chih-Pei Cha, K. Inkjet-Printed Energy Storage Device Using Graphene/Polyaniline Inks. *J. Power Sources* **2014**, *248*, 483–488. [[CrossRef](#)]
61. Zhang, J.; Wang, J.; Yang, J.; Wang, Y.; Chan-Park, M.B. Three-Dimensional Macroporous Graphene Foam Filled with Mesoporous Polyaniline Network for High Areal Capacitance. *ACS Sustain. Chem. Eng.* **2014**, *2*, 2291–2296. [[CrossRef](#)]
62. Anjali, M.K.; Bharath, G.; Rashmi, H.M.; Avinash, J.; Naresh, K.; Raju, P.N.; Raghu, H.V. Polyaniline-Pectin Nanoparticles Immobilized Paper Based Colorimetric Sensor for Detection of Escherichia Coli in Milk and Milk Products. *Curr. Res. Food Sci.* **2022**, *5*, 823–834. [[CrossRef](#)] [[PubMed](#)]
63. Bilbao, E.; Kapadia, S.; Riechert, V.; Amalvy, J.; Molinari, F.N.; Escobar, M.M.; Baumann, R.R.; Monsalve, L.N. Functional Aqueous-Based Polyaniline Inkjet Inks for Fully Printed High-Performance PH-Sensitive Electrodes. *Sens. Actuators B Chem.* **2021**, *346*, 130558. [[CrossRef](#)]
64. Crowley, K.; Morrin, A.; Hernandez, A.; O'Malley, E.; Whitten, P.G.; Wallace, G.G.; Smyth, M.R.; Killard, A.J. Fabrication of an Ammonia Gas Sensor Using Inkjet-Printed Polyaniline Nanoparticles. *Talanta* **2008**, *77*, 710–717. [[CrossRef](#)]
65. Le, D.D.; Nguyen, T.N.N.; Doan, D.C.T.; Dang, T.M.D.; Dang, M.C. Fabrication of Interdigitated Electrodes by Inkjet Printing Technology for Application in Ammonia Sensing. *Adv. Nat. Sci. Nanosci. Nanotechnol.* **2016**, *7*, 025002. [[CrossRef](#)]
66. Crowley, K.; Morrin, A.; Shepherd, R.L.; In Het Panhuis, M.; Wallace, G.G.; Smyth, M.R.; Killard, A.J. Fabrication of Polyaniline-Based Gas Sensors Using Piezoelectric Inkjet and Screen Printing for the Detection of Hydrogen Sulfide. *IEEE Sens. J.* **2010**, *10*, 1419–1426. [[CrossRef](#)]
67. Sarfraz, J.; Tobjork, D.; Osterbacka, R.; Linden, M. Low-Cost Hydrogen Sulfide Gas Sensor on Paper Substrates: Fabrication and Demonstration. *IEEE Sens. J.* **2012**, *12*, 1973–1978. [[CrossRef](#)]
68. Kit-Anan, W.; Olarnwanich, A.; Sriprachubwong, C.; Karuwan, C.; Tuantranont, A.; Wisitsoraat, A.; Srituravanich, W.; Pimpin, A. Disposable Paper-Based Electrochemical Sensor Utilizing Inkjet-Printed Polyaniline Modified Screen-Printed Carbon Electrode for Ascorbic Acid Detection. *J. Electroanal. Chem.* **2012**, *685*, 72–78. [[CrossRef](#)]

69. Kulkarni, M.V.; Apte, S.K.; Naik, S.D.; Ambekar, J.D.; Kale, B.B. Ink-Jet Printed Conducting Polyaniline Based Flexible Humidity Sensor. *Sens. Actuators B Chem.* **2013**, *178*, 140–143. [[CrossRef](#)]
70. Li, L.; Pan, L.; Ma, Z.; Yan, K.; Cheng, W.; Shi, Y.; Yu, G. All Inkjet-Printed Amperometric Multiplexed Biosensors Based on Nanostructured Conductive Hydrogel Electrodes. *Nano Lett.* **2018**, *18*, 3322–3327. [[CrossRef](#)]
71. Suman; O'Reilly, E.; Kelly, M.; Morrin, A.; Smyth, M.R.; Killard, A.J. Chronocoulometric Determination of Urea in Human Serum Using an Inkjet Printed Biosensor. *Anal. Chim. Acta* **2011**, *697*, 98–102. [[CrossRef](#)]
72. Määttänen, A.; Vanamo, U.; Ihalainen, P.; Pulkkinen, P.; Tenhu, H.; Bobacka, J.; Peltonen, J. A Low-Cost Paper-Based Inkjet-Printed Platform for Electrochemical Analyses. *Sens. Actuators B Chem.* **2013**, *177*, 153–162. [[CrossRef](#)]
73. Morrin, A.; Wilbeer, F.; Ngamna, O.; Moulton, S.E.; Killard, A.J.; Wallace, G.G.; Smyth, M.R. Novel Biosensor Fabrication Methodology Based on Processable Conducting Polyaniline Nanoparticles. *Electrochem. Commun.* **2005**, *7*, 317–322. [[CrossRef](#)]
74. Oh, W.K.; Kim, S.; Shin, K.H.; Jang, Y.; Choi, M.; Jang, J. Inkjet-Printed Polyaniline Patterns for Exocytosed Molecule Detection from Live Cells. *Talanta* **2013**, *105*, 333–339. [[CrossRef](#)] [[PubMed](#)]
75. Song, E.; Tortorich, R.P.; Da Costa, T.H.; Choi, J.W. Inkjet Printing of Conductive Polymer Nanowire Network on Flexible Substrates and Its Application in Chemical Sensing. *Microelectron. Eng.* **2015**, *145*, 143–148. [[CrossRef](#)]
76. Song, E.; da Costa, T.H.; Choi, J.W. A Chemiresistive Glucose Sensor Fabricated by Inkjet Printing. *Microsyst. Technol.* **2017**, *23*, 3505–3511. [[CrossRef](#)]
77. Teengam, P.; Siangproh, W.; Tuantranont, A.; Henry, C.S.; Vilaivan, T.; Chailapakul, O. Electrochemical Paper-Based Peptide Nucleic Acid Biosensor for Detecting Human Papillomavirus. *Anal. Chim. Acta* **2017**, *952*, 32–40. [[CrossRef](#)] [[PubMed](#)]
78. Wang, L.; Ma, Y.; Chen, M.; Yao, H.; Zheng, X.; Yang, W. An Inkjet Printing Soft Photomask and Its Application on Organic Polymer Substrates. *Sci. China Chem.* **2010**, *53*, 1695–1704. [[CrossRef](#)]
79. Zea, M.; Texidó, R.; Villa, R.; Borrós, S.; Gabriel, G. Specially Designed Polyaniline/Polypyrrole Ink for a Fully Printed Highly Sensitive PH Microsensor. *Cite This ACS Appl. Mater. Interfaces* **2021**, *13*, 33535. [[CrossRef](#)]
80. Zhang, S.; Cao, S.; Zhang, T.; Fisher, A.; Lee, J.Y. Al<sup>3+</sup> Intercalation/de-Intercalation-Enabled Dual-Band Electrochromic Smart Windows with a High Optical Modulation, Quick Response and Long Cycle Life. *Energy Environ. Sci.* **2018**, *11*, 2884–2892. [[CrossRef](#)]
81. Lee, H.J.; Lee, C.; Song, J.; Yun, Y.J.; Jun, Y.; Ah, C.S. Electrochromic Devices Based on Ultraviolet-Cured Poly(Methyl Methacrylate) Gel Electrolytes and Their Utilisation in Smart Window Applications. *J. Mater. Chem. C* **2020**, *8*, 8747–8754. [[CrossRef](#)]
82. Cong, S.; Tian, Y.; Li, Q.; Zhao, Z.; Geng, F. Single-crystalline Tungsten Oxide Quantum Dots for Fast Pseudocapacitor and Electrochromic Applications. *Adv. Mater.* **2014**, *26*, 4260–4267. [[CrossRef](#)] [[PubMed](#)]
83. Cai, G.; Park, S.; Cheng, X.; Eh, A.L.S.; Lee, P.S. Inkjet-Printed Metal Oxide Nanoparticles on Elastomer for Strain-Adaptive Transmissive Electrochromic Energy Storage Systems. *Sci. Technol. Adv. Mater.* **2018**, *19*, 759–770. [[CrossRef](#)]
84. Small, W.R.; Masdarolomoor, F.; Wallace, G.G.; In Het Panhuis, M. Inkjet Deposition and Characterization of Transparent Conducting Electroactive Polyaniline Composite Films with a High Carbon Nanotube Loading Fraction. *J. Mater. Chem.* **2007**, *17*, 4359–4361. [[CrossRef](#)]
85. Shim, G.H.; Han, M.G.; Sharp-Norton, J.C.; Creager, S.E.; Foulger, S.H. Inkjet-Printed Electrochromic Devices Utilizing Polyaniline-Silica and Poly(3,4-Ethylenedioxythiophene)-Silica Colloidal Composite Particles. *J. Mater. Chem.* **2008**, *18*, 594–601. [[CrossRef](#)]
86. Abargues, R.; Rodríguez-Cantó, P.J.; García-Calzada, R.; Martínez-Pastor, J. Patterning of Conducting Polymers Using UV Lithography: The in-Situ Polymerization Approach. *J. Phys. Chem. C* **2012**, *116*, 17547–17553. [[CrossRef](#)]
87. Cho, J.; Shin, K.H.; Jang, J. Polyaniline Micropattern onto Flexible Substrate by Vapor Deposition Polymerization-Mediated Inkjet Printing. *Thin Solid Film.* **2010**, *518*, 5066–5070. [[CrossRef](#)]
88. Jeon, S.; Park, S.; Nam, J.; Kang, Y.; Kim, J.M. Creating Patterned Conjugated Polymer Images Using Water-Compatible Reactive Inkjet Printing. *ACS Appl. Mater. Interfaces* **2016**, *8*, 1813–1818. [[CrossRef](#)]
89. Rajzer, I.; Rom, M.; Menaszek, E.; Pasierb, P. Conductive PANI Patterns on Electrospun PCL/Gelatin Scaffolds Modified with Bioactive Particles for Bone Tissue Engineering. *Mater. Lett.* **2015**, *138*, 60–63. [[CrossRef](#)]
90. Xu, Q.; Ihalainen, P.; Smått, J.H.; Määttänen, A.; Sund, P.; Wilén, C.E.; Peltonen, J. Template-Induced Fabrication of Nanopatterned Polymeric Films by Inkjet Printing. *Appl. Surf. Sci.* **2014**, *313*, 237–242. [[CrossRef](#)]
91. Nguyen, T.V.; Van, Q.L.; Peng, S.; Dai, Z.; Ahn, S.H.; Kim, S.Y. Exploring Conducting Polymers as a Promising Alternative for Electrochromic Devices. *Adv. Mater. Technol.* **2023**, 2300474. [[CrossRef](#)]

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