

Review

Inkjet Printing for Batteries and Supercapacitors: State-of-the-Art Developments and Outlook

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Abstract

Inkjet printing enables contactless deposition onto fragile substrates for printed energy-storage devices and supports flexible batteries and supercapacitors with reduced material use. This review examines multilayer and interdigital architectures and analyzes how ink rheology, droplet formation, colloidal interactions, and the printability window govern performance. For batteries, reported inkjet-printed electrodes commonly deliver capacities of $\sim 110\text{--}150\text{ mAh g}^{-1}$ for oxide cathodes at $C/2\text{--}1\text{ C}$, with coulombic efficiency $\geq 98\%$ and stability over $10^2\text{--}10^3$ cycles; silicon anodes reach $\sim 1.0\text{--}2.0\text{ Ah g}^{-1}$ with efficiency approaching 99% under stepwise formation. Typical current densities are $\sim 0.5\text{--}5\text{ mA cm}^{-2}$ depending on areal loading, and multilayer designs with optimized drying and parameter tuning can yield rate and discharge behavior comparable to cast films. For supercapacitors, inkjet-printed microdevices report volumetric capacitances in the mid-hundreds of F cm^{-3} , translating to $\sim 9\text{--}34\text{ mWh cm}^{-3}$ and $\sim 0.25\text{--}0.41\text{ W cm}^{-3}$, with 80–95% retention after 10,000 cycles and coulombic efficiency near 99%. In solid-state configurations, stability is enhanced, although often accompanied by reduced areal capacitance. Although solids loading is lower than in screen printing, precise material placement together with thermal or photonic sintering enables competitive capacity, rate capability, and cycle life while minimizing waste. The review consolidates practical guidance on ink formulation, printability, and defect control and outlines opportunities in greener chemistries, oxidation-resistant metallic systems, and scalable high-throughput printing.



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1. Introduction

Inkjet printing is widely used in printed electronics and enables the production of portable, flexible, and potentially environmentally friendly devices [1]. This review explores how precise adjustment of printing parameters such as waveform design and electrical settings, jetting frequency, droplet spacing, printing resolution, nozzle-to-substrate gap, and substrate choice, together with ink formulations, can be harnessed to optimize both the electrochemical performance and structural integrity of inkjet-printed energy storage devices.

Batteries and supercapacitors are fundamental pillars of portable electronics and wearable technologies, and their demand will continue to grow as miniaturization, flexibility, and sustainability become critical requirements. Flexible energy-storage devices require manufacturing processes that combine high resolution, low cost, and compatibility with sensitive substrates; printed electronics, particularly inkjet printing, enable these objectives.

Recent studies on printed solid-state batteries highlight that printing techniques provide accessible, versatile, and eco-efficient methods for the fabrication of microscopic batteries and other electronic devices, while facing common challenges such as ink rheology [2], device durability [3], scalability, and material uniformity [4,5].

Alternative additive manufacturing techniques have been explored to meet different design and performance requirements. Screen printing, aerosol jet printing, and emerging 3D printing methods provide complementary capabilities, each with specific strengths and limitations. Screen printing transfers high-viscosity inks at or above 500 mPa·s through a mesh that contacts the substrate, producing nanoliter-scale deposits, relatively thick layers, minimum feature sizes near 40 µm, and high throughput [6]. Aerosol jet printing atomizes the ink and uses a sheath gas to hydrodynamically focus sub-picoliter to picoliter droplets, reaching about 10 µm features; in specialized setups with annular acoustic focusing, line widths below 6 µm are possible, at the cost of added process complexity and overspray control [7,8]. 3D printing techniques, although not the focus of this review, are gaining momentum because they can build thicker electrodes and precisely control microstructures; however, they typically require more complex equipment and remain strongly constrained by ink rheology compared with inkjet printing [9]. A comparative analysis of printing methods for MXene-based micro-supercapacitors showed that inkjet printing requires less material, offers environmentally friendly and low-cost operation, but suffers from nozzle clogging, whereas screen printing provides higher throughput at the expense of resolution and roughness [10].

In this context, inkjet printing has emerged as a particularly promising technique, as it enables the deposition of functional materials onto rigid or flexible substrates and the fabrication of electronic components such as batteries, sensors, antennas, conductive traces, RFID tags, and transistors [11,12], offering precise control over electrode patterns and geometry [13].

A characteristic of inkjet printing is its contactless deposition. The printhead remains separated from the substrate during droplet ejection [14]; this minimizes mechanical loading and local heating, protects pressure- and heat-sensitive media [15], and enables conformal patterning on irregular surfaces [16].

Additionally, this capability supports printing on glass, ceramics, wood finishes, thin metal foils, and polymer films, works with lower-viscosity inks of about 5 to 15 mPa·s, achieves feature sizes typically in the ~20 to 60 µm range with high placement accuracy, and offers straightforward digital control suited to battery and supercapacitor electrodes [17,18]. However, the nozzle–substrate separation also imposes challenges: droplet flight can be influenced by ambient airflow, and partial evaporation during flight may alter ink concentration. These issues necessitate careful control of ambient conditions and waveform design [19].

In the past two years, several milestones have demonstrated the practical maturity of inkjet printing for energy storage. Progress has concentrated on four complementary fronts: ink chemistry and printable materials, jetting control and print fidelity, device architectures with electrochemical validation, and translation toward scalable manufacturing. Together, these advances position inkjet printing as a technically robust route to flexible, thin energy devices with consistent performance.

In ink formulation and printable materials, aqueous and additive-free MXene inks have matured into reliable platforms for micro-supercapacitors and flexible current collectors, combining high conductivity and electrochemical accessibility with good jetting across practical nozzle diameters [20–23]. Hybrid and doped systems, such as NiS/N-MXene for asymmetric devices and water-based graphene or graphene-derivative inks, extend

voltage window, areal capacitance, and environmental compatibility in printed energy storage [24,25].

For batteries, inkjet-addressable dispersions for Si-based anodes, Li-rich and Ag-assisted interfaces, and MXene current collectors have enabled thin-film and flexible Li-ion cells with improved interfacial stability and more uniform current distribution [26–30]. Researchers emphasize that stable droplet formation typically occurs for inverse Ohnesorge numbers $Z \approx 1$ –14; for water-based MXene inks, researchers increased viscosity to reduce Z and avoid satellite droplets or nozzle clogging, enabling high-resolution patterns on cotton or polymer substrates [31,32]. The print quality also depends on flake size and the number of printed passes: larger $\text{Ti}_3\text{C}_2\text{T}_x$ MXene flakes yielded lower resistance, and mixing different flake sizes while optimizing the number of passes produced specific areal capacitances of 60 and 32 mF cm^{-2} at 2 and 20 mV s^{-1} , respectively [10].

In waveform optimization, droplet control, and print fidelity, tailored piezoelectric drive waveforms and predictive drop-control algorithms have expanded stable jetting over wider viscosity and elasticity ranges, reduced satellite formation, and delivered repeatable line thickness at higher carriage speeds [33,34]. Ultra-high-velocity jetting and combined rheology–waveform characterization link fluid relaxation, meniscus dynamics, and ligament breakup to practical set points, which supports robust recipe transfer across platforms [35–37]. Complementary insights into continuous-inkjet instability maps and substrate-pattern wetting provide guidelines for texture- and chemistry-directed deposition that sharpen edges and mitigate bulging and coffee-ring artifacts in functional lines [38,39].

In device architectures and electrochemical performance, inkjet printing has delivered interdigitated and multilayer micro-supercapacitors with engineered porosity and heterostructures, for example, MXene–oxide and MXene–polymer, achieving high areal and volumetric capacitances and fast rate capability on flexible supports, including textiles [22,40–42]. For batteries, inkjet-printed Si and Si-composite anodes, Ag-modified current-collector interfaces for anode-free concepts, and MXene current collectors have shown improved cyclability, lower impedance, and enhanced tolerance to deformation [26,27]. Beyond laboratory coupons, demonstrations of inkjet-printed Zn– MnO_2 planar cells and water-based electrode inks underscore progress toward safer chemistries and greener processing [43–45].

In scalability and manufacturability, advances in materials and process control are translating to larger-area, higher-density layouts. Large-area printed optoelectronics contribute registration, sintering, and throughput strategies that are transferable to energy storage. Flexible and wearable electronics provide validated pathways for adhesion, encapsulation, and mechanical reliability on soft substrates [46,47]. Before jetting, a priori printability metrics for battery dispersions, for example water-based anodes, enable faster down-selection and risk reduction prior to device trials [30], while rheology-guided green conductive inks facilitate sustainable scale-up [48].

In parallel with these advances, several bottlenecks persist. Critical parameters such as particle size distribution, viscoelastic behavior, rheological profiles, and colloidal stability must be meticulously controlled to ensure both unimpeded printer operation and the preservation of desired material characteristics after deposition. For high solids functional systems, including MXenes, oxides, silicon, and conducting polymers, attaining jettable viscosity and elasticity while maintaining electronic percolation, ion transport pathways, and robust adhesion remains non-trivial. Shear thinning responses, particle anisotropy, and surfactant selection frequently impose tradeoffs between jet stability and film performance, which complicates scale transfer and long print runs [22,23,30,48].

Droplet formation physics and nozzle health introduce additional constraints. The breakup process is highly sensitive to waveform shape, back pressure, and transient meniscus dynamics, which can trigger satellites, misting, or misfires as throughput increases. Long-term reliability continues to depend on waveform recipes with adequate stability margins and on clog-resistant formulations, especially for plate-like or chemically reactive materials that promote aggregation or edge drying at the nozzle [33,35].

Microstructure control during wetting and drying is another bottleneck with direct consequences for electrochemical performance. Uniformity across length scales, including grain or flake orientation and porosity gradients, depends on wetting hysteresis, substrate topography, and evaporation kinetics. Deviations from uniform drying promote resistive bottlenecks, heterogeneous current distribution, and local delamination during cycling, which undermines rate capability and lifetime [39,49].

Mechanical durability and interfacial integrity under realistic duty cycles remain insufficiently standardized. Under bending, twisting, and cyclic strain, printed stacks can initiate cracks at interfaces or at discontinuities in current collectors. MXene-based collectors and silver-assisted wetting have improved compliance and contact quality, yet consensus benchmarks for long-term durability on flexible substrates are still emerging and do not fully capture multiaxial fatigue or out-of-plane deformation relevant to textiles and ultrathin films [28,29].

Standardization and metrology further limit cross-platform reproducibility. Proprietary implementations of waveform generation and differences in drop watch imaging and calibration hinder recipe portability and data sharing. Rigorous, shareable datasets and benchmark protocols tailored to energy device-grade inks are in a formative stage, which slows down comparative evaluation and technology transfer across laboratories and toolsets [38,50].

This review focuses specifically on batteries and supercapacitors because both are key energy storage devices whose demand continues to grow, particularly in markets for portable and wearable electronics. These devices share common operational and material requirements, and inkjet printing offers distinct advantages for their fabrication, including high-resolution patterning, low-temperature processing, and compatibility with flexible substrates. Unlike other reviews that either address multiple device types without a dedicated focus or discuss inkjet printing in a broad context without targeting specific applications, this work integrates both perspectives into a single, compact source. It critically examines the evolution of inkjet printing for these two classes of devices, consolidating advances in materials, rheology, waveform optimization, and device architecture into a chronological framework. This approach provides readers with a coherent, application-focused reference that links technical parameters to electrochemical performance, fulfilling a need for a comprehensive resource on inkjet printing for high-demand energy storage technologies.

To ensure a comprehensive and unbiased overview, this review was conducted through a structured literature search covering the period from 2015 to May 2025. Sources were collected from Google Scholar, the SOFIA portal of Université du Québec à Trois-Rivières, and Research Rabbit, with major publishers such as Elsevier, Wiley, Springer, MDPI, ACS, and IEEE being included. Search queries combined terms such as “inkjet printing”, “batteries”, “supercapacitors”, “flexible”, and “energy storage” to capture both fundamental and applied works. Only peer-reviewed journal articles and conference proceedings in English were retained, while studies not directly addressing inkjet printing of energy storage devices were excluded. Seminal contributions predating 2015 were included selectively when necessary to contextualize droplet formation, rheology, and waveform fundamentals. This methodology yielded a representative corpus of studies that underpin

the chronological analysis of materials, processes, and device performance presented in the following sections.

2. Inkjet Printing

Inkjet systems come in different types based on backend technology, drop-ejection mechanisms, printhead architectures, and application areas. Two well-known types are Continuous Inkjet (CIJ) and Drop-on-Demand (DoD) (Figure 1). CIJ systems produce a continuous stream of ink droplets that are electrically charged and subsequently deflected to form the desired printing pattern [51]. Any droplets not required for printing are then recirculated back into the ink reservoir. On the other hand, DoD systems, create drops only when needed from 25 to 120 μm droplets at a rate of 0–2000 drops per second, using thermal or piezo-electric actuators to control ink ejection [34,52,53].

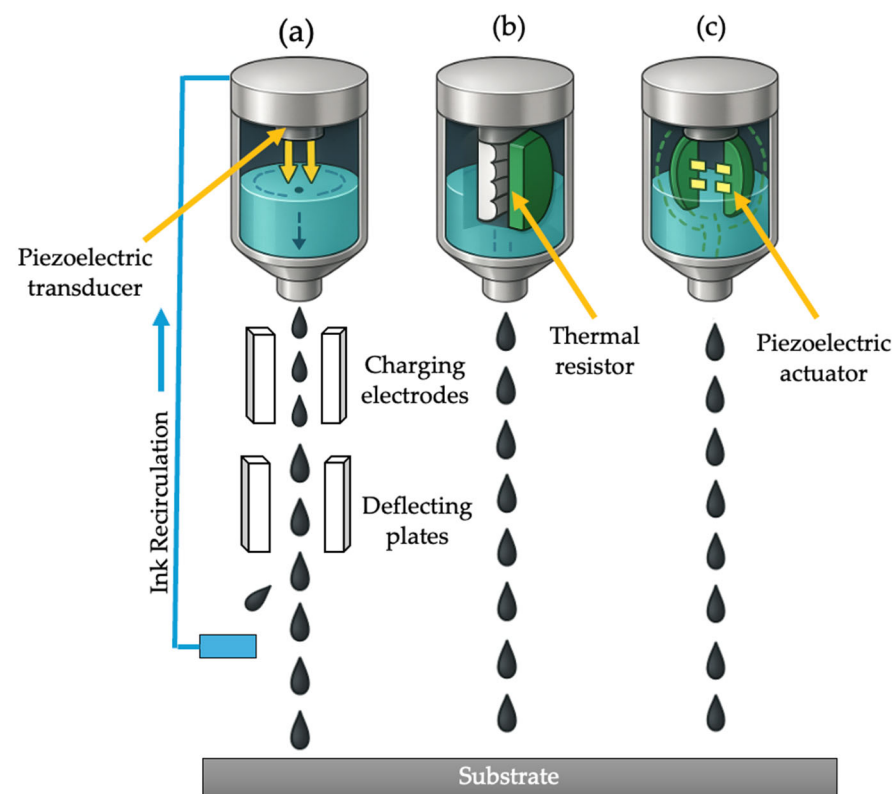


Figure 1. Schematic representation of the operating principles of (a) Continuous Inkjet (CIJ) with piezoelectric transducer, charging electrodes, deflecting plates, and ink recirculation; (b) Thermal Inkjet (TIJ) with resistive heating element; and (c) Piezoelectric Inkjet (PIJ) with piezoelectric actuator. The diagram highlights the internal actuation mechanism, droplet path, and substrate deposition. Redrawn and adapted from Ref. [1]. Copyright (6098420614292).

In CIJ mode, constant pressure is applied to the fluid reservoir, leading to the ejection of a pressurized column from the nozzle. This jet breaks into droplets due to Rayleigh instability, which is a physical phenomenon where perturbations in the flow cause the formation of smaller droplets [38]. Droplet formation can be optimized by vibrating, disturbing, or modulating the jet at a frequency close to the spontaneous droplet formation rate, thus synchronizing with the forced vibration and ejecting uniform mass ink droplets [54]. This control is crucial in inkjet printing, where droplets are produced at consistent intervals and their deposition must be carefully controlled after their separation from the jet.

Additionally, CIJ deposition offers a high throughput rate, making it widely used in applications such as food and pharmaceutical labeling. The materials used must be capable of carrying a charge, and the fluid diverted to the collector must be either discarded

or reprocessed, which can pose challenges when the fluid is expensive or when waste management is problematic. In terms of droplet formation, commercially available systems typically produce droplets of about 150 μm in diameter at a rate of 80–100 kHz, although frequencies up to 1 MHz and droplet sizes ranging from 6 μm (10 fL) to 1 mm (0.5 μL) have also been reported [55].

Thermal inkjet technology (TIJ) uses a heated resistor to create bubbles that force ink out of the nozzle; the print head design meticulously controls where and how the ink is released [47]. The ink chamber is precisely engineered, including a small orifice at the nozzle tip. When the resistor inside the chamber heats up, the solvent in the ink rapidly vaporizes, forming a bubble. This steam bubble exerts pressure on the ink, forcing it through the nozzle orifice. Both the pressure and volume are finely regulated to prevent excessive ink evaporation or the creation of excessive pressure that could damage the print head. On the other hand, piezoelectric inkjet systems (PIJ) utilize piezoelectric materials embedded within the print head that deform upon voltage application, changing their shape [56].

This deformation generates pressure waves, which, without the need for heat, eject ink droplets from the nozzle. This method prevents ink degradation associated with heat exposure and enables the use of a broader range of inks, including those sensitive to high temperatures. Consequently, piezoelectric systems enable precise control and broad compatibility across various printing applications, supporting accuracy and the ability to handle diverse ink types [36].

Each method has benefits and limitations. Thermal inkjet has evolved beyond desktop devices and now powers high-volume industrial presses such as the HP Page Wide T1195i (HP Inc., Palo Alto, CA, USA) for corrugated preprint, with a web width of 2.8 m, speeds up to 305 m/min, native resolution of 1200 nozzles per inch, and six-color aqueous pigment sets [57]. These systems show that TIJ can support continuous single-pass production at industrial scale. Even so, the heat-driven ejection mechanism narrows the range of compatible chemistries and entails specific maintenance and durability regimes for the printhead [58]. Piezoelectric inkjet offers broader ink latitude, including UV, solvent and high-viscosity aqueous formulations, and is widely adopted in high-volume platforms that use thin-film piezo heads with ink recirculation [37]. Representative implementations include industrial-scale systems such as the RICOH Pro Z75 (Ricoh Company, Ltd., Tokyo, Japan), sheet-fed aqueous inkjet press with stainless steel piezo printheads delivering 1200 dpi at up to 4500 sheets per hour, and the EFI Nozomi C18000 (Electronics For Imaging, Inc., Fremont, CA, USA) corrugated press with 1.8 m sheet width for high-volume single-pass packaging production [59,60]. In press practice, throughput is governed primarily by nozzle count, web width and single-pass architecture, while the actuator type chiefly determines drop formation control, ink compatibility and head lifetime [37].

A recent development in inkjet technology is Super-fine Inkjet (SIJ), which uses an electrostatic process where electrostatic forces are pivotal in manipulating the formation and size of ink droplets. Initially, the ink is electrically charged within the print head without the need for mechanical components. These charged droplets are then expelled through a controlled electrostatic field, which adjusts both the trajectory and the speed of the droplets, enabling precise positioning on the substrate [61]. This control allows SIJ to produce droplets that can be smaller, and more uniform compared to traditional inkjet technologies [62]. Figure 1 shows each of the classifications and components.

Ejection control is achieved by applying varying levels of charge to the droplets, allowing for the manipulation of their behavior upon expulsion from the print head. Droplets with a higher charge are repelled more strongly by the electrostatic fields, facilitating the controlled disintegration of a larger droplet into smaller droplets. Additionally, the charge directly influences how and where the droplets are deposited on the substrate, enabling them to be directed with high precision to specific locations, thereby improving the quality and resolution of the printed pattern. This ability to finely adjust the size and placement of each droplet results in high-resolution prints with well-defined lines and accurate features. This level of control is useful in applications where fine details are crucial, such as in the printing of electronic circuits and detailed reproductions of photographic and artistic images. Collectively, these features indicate that SIJ is a printing approach with electrostatic droplet control suitable for a range of industrial and artistic applications [61].

Inkjet printing technology is used beyond small-scale applications to encompass large-format printing on textiles, posters, and more, handling materials up to 5 m wide and producing over $120 \text{ m}^2 \text{ h}^{-1}$ [1]. Additionally, it is commonly used for variable data printing for labels and codes on irregular shapes, and it integrates with traditional printing techniques for mass production.

The evolution of inkjet printing technology from traditional graphics applications to advanced industrial manufacturing has enabled applications for battery and supercapacitor electrode fabrication. In energy storage applications, inkjet printing offers advantages, including precise material deposition, contactless processing, and reduced material waste, making it well-suited for manufacturing thin-film electrodes and customized energy storage devices [63,64]. Table 1 shows a comparative analysis that examines how inkjet printing compares to other manufacturing techniques for energy storage applications, highlighting the critical trade-offs between resolution, solid loading, and production throughput that influence technique selection.

For battery and supercapacitor designers, the analysis of various printing techniques reveals three critical trade-offs that directly impact inkjet printing's positioning in the energy storage manufacturing landscape.

First, high-viscosity, high-solid-loading inks are most effectively handled by screen-printing and extrusion-based methods, which can accommodate solid loadings up to 80 wt% while maintaining printability. In contrast, inkjet printing is fundamentally limited to lower solid loadings (typically 5–15 wt%), requiring it to achieve competitive performance through alternative strategies such as multilayer printing and precise material placement. A recent study demonstrated that while screen-printed LiFePO_4 cathodes with 65 wt% solid loading achieved areal capacities of 5.2 mAh cm^{-2} , inkjet-printed LiFePO_4 electrodes with optimized multilayer deposition reached 4.8 mAh cm^{-2} through 20 printing passes, albeit with significantly reduced throughput [65].

Second, fine feature sizes below $20 \text{ }\mu\text{m}$ represent a key advantage of inkjet printing in energy storage applications. While conventional drop-on-demand inkjet systems achieve resolutions of 5–50 μm , aerosol-jet printing excels with features down to $10 \text{ }\mu\text{m}$, and Super-fine Inkjet (SIJ) technology pushes this boundary to sub-micrometer scales (0.5–10 μm). This high-resolution capability enables the fabrication of microelectrodes and interdigitated structures that maximize surface area while minimizing ionic transport distances. Recent work using SIJ technology achieved printed conductive lines as narrow as $3 \text{ }\mu\text{m}$ for silver ink applications, illustrating feasibility for miniaturized energy storage device. However, these high-resolution inkjet techniques typically sacrifice throughput and solid loading capacity compared to conventional manufacturing methods.

Table 1. Technical Comparison of Printing Methods for Battery and Supercapacitor Electrode Manufacturing.

Printing Technique	Key Advantages	Main Limitations	Typical Printing Parameters	Battery Parameters	Supercapacitor Parameters
Drop-on-Demand Inkjet [64,66]	High resolution (5–50 μm), accurate material deposition, low material waste	Limited to low solid loading (5–15 wt%), requires Ohnesorge number (Z) within 1–14	Viscosity: 1–20 mPa·s; Throughput: 0.1–2 mL/min; X-Y Resolution: 10–50 μm ; Z Resolution: 0.1–10 μm	Capacity 300–2500 mAh g ^{−1} ; current 0.2–1 A g ^{−1} ; energy 94–330 Wh kg ^{−1} ; power 220–1650 W kg ^{−1} ; cycle life 200–500 cycles; coulombic efficiency $\approx$ 99% [43,67]	Areal capacitance 22–29 mF cm ^{−2} ; mass capacitance 829–1294 F g ^{−1} ; volumetric capacitance 608–746 F cm ^{−3} ; cycle life > 1000 cycles [68]
Super-fine Inkjet (SIJ) [1,62]	Ultra-high resolution (0.5–10 μm), broad viscosity range (0.5–10,000 mPa·s), femtoliter droplets	Complex setup; limited commercial availability; lower throughput	Viscosity: 0.5–10,000 mPa·s; Droplet volume: 0.1 fL–10 pL; X-Y Resolution: 0.5–10 μm ; Z Resolution: 0.1–1 μm	Zn–Ag 3D microbattery: ~60% higher capacity than planar; current $\approx$ 1.1 mA cm ^{−2} [69]	3D micro-SC: areal capacitance 11–14 mF cm ^{−2} ; capacitance retention ~ 80% after 2000 cycles [70]
Aerosol-Jet Printing [71]	Broad viscosity tolerance (1–1000 mPa·s), high precision (10–250 μm), compatible with non-planar substrates	Complex instrumentation; limited to thin films; moderate throughput	Viscosity: 1–1000 mPa·s; Solid loading: 10–40% wt%; X-Y Resolution: 10–250 μm ; Z Resolution: 0.1–5 μm	Capacity $\approx$ 75–100 mAh g ^{−1} ; current $\approx$ 50–150 mA g ^{−1} (C/2–1C); cycle life >100 cycles [72]	MXene micro-SC: areal capacitance ~ 122 mF cm ^{−2} ; volumetric capacitance ~ 611 F cm ^{−3} [73]
Screen Printing [74,75]	High solid loading capacity (50–80 wt%), ideal for thick layers, scalable manufacturing	Limited resolution (>50 μm); requires specific rheological tuning	Viscosity: 1000–100,000 mPa·s; Throughput: 100–1000 cm ² /min; X-Y Resolution: >50 μm ; Z resolution (per-pass layer thickness): 10–50 μm , up to ~100 μm for thick films.	Capacity 120–170 mAh g ^{−1} at 2–6C; current 0.7–2.2 A g ^{−1} ; cycle life > 100 cycles [76]	Areal capacitance 200–250 mF cm ^{−2} ; cycle life > 5000 cycles [77]
Gravure Printing [78]	High-speed roll-to-roll processing (~400 m/min), excellent film uniformity, scalable	High initial cost; limited to thin films; handling of complex inks can be challenging	Viscosity: 50–500 mPa·s; X-Y Resolution: 2–100 μm ; Z Resolution: 1–50 μm	Na-ion battery anodes: initial capacity 400–440 mAh g ^{−1} ; stabilized ~ 120 mAh g ^{−1} after 100 cycles	MoS ₂ @S-rGO micro-SC: areal capacitance 6.6 mF cm ^{−2} ; energy density 0.58 mWh cm ^{−3} ; power density 13.4 mW cm ^{−3} ; cycle life > 1000 cycles

Table 1. Cont.

Printing Technique	Key Advantages	Main Limitations	Typical Printing Parameters	Battery Parameters	Supercapacitor Parameters
Extrusion Printing [79,80]	Very high viscosity inks (1000–10,000 cP), 3D structures, high loading	Slow printing speed, limited resolution (>100 µm)	Viscosity: 1000–50,000 mPa·s, Solid loading: 60–90 wt%, X-Y Resolution: >100 µm; Z Resolution: 50–500 µm	Capacity 30–90 mAh g ^{−1} ; energy density ~ 110 Wh kg ^{−1} ; cycle life > 100 cycles [81]	Planar rGO micro-supercapacitor: volumetric capacitance ≈ 41.8 F cm ^{−3} ; areal energy density ≈ 7.6 mWh cm ^{−2} ; areal power density ≈ 29.2 mW cm ^{−2} [82]
Continuous Inkjet [50]	High-speed printing (80–100 kHz), suitable for large-area coverage	Compatible only with conductive inks; high material waste; low resolution X-Y (>100 µm)	Viscosity: 1–10 mPa·s; Throughput: 10–50 mL/min; X-Y Resolution: >100 µm; Z Resolution: N/A (continuous)	N/A	N/A

Third, the optimal trade-off between resolution, loading, and speed defines inkjet printing as a technically advantageous approach within the field of energy storage device fabrication. Unlike screen-printing, which dominates commercial supercapacitor electrode fabrication with 50–100 μm features and >60 wt% solid loadings at industrial throughput rates, inkjet printing offers precision and material flexibility at the expense of manufacturing speed. The technology's strength lies in applications requiring precise material placement, customized geometries, and reduced material waste rather than high-volume production. For example, inkjet-printed flexible micro-supercapacitors demonstrate areal capacitances of 46.6 mF cm^{-2} while maintaining 86.8% capacity retention after 1000 bending cycles at 180° performance levels that would be challenging to achieve with conventional manufacturing methods [83]. Similarly, aerosol-jet printed MXene micro-supercapacitors achieve volumetric capacitances of 611 F cm^{-3} with precise control over electrode geometry and thickness [84], illustrating inkjet printing's utility for specialized energy storage applications where precision and customization outweigh throughput considerations.

Subsequent sections delve into droplet-formation mechanisms and waveform control in inkjet systems, explore ink-formulation strategies tailored to energy storage, and present a chronological critical analysis of inkjet-printed batteries and supercapacitors.

3. Ink Rheology

The formulation of functional battery inks relies on colloidal science principles that govern particle interactions, stability, and flow behavior under the short-timescale, high-shear conditions encountered during inkjet printing. This subsection outlines links between ink composition to rheological properties and printability parameters, with emphasis on the relationship between formulation variables and the dimensionless Z number that defines the inkjet printing window.

3.1. Colloidal Fundamentals for Ink Formulation

Particles experience van der Waals attraction; without stabilization they aggregate and sediment. Electrostatic stabilization uses surface charges to generate repulsive double layers; a zeta potential $|\zeta| \gtrsim 30 \text{ mV}$ is generally sufficient for colloidal stability [85,86]. In battery inks the ionic strength is high and many materials (graphite, LiFePO_4 , MXenes) have low surface charge, so electrostatics alone is often inadequate. Steric stabilization solves this by adsorbing polymer chains onto particle surfaces. Polymers such as carboxymethyl cellulose (CMC), poly(acrylic acid) (PAA), poly(vinylpyrrolidone) (PVP), poly(vinylidene fluoride) (PVDF) and the conducting polymer PEDOT:PSS form a solvated layer (typically 5–10 nm thick) that reduces van der Waals attraction and prevents close approach [87,88]. Dispersant choice depends on particle chemistry, for example: CMC and PAA adsorb to carbon black and LiFePO_4 via hydrogen bonding, PVP interacts through polar interactions, and PEDOT:PSS provides both steric stabilization and electrical conductivity [89,90].

Aggregates promote sedimentation, giving non-uniform solids content and nozzle clogging. For dilute suspensions, the settling velocity obeys to Stokes-type law,

$$v_{\text{sed}} = \frac{2}{9} \frac{\Delta\rho g r^2}{\eta} \quad (1)$$

where $\Delta\rho$ is the density difference between particles and solvent, g is the gravitational acceleration, r is the particle radius, and η is the continuous-phase viscosity [91].

Suppression of sedimentation can be achieved through several approaches: reducing particle size (nano-scale carbon additives), increasing continuous phase viscosity η through polymer addition such as ethylene glycol or glycerol, density matching between phases, or forming weak reversible networks with fumed silica or cellulose nanofibrils that create

a yield stress sufficient to suspend particles without impeding jetting [92,93]. However, these strategies must be balanced against printability requirements, as excessive viscosity or network strength can prevent proper droplet formation. The critical balance involves maintaining sufficient stability during storage while ensuring the network breaks down under the shear conditions in the printhead.

Maintaining viscosity within the jettable window is only one aspect of ensuring print quality; surface tension control is equally critical for stable droplet formation and substrate wetting. Surfactants lower surface tension and control wetting on substrates. They can be anionic, for example sodium dodecyl sulfate SDS, cationic such as cetyltrimethylammonium bromide CTAB, non-ionic like Triton X-100 or Tween 20, or fluorosurfactants such as Zonyl. Their interaction with polymers depends on charge; SDS strongly binds to methyl cellulose, while CTAB interacts hydrophobically [94].

Dynamic surface tension at millisecond timescales governs drop formation and wetting behavior during printing. Unlike equilibrium surface tension measurements, dynamic measurements at surface ages of 5–100 ms reveal the actual wetting behavior during the rapid processes of drop ejection and substrate impact. Optimal dynamic surface tension values for battery inks typically range from 30 to 45 mN/m at 10–100 ms timescales [95]. The dosing strategy involves adding surfactants near or below the critical micelle concentration (CMC) to balance effective surface tension reduction with minimal impact on ionic conductivity and electrochemical compatibility. Excessive surfactant concentration can lead to foam formation, residual contamination affecting electrode performance, and interference with particle-binder interactions [96].

Viscosity η controls both jetting and film formation. At low particle volume fraction ϕ , the relative viscosity $\eta_r = \eta/\eta_0$ of hard-sphere suspensions follows the Einstein–Batchelor expansion:

$$\frac{\eta}{\eta_0} \approx 1 + 2.5\phi + 6.2\phi^2 \quad (2)$$

where η_0 is the viscosity of the pure solvent. The linear coefficient equals the intrinsic viscosity $[\eta]$, which is $[\eta] \approx 2.5$ for monodisperse spheres. More generally, one may write $\eta_r \approx 1 + [\eta]\phi + \kappa_H\phi^2$, where κ_H , captures weak hydrodynamic pair interactions [97].

At higher ϕ , many-body hydrodynamic and excluded-volume interactions cause rapid thickening. The divergence as $\phi \rightarrow \phi_m$ is described by the Krieger–Dougherty model:

$$\frac{\eta}{\eta_0} = \left(1 - \frac{\phi}{\phi_m}\right)^{-[\eta]\phi_m} \quad (3)$$

where $[\eta]$ is the intrinsic viscosity and ϕ_m is the maximum packing fraction. For randomly packed monodisperse spheres $\phi_m \approx 0.64$ (often 0.58–0.64 depending on protocol). Polydispersity in (approximately) spherical particles typically increases ϕ_m and can lower η_r at a fixed ϕ . In contrast, high-aspect-ratio flakes and rods (e.g., graphene and carbon nanotubes) markedly increase $[\eta]$ and often decrease ϕ_m , which strongly raises η_r at a given solids loading [97].

Beyond controlling surface tension, polymeric binders (PVDF, CMC/SBR, PAA, PVP, and PEDOT:PSS) are essential for film integrity and adhesion, providing structural cohesion to the printed layer. Most exhibit shear-thinning at $\sim 10^3$ – 10^4 s^{−1} during jet ejection, which supports droplet formation while preserving a higher on-substrate viscosity to limit spreading; for example, PEDOT:PSS inks decrease from ~ 14 mPa·s at low shear to ~ 7 mPa·s at high shear [98]. Excess binder or very high molecular weight introduces elasticity that promotes long ligaments and satellite droplets, whereas insufficient binder yields fragile films. Typical printhead windows are 5–15 mPa·s viscosity, 30–45 mN m^{−1} surface tension, and ~ 1.0 – 1.2 g cm^{−3} density [98]. PVDF performance depends on solvent

(e.g., N-methyl-2-pyrrolidone), molecular weight, and coupling agents; high viscosities or reactions with lithium salts can cause swelling or degrade conductivity [96]. Dispersants and surfactants must also be electrochemically compatible: residual anionic surfactants (e.g., SDS) can drive first-cycle side reactions, inflate initial capacity, and reduce coulombic efficiency; therefore, selection and removal protocols should minimize residual films that harm long-term performance [99].

Collectively, these formulation choices set the high-shear viscosity and the millisecond-scale surface tension experienced at the nozzle. To translate composition into printability, the Ohnesorge number and its inverse Z provide the framework that delineates the jettable window and the defect modes they suppress.

3.2. Printability Window and Defect Control

The performance requirements for ink in inkjet printing primarily revolve around its viscosity and surface tension. It has been found that effective inkjet printing can be achieved by monitoring the Ohnesorge number, denoted as Oh . This dimensionless number is utilized to characterize the significance of viscous forces relative to inertial and surface forces in processes such as inkjet printing. The formula to calculate the Ohnesorge number is as follows [100]:

$$Re = \frac{v\rho a}{\eta} \quad (4)$$

$$We = \frac{v^2\rho a}{\gamma} \quad (5)$$

$$Oh = \frac{\sqrt{We}}{Re} = \frac{\eta}{\sqrt{\gamma\rho l}} \quad (6)$$

where η is the fluid's viscosity, ρ is the fluid's density, γ is the surface tension of the fluid and l is the characteristic diameter of the injector or nozzle. In the early stages of research on Drop-on-Demand (DOD) printing technology, significant contributions were made by Fromm and later by Reis & Derby, focusing on the dynamics of droplet formation. Fromm's pivotal work centered on the Ohnesorge Number, which he identified as a critical parameter for characterizing the fluid dynamics involved in droplet formation. He utilized the inverse of this number, designated as $Z = 1/Oh$, to develop a criterion for stable droplet generation, proposing that $Z > 2$ ensures stable droplet formation without excessive satellite droplet creation or other instabilities [101].

Building on Fromm's foundational research, Reis & Derby employed numerical simulations to further refine and expand the understanding of this model. Their studies suggested a more detailed range for the Z parameter, establishing that stable droplet formation is best achieved when Z falls between 1 and 10. This range indicates a balanced interplay between fluid viscosity, surface tension, and inertia, which are critical for producing uniform and predictable droplets essential for high-quality DOD printing. Their work helped clarify the optimal conditions under which DOD printers operate, providing a more nuanced understanding that aids in the design and operation of these systems. Within this window, defects such as satellites and coffee-ring are minimized, supporting uniform, predictable droplets and high-quality DOD printing.

Satellite Formation occurs when small secondary droplets detach from the main drop during printing, leading to unintended prints or spots on the substrate, thereby degrading the quality of the printed pattern. The Coffee-Ring Effect refers to an irregular pattern that can form when a liquid drop dries on a surface, leaving a dark ring at the edge [93]. This phenomenon occurs because the particles within the drop are dragged towards the edge as the solvent evaporates, which can result in irregular and uneven edges in the printed images or patterns.

These phenomena are particularly problematic in high-precision applications, such as electronic circuit manufacturing, where the exact placement and quality of each drop are critical to the performance of the final device. Therefore, controlling viscosity, surface tension, and other ink properties through the proper regulation of the Ohnesorge number helps mitigate these effects and improve print quality and precision.

Figure 2a provides a detailed visualization of the evolution of a droplet's diameter after impacting a substrate, where the initial velocity V_i and diameter D_i set the stage for the dynamic interactions that follow. The yellow line depicts how the diameter changes over time, stabilizing eventually due to capillary forces. These forces stem from two critical types of molecular interactions: cohesion and adhesion. Cohesion refers to the attraction among the molecules within the liquid itself, while adhesion describes the attraction between the liquid's molecules and those of the substrate. These molecular forces underpin the phenomenon of capillarity, which dictates how a liquid behaves when in contact with a solid surface. If the adhesive forces between the liquid and the solid are stronger than the cohesive forces within the liquid, the liquid tends to spread across the surface. Conversely, if cohesive forces within the liquid are stronger, the liquid contracts and forms more compact droplets.

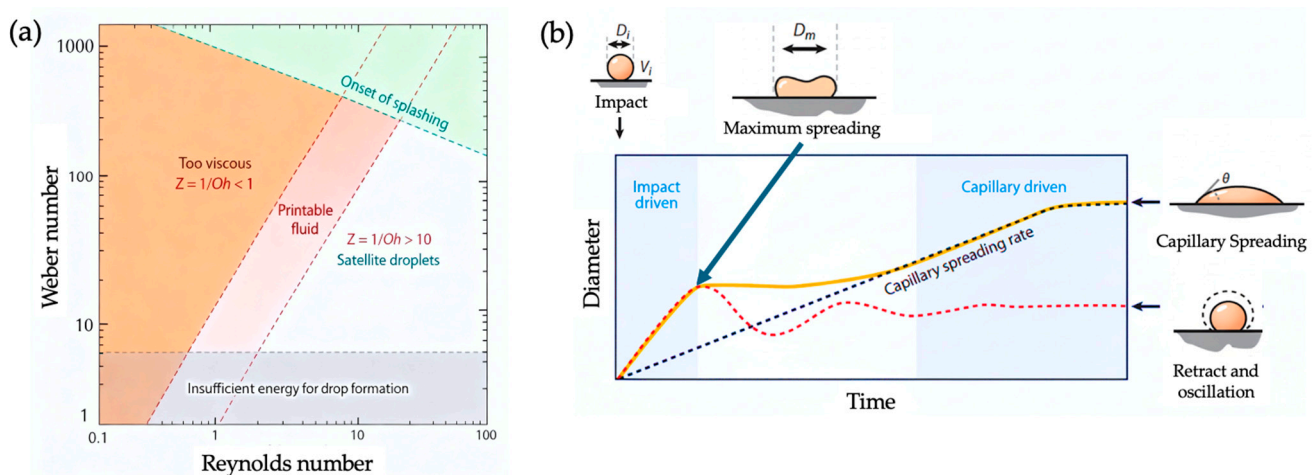


Figure 2. (a) Printable-fluid window in Reynolds–Weber coordinates illustrating the $Z = 1/Oh$ stability range for DOD inkjet printing. (b) Sequence of events following droplet impact on a substrate, showing impact-driven spreading, relaxation oscillations, and capillary-driven spreading. Redrawn and adapted from Ref. [100].

Figure 2b depicts the temporal evolution of a droplet after impact on the substrate. The initial velocity V_i and the initial diameter D_i set the inertial stage of the event. The yellow curve tracks the droplet diameter through time: it rises rapidly to a maximum spread D_m during inertia-dominated spreading, then slows as capillary forces take control and the footprint approaches a steady value. The dashed black trace indicates the capillary spreading rate. The dotted red trace shows relaxation and oscillation as viscous dissipation damps the motion. The contact angle θ summarizes wetting behavior. Small θ signals strong wetting and lateral spreading, whereas large θ produces compact droplets with steeper edges. Together, V_i , D_i , D_m and θ , along with the three traces, summarize the sequence from impact-driven spreading to capillary adjustment and finally viscous relaxation to equilibrium [100].

In practice, maintaining the ink's viscosity and surface tension within this stability window is important for reliable drop formation and deposition [102]. Staying inside this range minimizes satellite droplets and coffee-ring effects, leading to uniform, high-resolution

electrode patterns. For battery and supercapacitor inks, tuning η to 5–15 mPa·s [35] and γ to 25–45 mN/m [103] helps balance printability, device performance, and throughput.

The two panels delineate operating conditions for inkjet deposition of electrodes. Droplet generation should remain within the printability band by adjusting viscosity and surface tension, and by selecting the drive waveform, jet velocity, and nozzle diameter to suppress satellites and splashing. At the shear rates present in the nozzle, the ink viscosity should remain effectively constant and within the process window. The dispersed solids should be substantially smaller than the nozzle diameter to prevent clogging. On the substrate side, moderate contact angles achieved through surface preparation or controlled surfactant dosing promote sufficient spreading for coverage without edge roughening.

Control of the post-impact regime is also required. Drop spacing should be set as a function of the measured maximum spread D_m so that neighboring drops coalesce uniformly without flooding. Evaporation should be balanced using solvent blends, mild substrate heating, and brief between-pass drying to limit radial particle transport and suppress coffee-ring deposition. These practices yield films with uniform thickness, strong adhesion, and smooth line edges. In current collectors, this lowers sheet resistance and improves mechanical robustness; in active layers, it enhances pore connectivity and ionic accessibility, reducing interfacial and transport losses. The result is more consistent electrochemical performance in inkjet-printed batteries and supercapacitors.

In practical terms, viscosity and surface tension jointly govern print quality, electrode uniformity, and device performance. Higher viscosity at ejection, combined with shear-thinning behavior, stabilizes jetting and yields uniform line width; excessive viscosity or elasticity, however, produces ligaments and satellite droplets that roughen edges and create local thickness spikes. Lower surface tension improves wetting and coalescence between passes, which reduces pinholes; however, too low surface tension can cause overspreading, line merging, and loss of feature fidelity. When wetting and spreading are balanced, solids distribute uniformly, and porosity is even across the film. In current collectors this lowers sheet resistance and improves mechanical robustness; in active layers it enhances pore connectivity and ionic accessibility. Coffee-ring deposition creates binder-rich rims and porous centers, increasing tortuosity and contact resistance, which degrade specific capacity or capacitance and accelerate performance fade. Residual surfactants can promote parasitic reactions and reduce first-cycle coulombic efficiency in batteries, so their dosage and removal must be carefully controlled. With the printable window in place, attention turns to waveform design and its impact on droplet ejection, satellites, and electrode porosity.

4. Voltage Waveforms in Piezoelectric Drop-on-Demand Inkjet Printing

This section applies to piezoelectric drop-on-demand (PIJ) printheads. Thermal inkjet (TIJ), continuous inkjet (CIJ), and super-fine inkjet (SIJ) use different actuation principles and are not governed by the bipolar/unipolar voltage sequences described here.

A key aspect of piezoelectric drop-on-demand inkjet printing is the precise control of droplet formation by the voltage waveform applied to the piezoelectric actuator. These pulses not only determine the timing of ink ejection but also directly impact the volume and velocity of each droplet. Additionally, they play a crucial role in avoiding printing defects such as satellite droplets and ligaments, which otherwise compromise the sharpness and uniformity of the printed patterns. Hence, clearly selecting and designing voltage pulses is fundamental to achieving high-quality printing and operational efficiency across various applications.

Before describing the specific pulse types utilized in inkjet printing, it is useful to outline their general influence on droplet ejection. During dead periods (when no voltage is applied), ink remains at rest in the nozzle, allowing residual oscillations of the meniscus to stabilize, thus preventing the formation of unwanted droplets. In PIJ, when a positive voltage is applied, it generates outward pressure that forces the ink through the nozzle, forming the primary droplet. Both the magnitude and duration of this voltage pulse directly affect the droplet's volume and velocity; however, if these parameters are not carefully controlled, meniscus oscillations can cause satellite droplets to form.

Conversely, applying a negative voltage generates inward pressure, pulling ink back into the nozzle and suppressing residual vibrations. This prevents unwanted ligaments or satellite droplets, ensuring only a single, well-defined primary droplet is ejected. Specifically, bipolar pulses which alternate between positive and negative voltages combine these beneficial effects to achieve finer control over fluid dynamics, ultimately improving the precision of ink ejection.

To reflect this, Figure 3a presents the unipolar M-shaped pulse, characterized by a single square voltage pulse [104]. This pulse begins with a rapid rise in voltage to a maximum level V_1 , followed by a maintenance period at t_2 , and a final drop to zero at t_3 . This simple design ejects ink droplets by generating sufficient pressure to force the ink out of the nozzle. However, the simplicity of this pulse can lead to the formation of satellite droplets, especially in low-viscosity fluids, thus affecting the accuracy and quality of the print [33,105].

In Figure 3b, the unipolar M-shaped pulse is shown, consisting of two voltage pulses with an intermediate pause [106]. The first pulse, with a rapid rise to V_1 , resembles the single pulse and is responsible for the initial droplet ejection. Following this, a second pulse of lower amplitude exhibits similar rise and fall times but shorter duration. Lumped-element modeling of recirculating printheads, preferred for high-solid battery ink, demonstrates that inserting this second pulse with a dwell of $\approx 4 \mu\text{s}$ and an amplitude $\approx 1/3$ of the main pulse damps residual pressure in $<6 \mu\text{s}$, enabling higher firing frequencies essential for industrial throughput [105]. This second pulse helps to suppress residual oscillations and satellite droplet formation, stabilizing the ink ejection and thereby improving print quality [107]. This type of pulse is particularly useful when a reduction in droplet volume is required.

Figure 3c depicts the bipolar M-shaped pulse, which differs from the unipolar version by the polarity inversion between the pulses. The initial negative pulse, followed by a positive one, enables more precise control over droplet volume. This polarity inversion is useful for counteracting capillary forces that cause oscillations in the nozzle meniscus, thereby minimizing droplet size and enhancing printing precision. This pulse type is suitable for applications requiring precise droplet size control [108].

Similarly, Figure 3d describes the unipolar W-shaped pulse, which includes three voltage segments. It starts with a rise to V_1 , followed by a low-voltage period t_2 , a second rise to V_2 , and a drop to V_3 . This design offers detailed control over the ink dynamics, effectively reducing droplet volume. By adjusting the time t_2 , satellite droplet formation can be suppressed, ensuring greater precision in ink deposition and minimizing unwanted dispersion [109].

Extending this strategy, Figure 3e introduces the bipolar W-shaped pulse, structurally like the unipolar version but incorporating polarity changes. It begins with a negative voltage, followed by a pause and a positive voltage, before returning to a negative level and finally to zero. This configuration helps prevent the formation of ink ligaments, structures that can disrupt print quality. By alternating the polarity, the separation of the primary droplet is enhanced, resulting in greater definition and precision of the printed dot.

Finally, Figure 3f and Figure 3g illustrate the bipolar-1 and bipolar-2 N-shaped pulses, respectively. Both feature a sequence of alternating positive and negative polarities. For the bipolar-1 pulse, the initial positive pulse is followed by a negative one, useful for ejecting high-viscosity inks due to the significant voltage differential. The bipolar-2 pulse follows a similar pattern but offers additional control over suppressing residual vibrations, ensuring that the ink chamber pressure reaches an optimal level for uniform droplet ejection. These pulses are used in applications requiring precise ink management and minimizing print defects.

Precise waveform design selecting the appropriate pulse shape, polarity, amplitude, and timing significantly influences droplet uniformity and electrode porosity in printed batteries. While industrial PIJ heads typically operate in the 15–30 V, 5–20 μs envelope [110,111], machine learning searches have shown that broader spaces (10–80 V; 2–60 μs) can be safely explored to find satellite-free solutions even for challenging high-viscosity formulations [33]. When these electrical guidelines are combined with inks that satisfy the printability window, waveform optimization reduces trial-and-error, can lower surface roughness and pore clogging, and yields denser, more uniform battery electrodes at comparable production speeds.

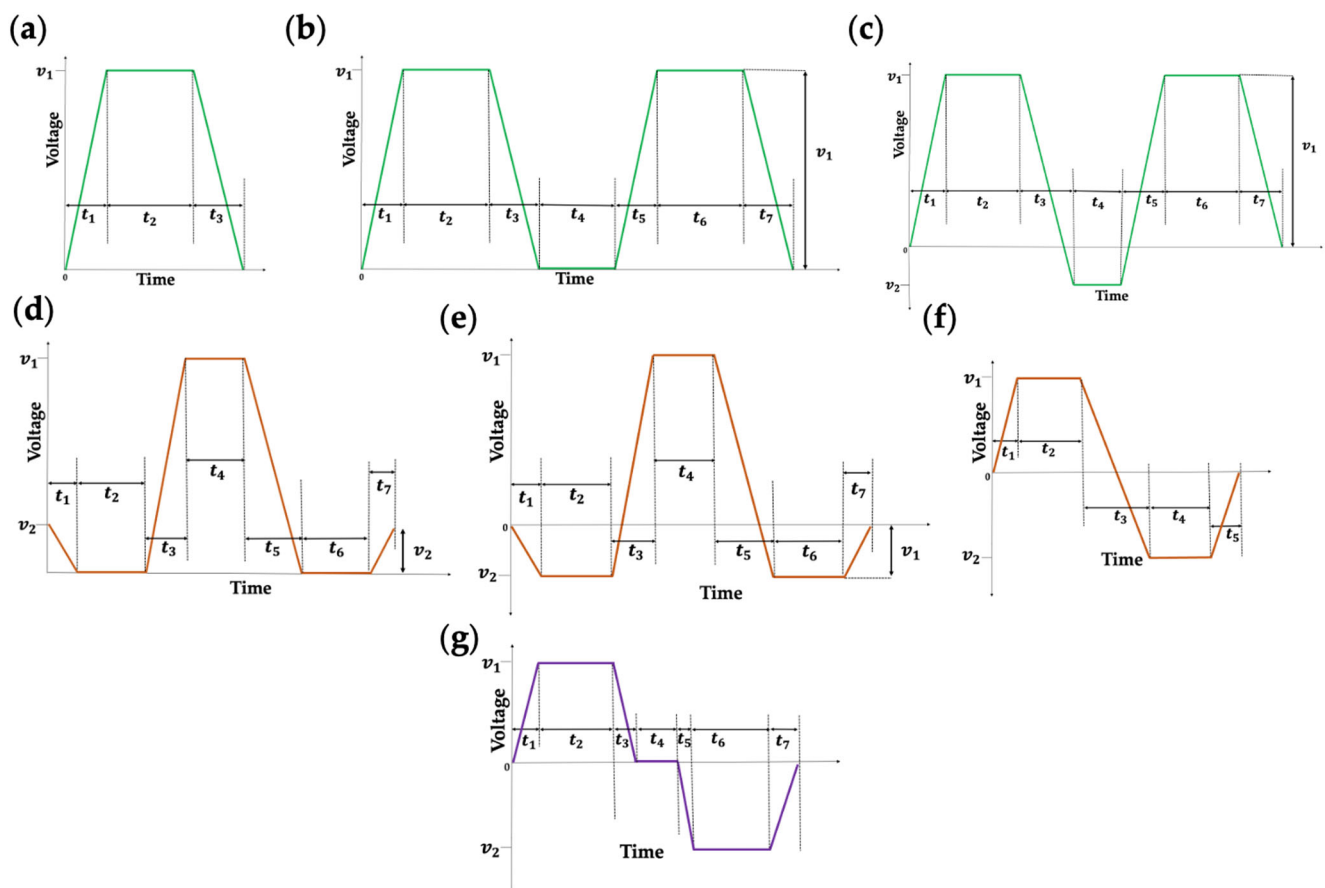


Figure 3. Schematic representations of various voltage waveforms applied in piezoelectric drop-on-demand inkjet printing: (a) single pulse, (b) unipolar M-shaped pulse, (c) bipolar M-shaped pulse, (d) unipolar W-shaped pulse, (e) bipolar W-shaped pulse, (f) bipolar-1 N-shaped pulse, and (g) bipolar-2 N-shaped pulse. These waveforms influence the dynamics of droplet ejection, suppress oscillations, and minimize defects such as satellite droplets and ligaments. Redrawn and adapted from Ref. [56].

Drive waveform parameters exert a decisive influence on the final properties of printed films by controlling droplet volume, velocity, and the relaxation of the meniscus. Increasing the main-pulse amplitude or pulse width raises the droplet volume and impact velocity, producing thicker wet films and promoting coalescence when drop spacing is tuned relative to the maximum spread, D_m (Figure 2). Appropriately timed secondary or dwell pulses damp residual chamber pressure and meniscus oscillations, reducing satellite droplets and ligaments and yielding cleaner edges and fewer microscopic voids. Bipolar sequences that briefly retract the meniscus before the ejection pulse further suppress overspray and aid separation of the primary droplet, improving dot fidelity [112]. At the device scale, the result is lower sheet resistance and equivalent series resistance, higher areal loading without cracking, improved ionic transport, and enhanced capacitance retention over cycling [39,49].

Recent literature offers quantitative insights. Giannakou and colleagues printed NiCo_2O_4 nanoparticle inks with a Dimatix DMP-2850 using 10 pL cartridges; they increased the viscosity of water with propylene glycol and added a surfactant to ensure a jetting number Z around 7 at 25 °C, avoiding satellite droplets [68]. Raising the temperature pushed Z above 10 and led to drop splitting. Particle sizes were kept below one-fiftieth of the nozzle diameter to prevent clogging. These conditions produced 42 μm dots with uniform coverage and films 295–477 nm thick after one or two passes. The resulting NiCo_2O_4 electrodes delivered areal capacitances of 22–29 mF cm^{-2} and mass-specific capacitances up to 1294 F g^{-1} with little degradation after 1000 cycles [68].

Mau and Seitz examined solvent volatility and concluded that inks based on highly volatile solvents rapidly form deposits on the nozzle, altering droplet trajectories and requiring frequent cleaning, whereas low-volatility solvents maintain stable ejection and consistent droplet volumes over extended periods. They emphasized that keeping Z within recommended ranges (e.g., $1 < Z < 10$ or $2 < Z < 20$) and maintaining the Weber number below the splashing threshold is essential to prevent satellite drops and splashing [112].

Yang et al. developed a drive waveform leveraging acoustic crosstalk to self-reinforce the actuation pressure while restraining satellite droplets and mitigating aerodynamic deceleration, thereby achieving monodisperse droplets at velocities up to 27.53 m s^{-1} . This approach expanded the printable viscosity range from 1 to 25 $\text{mPa}\cdot\text{s}$ to 1–40.3 $\text{mPa}\cdot\text{s}$ and the acceptable Ohnesorge number range from 0.1 to 1 to 0.03–1.18, suggesting that advanced waveform design can relax conventional limits [37].

For lithium-ion batteries, Pavlovskii et al. reviewed printable inks and noted that effective formulations for cathodes typically have viscosities of 10–12 $\text{mPa}\cdot\text{s}$, surface tensions of 28–33 mN m^{-1} , and particle sizes below 200 nm; they reported that poly(acrylic-co-maleic acid) binders yield better jetting stability than carboxymethyl cellulose [63]. These optimized inks enabled the fabrication of lithium-ion cells with areal capacities around 300 mAh cm^{-2} . In micro-supercapacitors, Wang et al. printed $\delta\text{-MnO}_2$ nanosheets from propylene glycol/water mixtures onto polyimide, achieving volumetric capacitances of 2.4 F cm^{-3} and 88% retention after 3600 cycles [113]. These examples illustrate how waveform optimization, combined with careful control of ink rheology and drop spacing, leads to uniform lines, reduced voids and defects, and improved electrochemical performance across both supercapacitors and batteries.

5. Development of Inks in Inkjet Printing

Inkjet printing has evolved to the point of establishing itself as a crucial technique for the manufacture of electronic devices due to its precision, versatility, and profitability. This section examines advancements in inkjet printing for electronic devices, focusing on the preparation, composition, and application of inks. By integrating findings from

various studies chronologically, the advantages and challenges associated with different ink formulations and printing conditions are highlighted.

In the formulation of battery electrodes via inkjet printing, the electrode slurry composition and particle size are critical parameters that govern both printability and final device performance. The slurry typically comprises an active material, which provides the requisite lithium ions and electrons for battery operation; a conductive agent to facilitate electronic mobility; a binder conferring structural integrity and mechanical properties; and dispersing agents and solvents ensuring slurry stability and homogeneity (Figure 4a,b). Uniform distribution of active material and conductive agent is imperative to prevent performance degradation, while binder selection must balance mechanical stability and chemical compatibility with other components.

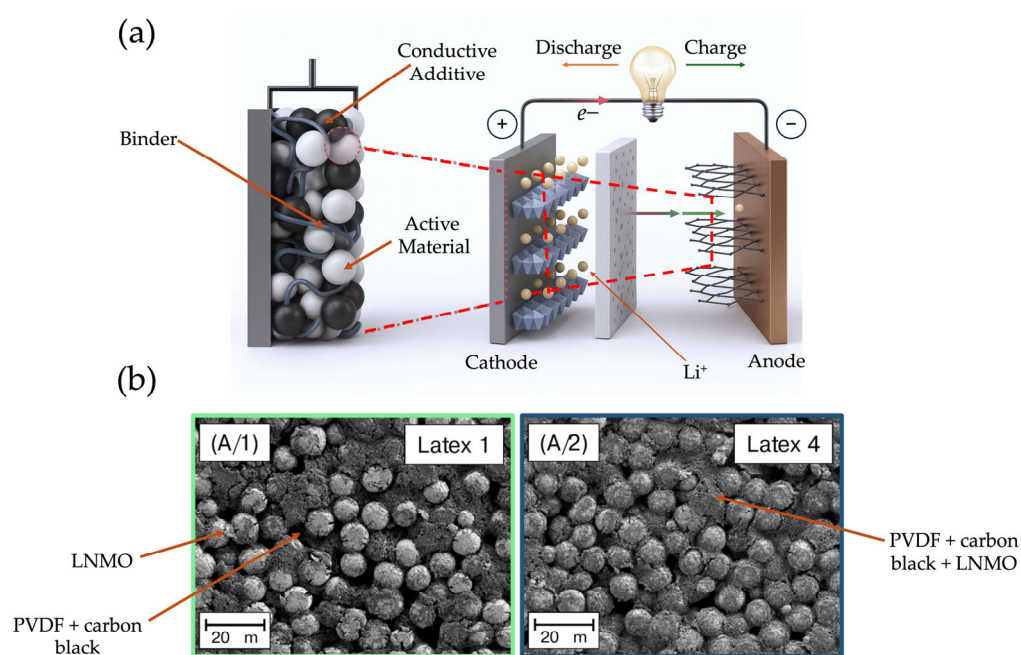


Figure 4. (a) Schematic representation of a composite electrode illustrating the arrangement of active material, conductive additive, and binder. The view shows how these components are integrated into the electrode microstructure. The diagram on the right situates the electrode within a complete battery cell, where it can function as either the cathode or the anode, enabling electron transport through the external circuit and ion migration through the separator. Redrawn and adapted from Ref. [114]. (b) SEM micrographs of LNMO composite electrodes prepared with two PVDF latex binders of different polarity, mixed with carbon black as a conductive additive. Left (Latex 1, less polar): PVDF and carbon black tend to agglomerate into clusters, leading to incomplete coverage of the $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) particle surfaces. Right (Latex 4, more polar): the binder–carbon mixture adheres more uniformly to the LNMO particles, forming a continuous conductive network and improving particle coverage [115].

The systematic process for ink selection and formulation optimization specifically tailored for inkjet printing technology, focusing on the fabrication of battery electrodes. This schematic is divided into four critical sections, each addressing a fundamental aspect of ink formulation to support printability and adequate electrochemical performance.

The process begins with the particle size section, where the initial step is to verify the particle size of the constituted powders to ensure they meet the printer's specific requirements. If the particle size is not within the acceptable range, the powders must be milled to the desired size or purchased in the correct particle size. It is crucial that the particle size is optimal concerning the electrochemical performance; if it is not, the process should be reconsidered as this is a fundamental requirement. Particle size is a determinant factor in

ink rheological properties, directly influencing electrode printability and electrochemical performance. Smaller particles can enhance electrochemical performance by reducing ionic and electronic transport distances but may also increase the risk of undesired side reactions. The IJP technique necessitates very small particle sizes to prevent nozzle clogging, potentially requiring additional milling processes that may alter the chemical and structural properties of the materials. The challenge lies in optimizing particle size to achieve a balance between optimal electrochemical performance and printing feasibility. Strategies such as particle coating with materials like SiO₂ or Al₂O₃ can improve performance, though potentially at the expense of ionic conductivity. Grinding techniques to reduce particle size must be applied judiciously to avoid structural damage that could degrade the material's electrochemical properties.

Next, the focus shifts to viscoelasticity. This step involves selecting a binder that balances electrochemical performance with mechanical robustness in the final electrode. It is essential to determine whether the ink formulated with the chosen binder exhibits strain-hardening, which can impair drop formation, layer integrity, and long-term durability of printed electrodes. If strain-hardening is observed, a lower-molecular-weight or alternative binder should be adopted. Regarding rheological characteristics, the solvent system must dissolve or swell the binder and be compatible with the solid particles [116]. Achieving target viscosity and surface tension within the inkjet jetting window is critical for a stable ink formulation [117]. If the neat solvent does not meet these requirements, appropriate solvents or rheology modifiers may be introduced, provided they do not compromise electrochemical performance, thus preserving the balance between printability and functionality.

Finally, the selection of a dispersant compatible with the binder/solvent system is used to stabilize the suspension and ensure proper particle dispersion. A well-dispersed ink minimizes agglomeration and sedimentation, enabling consistent droplet formation, feature definition, and electrode performance. If stability is inadequate, the dispersant type and loading should be adjusted, or compatible surfactants may be added, while confirming that these changes do not degrade electrochemical performance.

Through these sequential decisions, this workflow provides a structured route to define ink compositions for inkjet printing of battery electrodes. The resulting inks meet the physicochemical requirements for printability and deliver the electrochemical performance needed for effective electrode fabrication. The following sections examine representative formulations and highlight how the above parameters govern printability and electrochemical outcomes.

Regarding particle size, Zhao [118] highlighted the necessity of precisely controlling particle dimensions in inks intended for inkjet printing. They prepared electrode inks containing nano-sized Li₄Ti₅O₁₂ particles ranging from 50 to 300 nm, using intensive milling techniques to meet printer-specific size constraints. By utilizing a combination of dispersants and binders such as sodium-carboxymethyl-cellulose within a carefully adjusted co-solvent system (water, ethanol, diethylene glycol, triethanolamine, and IPA), they successfully created stable inks with appropriate viscosity and surface tension, preventing nozzle clogging and ensuring smooth deposition. The systematic optimization of particle size not only enhanced the electrochemical properties, achieving capacities around 174 mAh g⁻¹, but also ensured robust printability, reinforcing particle size as a foundational parameter in ink design.

The importance of carefully adjusting rheological characteristics and solvent choice was explored by Maximov et al. [119]. In their study, Li-rich cathode materials (Li_{1.25}Mn_{0.54}Ni_{0.13}Co_{0.13}O₂) were dispersed in N-methyl-2-pyrrolidone along with carbon nanotubes and PVDF binder. To tune viscosity into the optimal printing range (8–10 cP),

they systematically evaluated different glycol additives, ultimately selecting propylene glycol due to its favorable rheological properties and compatibility with their ink formulation. This solvent selection improved jetting stability, reduced satellite droplet formation, and maintained dispersion stability over extended periods. The resulting ink could be consistently printed onto aluminum foils with high fidelity, forming layered cathodes that demonstrated superior electrochemical performance, emphasizing solvent selection as pivotal for balancing ink printability and electrochemical performance.

Further developments by Nyabadza et al. [44] in 2025 provided valuable insights into the role of dispersion stability and substrate compatibility. They used a pulsed-laser ablation in liquid (PLAL) technique to produce uniform Mn-based nanoparticles (approximately 64 nm) dispersed in isopropanol (IPA). The selected solvent (IPA) significantly improved dispersion stability, achieving a ζ -potential of approximately 44 mV, crucial for maintaining colloidal stability and preventing sedimentation issues encountered with water-based alternatives. Furthermore, the researchers enhanced ink adhesion and printability by carefully modifying the substrate surface laser-textured copper current collectors with increased roughness (up to 2.1 μm), demonstrating that the interplay between dispersion and substrate surface properties is essential to achieve consistent printing outcomes and stable electrochemical interfaces.

Building upon this work, Nyabadza et al. [45] introduced a real-time monitoring strategy for feedback-controlled ink formulation, combining continuous-flow PLAL with inline dynamic light scattering (DLS) and UV-Vis spectroscopy. This approach allowed precise control over nanoparticle size (~ 3 nm), viscosity (1.3 mPa s), and ζ -potential (~ 44 mV). Their statistically driven optimization process (a 3×3 design-of-experiments) identified ideal printing conditions (40 kHz jetting frequency, 28 °C nozzle temperature, and 30 printed layers). Their methodology ensured reproducibility and robust electrochemical performance of printed films on flexible substrates, demonstrating the feasibility of transitioning laboratory-scale ink formulations to industrial-scale applications through real-time adaptive control.

Inkjet-printable inks are also moving decisively toward eco-friendly, water-based formulations that replace toxic solvents and synthetic surfactants without sacrificing jetting stability or device performance. A prominent route is to use biobased dispersants that both stabilize graphitic colloids in water and modulate dynamic surface tension within the inkjet window. For example, alkali lignin functions as a natural amphiphile for graphene oxide (GO), producing aqueous GO inks whose ζ -potential and wetting can be tuned for reliable DoD printing; temperature sensors printed from such inks on flexible substrates exhibit reproducible thermosensitive response after mild post-treatments [120]. Protein-stabilized nanoclusters offer a complementary bio-surfactant strategy: a fully inkjet-printable, water-based bio-ink combining protein nanoclusters with exfoliated graphene enables all-printed paper analytical devices, evidencing that biomolecules can deliver both colloidal stabilization and electroactivity in benign media [121]. Together, these studies demonstrate how natural macromolecules can deliver the interfacial control normally attributed to fluorosurfactants while keeping rheology and surface tension in the jettable regime.

Beyond bio-surfactants, additive-free aqueous graphene derivatives have matured to fully inkjet-printed electrodes. Nitrogen-doped, carboxylate graphene (“NGA-ink”) dispersed in pure water and without low-boiling co-solvents yields uniform, all-inkjet-printed electrodes that perform robustly as electrochemical sensors, while the high surface density of carboxyl groups facilitates subsequent bio-functionalization—linking sustainability with manufacturability and assay integration [122]. In parallel, liquid-phase-exfoliated graphene formulated as a water-based ink has been printed into continuous films that deliver high field-emission current densities ($\sim 7.2 \times 10^2 \text{ A cm}^{-2}$) and multi-hour stability, underscoring

that aqueous systems can meet stringent electronic transport requirements once flake size distributions and drying dynamics are controlled [24].

The same aqueous-first philosophy is now evident in energy-harvesting and storage-adjacent devices. Water-based GO and GO-PEDOT:PSS inks, prepared through low-cost, scalable chemistries, have been inkjet-printed into flexible moisture-energy generators that convert ambient humidity gradients into electrical output—evidence that green inks and low-temperature processing can support multilayer functional stacks on common polymer films [25]. Importantly, these demonstrations show that solvent selection (water with minimal benign additives), natural or no surfactant, and post-print mild anneals can be co-optimized with waveform design to maintain drop integrity and suppress coffee-ring effects, producing dense percolation networks even at modest solids loadings.

Finally, progress on predictive printability is closing the loop between green formulation and process robustness. A methodology for a priori evaluation of water-based graphite dispersions quantifies how particle volume fraction, ζ -potential, and dynamic surface tension map onto the Z-number window for stable DoD jetting, providing a transferable framework to de-risk aqueous, bio-stabilized systems before scale-up [30]. Coupled with the bio- and additive-free strategies above, such tools enable water-based conductive inks (graphene/GO and related carbons) to replace NMP-based slurries in printed energy devices without compromising line fidelity, interfacial resistance, or long-term stability.

Collectively, these studies support the structured approach presented earlier, emphasizing that optimal ink formulation requires carefully balancing particle size, viscoelastic properties, solvent compatibility, and dispersion stability, thereby ensuring both reliable printability and high electrochemical performance.

6. Development of Batteries in Inkjet Printing

The development of batteries via inkjet printing is relevant for producing energy-storage units in a wide range of shapes and sizes, a prerequisite for next-generation wearables, flexible IoT nodes, and other compact electronics. Figure 5 illustrates this capability: successive, digitally defined droplets build an interdigitated microbattery in which the cathode, electrolyte, anode, and final encapsulation are all deposited directly onto a single substrate. By eliminating discrete components such as pre-cut separators and manual stack assembly, inkjet printing streamlines fabrication, lowers cost, and improves yields. The paragraphs that follow trace the field's evolution chronologically, drawing on landmark studies to highlight key material choices, printing parameters, and performance milestones in printed batteries.

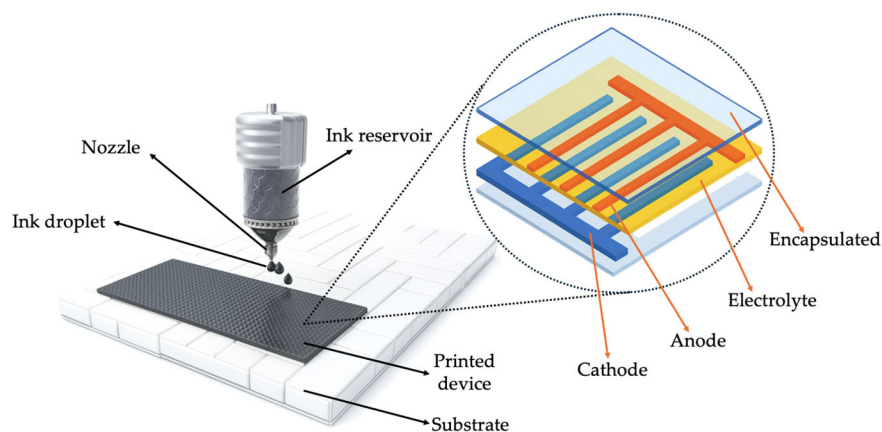


Figure 5. Inkjet printing process used to fabricate a multilayer interdigitated microbattery. The inset shows the layered architecture, including printed cathode, anode, electrolyte, and encapsulation on a substrate.

Table 2 shows this chronological rise in electrochemical performance for inkjet-printed batteries. Early entries such as planar zinc–manganese dioxide cells and thin-film lithium iron phosphate or lithium titanate deliver around 330 Wh kg^{-1} at about 220 W kg^{-1} for zinc–manganese dioxide, about 140 mAh g^{-1} at 1 C with roughly 98.6 percent coulombic efficiency for lithium iron phosphate [43], and about 128 mAh g^{-1} at $\text{C}/2$ for lithium titanate over 100 cycles [123]. More recent entries use higher capacity chemistries and three-dimensional architectures: vanadium pentoxide with MXene maintains about 112 mAh g^{-1} at 10.5 C for hundreds of cycles [124], printed silicon films sustain around 1000 mAh g^{-1} for more than one thousand cycles [66] and silicon micropillars reach about 2000 mAh g^{-1} for one hundred cycles [27], while lithium-rich layered oxides provide about 200 mAh g^{-1} at 0.1 C with specific energy near 835 Wh kg^{-1} and around 1037 Wh L^{-1} [125]. When these results are compared with recent work using other manufacturing routes, such as porous lithium iron phosphate produced by three-dimensional printing that reports about 121.7 mAh g^{-1} at 0.5 C with coulombic efficiency exceeding 99.7 percent and energy near 350 Wh kg^{-1} [126], LiFePO_4 electrodes fabricated by fused deposition modeling that achieve about 87 mAh g^{-1} at $\text{C}/20$ and 45 mAh g^{-1} at $\text{C}/10$ with 42% irreversible capacity loss in the first cycle [127], or silicon anodes produced by fused-deposition modeling that achieve about 345 mAh g^{-1} at 20 mA g^{-1} with about 96 percent retention over 350 cycles [128].

Overall, these cross-route results suggest that well-designed inkjet electrodes are already competitive on rate and even specific energy, while many extrusion-based builds pay a penalty in coarse porosity, higher binder fractions, and first-cycle losses. The real gap for inkjet is not gravimetric capacity but areal loading and stack-level energy, since many demonstrations still operate below practical loadings and footprint. By contrast, 3D and fused-filament methods can push mass loading but often trade away kinetics and interfacial control, which limits power and retention. A fair comparison must normalize by areal loading, thickness, porosity, and test rate; under those metrics, the next milestones for inkjet are stable performance above $2\text{--}3 \text{ mAh cm}^{-2}$ at $\geq 1 \text{ C}$ with long-cycle durability and high yield on flexible substrates.

Between 2009 and 2015, Inkjet Printing emerged as a promising alternative to traditional electrode fabrication methods in battery manufacturing. Early investigations demonstrated that commercially available printheads could accurately deposit both active materials and electrolytic separators, thereby extending Inkjet Printing beyond its organic-ink origins. These pioneering efforts laid the groundwork for understanding how fundamental printing parameters such as droplet formation dynamics, ink rheology, and morphology control directly influence the electrochemical performance of printed devices.

In 2009, Ho et al. [69] harnessed a Super Inkjet system based on electrohydrodynamic (EHD) actuation to fabricate three-dimensional silver–zinc microbatteries. By generating femtoliter-scale droplets, this EHD approach achieved high morphological precision, enabling the construction of micropillars $40 \text{ }\mu\text{m}$ tall and $10 \text{ }\mu\text{m}$ in diameter, spaced roughly $100 \text{ }\mu\text{m}$ apart. The resulting architecture delivered a 60% increase in energy capacity compared to planar electrodes, thanks to its enhanced active-material surface area. However, the study also revealed a spatial-utilization drawback: only 2.5% of the available surface was occupied by active material. Crucially, Ho et al. showed that tight control over droplet volume combined with rapid solvent evaporation can suppress convective flows and surface-tension effects, permitting complex 3D structure formation without external templates.

Table 2. Representative Studies of Inkjet Printing in Battery Manufacturing: Device Types, Ink Formulations, and Electrochemical Outcomes.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
Lithium anode-free with Ag lithophilic sites [28]	Aqueous solution with fluoropolymer	Ag nanoparticles (40 wt%)	Not specified	Dimatix DMP-2850, 20 kHz, 30 V, droplet spacing 10–40 μm , sintering at 150 $^{\circ}\text{C}$ for 3 h	Copper foil (15 μm)	Sintering at 150 $^{\circ}\text{C}$ for 3 h under N_2	Coulombic efficiency 97% at 0.2 mA cm^{-2} for 250 cycles; Nucleation overpotential 13.5 mV
Flexible planar Zn-MnO ₂ battery [43]	DI-water	Triton X-100, PVB	3.3–8.2 mPa·s	Epson L130, high quality mode, 70 $^{\circ}\text{C}$ for 12 h	Bond paper (100 GSM)	Heat treatment at 70 $^{\circ}\text{C}$ for 12 h in vacuum	600 mA g^{-1} (rate); 184.24 mAh g^{-1} after 500 cycles at that rate; Coulombic efficiency 99.24% (cycle 25); Energy density 330.15 Wh kg^{-1} at 220 W kg^{-1}
Flexible lithium-ion battery LTO/LFP [29]	Deionized water	Sodium ascorbate, LDS	2.58 mPa·s	Breva thermal inkjet, 11.5 V, 2.2 μs	Flexible PET	Overnight drying	12.5 $\mu\text{A cm}^{-2}$; 6.09 mAh g^{-1} ; 100 cycles.
3D silicon anode [27]	DI-water	Triton X-100, PVB or PEDOT:PSS	3.3–8.2 mPa·s	CeraPrinter X-Series, max voltage 120 V, <15 μs	Copper foil	Heating at 80 $^{\circ}\text{C}$ for 16 h in vacuum	0.2 C (after 4 formation cycles at 0.05 C); Lithiation capacity 2000 mAh g^{-1} (active material) for 100 cycles with no fading.
V ₂ O ₅ /MXene heterostructure cathode for LIB [124]	Deionized water	Not specified	Not specified	Dimatix DMP-2800, 10 pL cartridge	Not specified	Drying at 60 $^{\circ}\text{C}$ overnight	10.5 C; 112 mAh g^{-1} ; 680 cycles with 91.7% capacity retention; Coulombic efficiency 96.5%
Li ₄ Ti ₅ O ₁₂ thin-film electrodes [123]	Water + ethylene glycol + 2-propanol	Li-PAA, LDS, CNT or CB	2.58–2.82 cP	Breva thermal inkjet, 11.5 V, 2.2 μs , 600 dpi	Copper substrate with graphite spray	Drying at 80 $^{\circ}\text{C}$ for 3 h in vacuum	C/2; 128 mAh g^{-1} for 100 cycles (negligible fade); Coulombic efficiency 100%

Table 2. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
Dendrite-free zinc anode [129]	Conductive silver ink	Ag nanoparticles	Not specified	Custom piezoelectric inkjet printer, ~20 V	3D carbon cloth	Annealing at 180 °C for 1 h	5.0 A g ⁻¹ ; 184 mAh g ⁻¹ after 1200 cycles
3D LiFePO ₄ cathodes [130]	Deionized water	NaCMC, Triton X-100	Not specified	PICO Pulse, 100 µm orifice	Carbon-coated aluminum current collector	Vacuum drying <1 mbar for 8 h at 100 °C	1 C; ~140 mAh g ⁻¹ for 165 cycles; Coulombic efficiency 98.6%
Li-rich cathode material [125]	NMP	PVP	8–10 cPs	Dimatix Material printer 2831, 10 pL cartridge	Aluminum foil	Heat treatment conditions varied	0.1 C; 200 mAh g ⁻¹ after 80 cycles; Energy density 1037 Wh L ⁻¹ ; Specific energy 835 Wh kg ⁻¹
All-graphene micro supercapacitor [131]	Deionized water, ethylene glycol, phosphoric acid	Nano-graphene oxide (nGO)	Not specified	Inkjet printing on Kapton, annealing to remove binder	Flexible Kapton	Annealing to remove binder	5 mV s ⁻¹ (scan rate); Areal capacitance 313 µF cm ⁻² ; Volumetric energy density ~0.2 mWh cm ⁻³ with power density ~4 mW cm ⁻³ ; 11,000 cycles with >65%
Silicon nanoparticle anode for Li-ion battery [66]	Deionized water	Polymer binders: PEDOT:PSS, PVP, CMC, Na-alginate	~10 mPa·s	HP Deskjet 2540, room temp, multiple passes	Copper foil	Vacuum drying at 60 °C overnight	0.1 C; 1000 mAh g ⁻¹ for >1000 cycles; Coulombic efficiency ≈ 98.6%
Lithium-sulfur micro cathodes [132]	Cyclohexylpyrrolidone (CHP)	Not specified	Not specified	Fuji Dimatix 2800, 10 pL cartridges	Flexible aluminum foil or SiO ₂ wafers	Gradual annealing at 150 °C (~4°/min)	C/2; ~700 mAh g ⁻¹ after 100 cycles

Table 2. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
LiFePO ₄ water-based cathodes [133]	Water + glycerol	CMC, carbon black, Triton X-100	~13 cP	Dimatix-2800 inkjet printer	Aluminum foil and CNT paper	Vacuum drying at 300 °C for 2 h	0.1 C; 151.3 mAh g ^{−1} (first discharge); Coulombic efficiency 94.4% note: test rate/window specified at 0.1 C, 2.0–4.0 V.
Solid electrolyte for microbatteries [134]	Ionogel sol precursor (TMOS + ionic liquid)	Silica-based ionogel	10–12 mPa·s optimal	Dimatix DMP2800, 15 kHz, 16 V, 800 µm gap	Porous composite electrodes	Polycondensation after printing	C/10; Areal capacity ≈ 300 mAh cm ^{−2} for 100 cycles (≈60 mAh g ^{−1} on LFP)
Zinc-silver 3D microbattery [69]	Not specified (silver nanopaste + n-tetradecane)	Not specified	Not specified	Super inkjet printing, sintering at 250 °C for 1 h	Glass substrates	Sintering at 250 °C for 1 h	0.1 mA (discharge); Energy density 3.95 mWh cm ^{−2} ; Cycles: multiple with rapid fade (<20% by ~7 cycles)

By 2015, the focus had shifted to aqueous-based inks and substrate compatibility. Gu et al. [133] formulated LiFePO_4 cathode inks composed of LiFePO_4 , carbon black, and CMC binder in an 8:1:1 ratio at 13 mPa·s viscosity and pH 9.13 for a Dimatix-2800 piezoelectric printer. They demonstrated that ink pH critically governs stability and performance, while substrate choice dictates interfacial chemistry: aluminum foil suffered parasitic LiAlO_2 and AlPO_4 formation, whereas carbon-nanotube paper enabled superior cycling stability (Figure 6a). In parallel, Delannoy et al. [134], advanced sol-gel ink strategies by printing silica-based ionogel electrolytes via a Dimatix DMP2800. Their 10–12 mPa·s inks infiltrated porous electrodes and concurrently formed $\sim 5\text{ }\mu\text{m}$ separator layers, achieving ionic conductivities 100 times higher than LiPON through controlled precursor transformation (Figure 6b). Together, these studies underscored that fine-tuning ink chemistry, substrate interactions, and post-print conversions is essential for reliable, high-performance device integration.

Overall, these pioneering studies revealed several technical challenges inherent to early-stage Inkjet Printing approaches. Uniform deposition required precise control over viscosity, surface tension, and substrate wetting. Additionally, spatial utilization inefficiencies, unwanted interfacial reactions, and the complexity of optimizing multiple printing parameters simultaneously highlighted critical areas needing improvement. Moreover, intensive thermal post-processing (250–300 °C) and the necessity for multiple printing passes underscored limitations in process efficiency and deposition density at this early stage. These fundamental insights set the stage for subsequent innovations aimed at overcoming these initial challenges.

Between 2019 and 2021, Inkjet Printing for battery electrodes evolved from simple single-layer patterns to complex multilayer constructs. The central goal became boosting areal capacity by stacking thicker, multilayer films without sacrificing electrochemical stability or coulombic efficiency, thereby directly addressing the first-generation's low active-material coverage ($\approx 2.5\%$) and limited capacity per unit area. Meeting this objective required meticulous rheological tuning, strategic layer-by-layer deposition protocols, and finely optimized printing parameters to avoid morphological faults such as cracking or delamination.

Ben-Barak et al. [130] contributed progress by utilizing Drop-on-Demand technology for constructing three-dimensional LiFePO_4 cathodes. Employing a piezoelectric PICO Pulse system, the researchers formulated inks comprising LiFePO_4 , carbon black, sodium carboxymethyl cellulose (NaCMC), and dispersing agent Triton X-100, achieving a 20 wt% solid loading. This multilayer approach involved meticulous layer-by-layer deposition, significantly enhancing electrode thickness to approximately 90 μm after 100 layers without compromising structural integrity. The printing parameters and gentle vacuum drying at 100 °C substantially reduced morphological defects compared to earlier thermal treatments. Figure 6c shows that the resulting electrodes demonstrated superior electrochemical performance, maintaining capacities exceeding 140 mAh g^{-1} at 1 C, coulombic efficiencies over 98%, and cycle stability over 165 cycles. Critically, the mechanical Drop-on-Demand approach effectively overcame traditional inkjet surface tension limitations, allowing high solid content inks to be deposited uniformly.

Similarly, in 2021 Chen et al. [129] introduced an innovative multilayer inkjet printing method for zinc anodes aimed at suppressing dendritic growth. Using a customized piezoelectric inkjet printer, the researchers deposited silver nanoparticles onto three-dimensional carbon cloth substrates. These nanoparticles acted as nucleation sites to ensure uniform zinc deposition, significantly reducing dendrite formation. The multilayer construction approach, combined with a moderate annealing treatment (180 °C), yielded homogeneously distributed silver nanoparticles with enhanced thermal and electrical properties. Electro-

chemical tests showed high results, including a capacity of 184 mAh g^{-1} after 1200 cycles at 5.0 A g^{-1} and minimal voltage hysteresis. This strategy improved current distribution and prevented localized hotspots, showcasing multilayer architectures' capability to enhance device reliability and thermal management significantly.

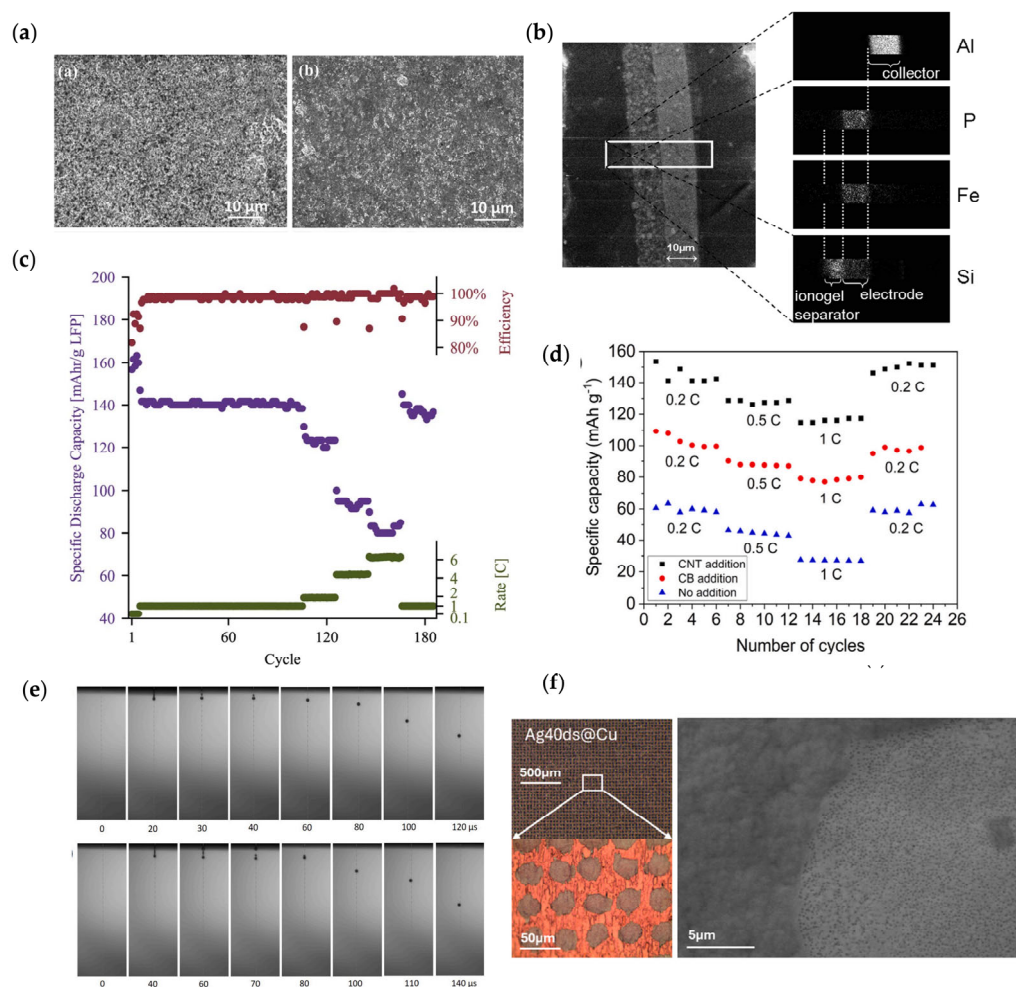


Figure 6. (a) Surface SEM images of LiFePO₄ ink on (left) Al foil vs. (right) CNT paper, showing parasitic LiAlO₂/AlPO₄ formation on Al and uniform active-material coverage on CNT substrate [133]. (b) Cross-section SEM and EDX elemental mapping of a tape-cast LiFePO₄ porous composite electrode with printed silica-based ionogel separator on top. (Left) SEM cross-section image showing the ~5 μm-thick ionogel layer (white rectangle) deposited over the electrode. (Right) Corresponding EDX elemental maps for Al (collector), P (ionogel), Fe (active LiFePO₄), and Si (ionogel), confirming the spatial distribution and composition of the separator [134]. (c) Cycle life of a multilayer, drop-on-demand printed LiFePO₄ cathode at 0.1 C: specific discharge capacity (left axis) and coulombic efficiency (right axis) over 180 cycles [130]. (d) Discharge capacity vs. number of cycles at different C-rates (0.2 C, 0.5 C, 1 C) for LTO electrodes formulated with carbon black (CB), multi-walled carbon nanotubes (MWCNT), and without additive. The MWCNT-based formulation shows the highest capacity (150.3 mAh g^{-1} at 0.2 C) and superior cycling stability over 100 cycles [123]. (e) Stroboscopic recording of droplet formation during inkjet printing of Si/C anode inks. Top series (0–120 μs): CMC-based ink. Bottom series (0–140 μs): PEDOT:PSS-based ink. Each frame shows the main droplet emerging, filament break-up, and clean detachment along the nozzle centerline (dotted line), with no satellite drops. This controlled jetting underpins the layer-by-layer assembly of 3-D silicon-carbon micropillars [27]. (f) Left, optical micrographs of the Ag-dot array produced with a Fujifilm Dimatix printer: a large-area view (scale bar 500 μm) and a magnified inset (scale bar 50 μm) reveal a uniform lattice of ~30 μm dots on a 40 μm pitch. Right, SEM image of a single sintered dot (scale bar 5 μm) showing its nanoporous morphology, which provides abundant nucleation sites for lithium and promotes dendrite-free growth [28].

Viviani et al. [123] systematically explored thin-film $\text{Li}_4\text{Ti}_5\text{O}_{12}$ electrode formulations, employing a thermal Brevia printer with optimized ink compositions featuring carbonaceous additives such as carbon black and multi-walled carbon nanotubes (MWCNT). Their careful rheological optimization allowed precise multilayer deposition ($\sim 1.5 \mu\text{m}$ per pass), yielding compact, crack-free electrode layers when MWCNT was used. Compared to carbon black, the MWCNT-based formulations demonstrated superior electrochemical performance, achieving capacities of 128 mAh g^{-1} at 0.5C and dramatically reduced charge transfer resistance (Figure 6d). This work critically underscored the importance of additive morphology, where the fibrous structure of MWCNT facilitated both smoother inkjet deposition and superior final electrode properties.

Collectively, these studies not only overcame the low active-material coverage and limited areal capacities of the first generation, but also established the multilayer, high-rheology, and additive-optimization strategies that pave the way for next-generation inkjet printing innovations targeting even higher resolution, enhanced print throughput, and broader material compatibility.

By 2024, inkjet printing was being used as a versatile fabrication tool, targeting three critical frontiers: 3D electrode architectures for high areal capacities, lithophilic patterning for lithium-metal anodes, and fully flexible, deformable batteries.

Sztymela et al. [27] reported three-dimensional silicon-based anodes using a CeraPrinter X-Series inkjet system equipped with a Dimatix Class SL piezoelectric printhead. They developed aqueous inks containing silicon or silicon-carbon core-shell nanoparticles combined with conductive carbon black and advanced binders like PEDOT:PSS. These formulations enabled precise micropillar fabrication with rapid pulse timings (Figure 6e), improving electrode capacities up to 2000 mAh g^{-1} . Critical analysis indicated that core-shell silicon-carbon structures notably enhanced cycling stability compared to pure silicon, although challenges remained with droplet generation stability and occasional nozzle clogging.

Meanwhile, In 2024 Mirbagheri et al. [28] advanced lithium-metal batteries by inkjet printing silver-based lithophilic sites onto copper current collectors (Figure 6f). Utilizing a Fujifilm Dimatix printer and nanoparticle-loaded fluoropolymer inks, they achieved uniform and controlled silver depositions. This strategy reduced lithium nucleation overpotential and improved coulombic efficiencies (97% over 250 cycles). The printed silver regions effectively guided lithium growth, preventing dendrite formation, demonstrating the potential of inkjet-printed lithophilic sites for scalable and dendrite-free lithium-metal batteries.

Viviani et al. [29] introduced flexible batteries with fully printed $\text{Ti}_3\text{C}_2\text{T}_x$ MXene current collectors using a Brevia thermal printer. The optimized aqueous MXene inks exhibited excellent processability and electrochemical stability. When coupled with active electrodes of LTO and LFP, these devices achieved a notable 21% improvement in gravimetric energy density compared to traditional copper collectors. Crucially, these batteries maintained their performance under significant mechanical bending, highlighting the viability of inkjet-printed flexible batteries for wearable applications.

Finally, Sarma Choudhury et al. [43] demonstrated fully flexible Zn-MnO₂ batteries printed onto commercial paper substrates using an Epson thermal printer. Their multilayer architecture, combining reduced graphene oxide collectors, MnO₂/PEDOT:PSS cathodes, zinc anodes, and gel electrolytes, yielded electrochemical performance of $330.15 \text{ Wh kg}^{-1}$ energy density at 220 W kg^{-1} . These batteries exhibited high mechanical flexibility, retaining capacity under repeated bending and folding, confirming their suitability for portable electronics.

Collectively, these studies illustrate distinct yet complementary approaches addressing different application needs. Three-dimensional stacking strategies significantly enhanced volumetric capacities, lithophilic printed patterns enabled high-energy lithium-metal chemistries, and substrate flexibility addressed emerging demands in portable and wearable electronics.

Despite these advances, several technical challenges persist, particularly nozzle clogging, spatial resolution limitations, and long-term mechanical durability. Addressing these issues requires further development in printing hardware, such as improved nozzle designs and integrated roll-to-roll printing systems. Future research will likely focus on hybrid additive manufacturing processes to scale up production capabilities, laying the groundwork for the commercial manufacturing of next-generation energy storage devices through advanced digital printing technologies.

7. Development of Supercapacitors in Inkjet Printing

Inkjet printing has advanced the fabrication of supercapacitors by enabling precise, mask-free deposition of active materials on a wide range of substrates. From the first demonstrations on flexible textiles to today's multifunctional, hybrid devices, each wave of research has addressed specific challenges: substrate compatibility, pattern resolution, ink chemistry, and device architecture, while exploring new performance limits. The next paragraphs show a chronology that highlights key milestones in inkjet-printed supercapacitor development.

Table 3 captures the shift from early carbon and manganese dioxide microdevices toward two-dimensional MXene and other pseudocapacitive systems, with marked gains in volumetric metrics and cycling stability. Recent all-MXene micro-supercapacitors reach about 562 F cm^{-3} with nearly full capacitance retention after ten thousand cycles and energy near $0.32 \text{ } \mu\text{Wh cm}^{-2}$ [31], asymmetric nickel sulfide on MXene with reduced graphene oxide reports about 429 F cm^{-3} and around 33.5 mWh cm^{-3} with roughly 80% retention [135], and textile-integrated MXene devices demonstrate solid-state operation with about 0.86 mF cm^{-2} , around $0.08 \text{ } \mu\text{Wh cm}^{-2}$ and about 99.2% coulombic efficiency [41]. Benchmarks from other techniques reinforce the comparison: aerosol-jet printed MXene micro-supercapacitors report around 611 F cm^{-3} [73], 3D printed NiCoP/Ti₃C₂ MXene architectures achieve about 2.0 F cm^{-2} at high mass loading with energy densities reaching 0.89 mWh cm^{-2} [21], and screen-printed supercapacitors based on tin sulfide decorated activated carbon deliver approximately 33.8 mF cm^{-2} with energy density of $9.2 \text{ } \mu\text{Wh cm}^{-2}$ [136]. These screen or extrusion printed supercapacitors often rely on thicker films to reach such areal capacitances and energy densities. In this context, the most recent inkjet-printed devices meet or exceed state-of-the-art volumetric performance while providing finer pattern fidelity and easier integration on flexible substrates, with remaining limitations tied to per-pass solids loading and throughput rather than intrinsic electrochemical behavior.

Three empirical regularities emerge from these reports. First, compact MXene microdevices are associated with high volumetric capacitance and stable cycling, with several studies clustering near $500\text{--}600 \text{ F cm}^{-3}$ and retention approaching 100% at 10,000 cycles. Second, the highest areal energy values are typically linked to increased thickness and mass loading in screen or 3D-printed electrodes, and these gains are often accompanied by higher equivalent series resistance (ESR) and slower rate response. Third, textile and other solid-state formats prioritize mechanical robustness and safety, which is reflected in modest areal energies together with high coulombic efficiency.

Within this landscape, inkjet-printed electrodes typically report thin, well-defined layers with competitive volumetric performance. The principal constraints arise at the process level rather than from intrinsic electrochemistry: per-pass solids loading, nozzle stability at high MXene or other pseudocapacitive contents, and uniformity across large arrays. Overall, recent inkjet-printed micro-supercapacitors show competitive volumetric performance while process constraints remain the main bottleneck. The following paragraphs examine, in detail, strategies reported to raise mass loading without impairing ion transport and to improve stability under humidity and repeated bending systems.

In 2010, Chen et al. [137] investigated the viability of inkjet printing for supercapacitors on textile and flexible substrates, breaking the traditional limitation of rigid substrates. Their groundbreaking work utilized single-walled carbon nanotubes (SWNTs) functionalized with sodium dodecyl sulfate (SDS) as surfactant, deposited onto cotton fabric and PET substrates using a commercial Epson Artisan 50 piezoelectric printer (Figure 7a). The technical contribution consisted of demonstrating that precise control of geometry, location, and electrical conductivity was achievable on unconventional substrates, attaining controllable thicknesses between 20 and 200 nm through multiple repeated printing passes ranging from 40 to 200 cycles. However, the authors identified the high sheet resistance observed on textile substrates ($815 \Omega/\text{sq}$) as a critical challenge, which dramatically contrasted with that obtained on PET ($78 \Omega/\text{sq}$) and significantly limited electrochemical performance. To address this fundamental issue, Chen et al. proposed several improvement strategies in their work: integration of RuO_2 nanowires to increase specific pseudo capacitance, systematic optimization of printing cycles to enhance nanotube percolation, and exploration of hybrid materials combining the conductive properties of SWNTs with pseudocapacitive capacitance. This SWNT/ RuO_2 hybrid materials approach achieved an increase in specific capacitance from 65 F g^{-1} (pure SWNT) to 138 F g^{-1} , laying the conceptual foundations for subsequent developments in hybrid ink formulations and post-printing treatments that would characterize the following decade of research.

Concurrently, Pech et al. [138] developed the first high-spatial-resolution printed interdigital micro-electrode, achieving finger widths as narrow as $40\text{--}100 \mu\text{m}$ on oxidized silicon substrates. Although this advancement in spatial resolution did not directly resolve the conductivity problem identified by Chen, it represented an important complement by demonstrating that inkjet printing could achieve micrometric precision comparable to conventional photolithographic techniques. Their technical approach employed high-specific-surface-area activated carbon ($1700\text{--}1800 \text{ m}^2 \text{ g}^{-1}$) dispersed in ethylene glycol with PTFE as binder and Triton X100 as surfactant, deposited using the AltaDrop[®] system from Altatech (Altatech, Montpellier, France). The key methodological innovation consisted of hydrophobic functionalization of the substrate with octadecyl trichlorosilane (OTS) and controlled heating at 140°C during deposition, ensuring selective deposition exclusively on predefined gold current collectors. The resulting devices exhibited maximum areal capacitance of 2.1 mF cm^{-2} and average of 0.4 mF cm^{-2} with a 2.5 V voltage window, values that, while representing significant progress in miniaturization and spatial resolution, remained in the millifarad per square centimeter range due to inherent limitations in the chemical formulation of the ink and the efficacy of applied thermal treatments. This energy density limitation motivated the intensive search for more efficient active materials and optimized ink formulations in subsequent years.

In 2014 Xu et al. [139], introduced the first systematic approach to hybrid materials through graphene/polyaniline (G/PANI) nanocomposites with precise surfactant control, establishing a new paradigm in ink formulation. Their technical contribution focused on hybrid inks utilizing graphene nanoplatelets (GNP) and polyaniline with sodium dodecyl benzenesulfonate (SDBS) as dispersing surfactant, with controlled rheological parameters: viscosity ~ 1 mPa·s and surface tension 36 mN/m. The fundamental innovation lay in demonstrating that SDBS acted as a molecular spacer between graphene sheets, increasing the specific surface area from $69 \text{ m}^2 \text{ g}^{-1}$ (pure GNP) to $287 \text{ m}^2 \text{ g}^{-1}$ (GNP/PANI), a 4.1-fold increase. The devices achieved specific capacitance of 82 F g^{-1} compared to 10 F g^{-1} for pure GNP, with power density of 124 kW kg^{-1} and excellent cycling stability after 1000 cycles. However, the authors identified the critical phenomenon of “dead surface” generated by excess SDBS surfactant, establishing a fundamental trade-off between stable dispersion.

Collectively, these foundational studies marked important advancements in the inkjet printing of supercapacitors. Chen et al. initially expanded substrate possibilities, Pech et al. refined pattern resolution and selective deposition techniques, and Xu et al. fine-tuned ink formulations and rheological control. Despite the progressive improvements, key challenges persisted, including interface-driven cycle stability issues, efficient thickness control, and the unresolved balance between surfactant dispersion and electrode capacitance.

In 2018, Diao et al. [140], developed robust reduced graphene oxide-polyaniline (RGO@PANI) composites using direct covalent anchoring via EDC/NHS chemistry, thus eliminating surfactant dependence and substantially improving interfacial stability. Their strategy ensured permanent molecular bonding and enhanced synergistic electrochemical properties, achieving volumetric capacitances up to 554 F cm^{-3} in interdigital configurations. This chemical innovation improved mechanical flexibility and maintained electrochemical stability beyond 2000 cycles, though at the cost of increased synthetic complexity associated with covalent chemistry.

By 2019, the focus in inkjet-printed supercapacitor research shifted decisively toward advanced hybrid nanocarbons and MXenes, introducing an advance in chemical sophistication and electrochemical performance. Liu et al. [141] notably employed carbon quantum dots (CQDs) as molecular spacers to mitigate graphene oxide (GO) restacking during solvent evaporation (Figure 7b). These CQDs, derived from KOH-activated fullerene C60, acted effectively as structural pillars, enhancing surface area and introducing additional pseudocapacitive sites. Crucially, their aqueous solubility and oxygen-rich functional groups facilitated surfactant-free, stable colloidal dispersions suitable for inkjet printing. Post-treatment at 200°C under argon improved electrochemical stability, resulting in capacitances of up to 4.2 mF cm^{-2} with consistent flexibility and cycle durability.

The same year, Zhang et al. [31] advanced this trajectory by introducing pristine $\text{Ti}_3\text{C}_2\text{T}_x$ MXenes, capitalizing on their inherently negative electrostatic charges to achieve stable dispersions without additives (Figure 7c). Through minimally aggressive delamination (MILD), they obtained primarily monolayer MXenes exhibiting demonstrated electronic conductivity and high volumetric capacitance. The absence of complex thermal post-processing simplified fabrication. Their inkjet-printed MXene electrodes achieved volumetric capacitances of up to 562 F cm^{-3} , with stable cycle performance and consistent flexibility. Despite these advantages, they faced gradual conductivity degradation due to surface oxidation when exposed to ambient air over extended periods.

Table 3. Representative Studies of Inkjet Printing in supercapacitor Manufacturing: Device Types, Ink Formulations, and Electrochemical Outcomes.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
Solar bifunctional supercapacitor with WO ₃ electrodes [42]	Cyclohexanone, terpineol	Ethyl cellulose	8–15 mPa·s	Dimatix DMP-2800, 140 V, 70μs, 300 °C/30 min	Glass-FTO	Thermal annealing 300 °C/30 min	0.3 mA cm ^{−2} (500 W m ^{−2}); areal capacitance 7.9 mF cm ^{−2} ; stability 500 cycles at 0.0003 A cm ^{−2} (1000 W m ^{−2}) with ~50% retention
Flexible V2CTx film electrode for supercapacitor [22]	N-methyl-2-pyrrolidone (NMP)	None	5.1–7.8 mPa·s	HP Deskjet 2132, 40 V, ambient	Al-coated PET	None	5 mV s ^{−1} ; areal capacitance 5787 μF cm ^{−2} ; 83% retention after 7000 cycles at 75 μA cm ^{−2} ; symmetric device: 23.4 μF cm ^{−2} at 5 mV s ^{−1} ; energy density 0.00034 μWh L ^{−1} ; power 0.01 μW L ^{−1}
Electrochromic device with zinc anode and WNO electrodes [142]	Water/ethylene glycol (70:30)	Triton X-100 (0.3 wt%)	2.6 mPa·s	Microdrop MD-K-130, 188 °C, 8 min grinding	Glass FTO with 3D MXene mesh	Reduction with hypo phosphorous acid (HPA)	0.2 A m ^{−2} ; ≈70 mAh m ^{−2} ; 1000 cycles with 91.2% capacity retention; round-trip energy efficiency 82.13%

Table 3. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
All-inkjet-printed Ti ₃ C ₂ MXene capacitor for textile energy storage [41]	Water (Ti ₃ C ₂ ink), Water/2-propanol (IPA) for electrolyte	Sulphate-nanocellulose (SNC), sodium ascorbate, lithium dodecyl sulphate (LDS)	Ti ₃ C ₂ : 2.7 ± 0.09 mPa·s, Electrolyte: 6.5 ± 0.1 mPa·s	Breva iJet2L printer, 50 °C heated substrate, UV curing for electrolyte	TPU-coated cotton fabric	UV polymerization of electrolyte	50 µA, 0–0.8 V; Areal capacitance 0.86 mF cm ^{−2} (average over 2000 cycles); Coulombic efficiency ≈99.2%; Areal energy 0.08 µWh cm ^{−2} ; Areal power 20 µW cm ^{−2} .
Asymmetric micro-supercapacitor (porous NiS/N-MXene//rGO) [135]	Ethanol, isopropanol	Polyvinylpyrrolidone (PVP)	Not specified	Jet lab II printer, 50 °C substrate, 192 dpi, 80 µm drop interval, 30 layers	PET, photo paper	KOH/PVA gel electrolyte	Volumetric capacitance 429 F cm ^{−3} ; Areal capacitance 16.6 mF cm ^{−2} ; Energy density 33.5 mWh cm ^{−3} at 249 mW cm ^{−3} ; ~80% retention after 10,000 cycles
Micro-supercapacitor (N,S-doped MXene ink) [23]	Ethanol, isopropanol	None (additive-free)	3.02 mPa·s	Jet lab II printer, 45 °C substrate, 25 layers, 300 µm electrode spacing	Photo paper, PET	H ₂ SO ₄ /PVA gel electrolyte	Energy density 8.9 mWh cm ^{−3} at 411 mW cm ^{−3} ; Volumetric capacitance 710 F cm ^{−3} ; 10,000 cycles with 94.6% retention

Table 3. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
Nanoporous carbon micro-supercapacitor [143]	Deionized water	Sulfuric acid, ethanol, urea (for N-doping)	Not specified	Bio Scaffolder 3.2, 150 Hz, 90 μ s, 55 V, 80 °C substrate	Glass	Pyrolysis 900 °C/2 h in argon	5 mV s ⁻¹ ; Areal device capacitance 3.9 mF cm ⁻² ; Volumetric capacitance 151.4 F cm ⁻³ ; Energy density 0.9 mWh cm ⁻³ at 0.4 W cm ⁻³ ; 10,000 cycles with 96% retention
Electric double-layer supercapacitor [18]	Cyclohexanone, terpineol	Ethyl cellulose	Not specified	HP Deskjet 3000, 600 dpi, 300 °C/30 min	Aluminum foil	Thermal annealing 300 °C/30 min	~23 F g ⁻¹ ; Coulombic efficiency 97%; 250 cycles; Capacity 50 nAh
Micro-supercapacitor (Fe-doped MnO ₂ nanosheets) [144]	Water: propylene glycol (1:10)	Triton X-100 (0.06 mg/mL)	1.7 mPa·s	Dimatix DMP-2800, 50 °C substrate, 20 μ m drop spacing, 5 layers	Polyimide	PEDOT:PSS overlayer, annealing at 120 °C	30 μ A cm ⁻² ; Areal capacitance 1.2 mF cm ⁻² ; Volumetric capacitance 9.2 F cm ⁻³ ; Energy density 1.13 mWh cm ⁻³ at 0.11 W cm ⁻³ ; 5200 cycles with 78.7% retention
All-MXene micro-supercapacitor [31]	Aqueous: water; Organic: NMP, DMSO, DMF, ethanol	None	NMP: ~2.2 Pa·s, ethanol: ~2.6 Pa·s, aqueous: 0.71 Pa·s	Dimatix DMP-2800 (FUJIFILM Dimatix, Santa Clara, CA, USA), Voxel8 (extrusion), ambient	AlO _x -coated PET, paper	None	Volumetric capacitance 562 F cm ⁻³ ; ~100% retention after 10,000 cycles; Energy density 0.32 μ Wh cm ⁻² at 11.4 μ W cm ⁻²

Table 3. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
Asymmetric supercapacitor with Ni-Co LDH/Ag/rGO electrodes [145]	Deionized water	Triton X-100, DF69 defoamer, ethylene glycol	Not specified	Jet-lab II, 140 V, −100 V echo, 70 μ s, 50 μ m step	Carbon cloth fabric	Annealing 800 $^{\circ}$ C/2 h N ₂ , then 160 $^{\circ}$ C/1 h	173 mAh g ^{−1} at 1 A g ^{−1} ; 79.8% capacity retention after 5000 cycles at 5 A g ^{−1}
Laser-induced graphene pseudocapacitive electrode [146]	Dimethyl-formamide (DMF)	None	Not specified	Microdrop MD-K-130, 188 $^{\circ}$ C substrate	Laser-induced graphene (LIG)	None	0.1 mA cm ^{−2} ; >97% capacitance retention after 10,000 cycles; Coulombic efficiency $\approx$ 100% after the first 500 cycles
Flexible δ -MnO ₂ micro-supercapacitor [113]	Water: propylene glycol (10:1)	Triton X-100 (0.06 mg/mL)	1.71 mPa·s	Dimatix DMP-2800, 10 pL, 40 μ m spacing, 50 $^{\circ}$ C	Glass, polyimide film	Annealing 350 $^{\circ}$ C/1 h N ₂	0.05 A cm ^{−3} ; Volumetric capacitance 2.4 F cm ^{−3} ; Energy density 1.8×10^{-4} Wh cm ^{−3} at 0.018 W cm ^{−3} ; $\sim$ 88% retention after 3600 cycles
Flexible graphene@ polyaniline nano composite supercapacitor [140]	Cyclohexanone, terpineol	Ethyl cellulose	Not specified	Dimatix DMP-2800, 40 V, 40 $^{\circ}$ C	Flexible gold film	Reduction with HPA	1 mV s ^{−1} ; Volumetric capacitance 554 F cm ^{−3} ; 2000 cycles with >96% retention; Energy density 76.94 Wh cm ^{−3} ; Power density 5593.7 W cm ^{−3}

Table 3. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
All-2D material capacitors (Graphene/hBN/Graphene) [147]	Water-based biocompatible inks	Shear-thinning biocompatible binder	Not specified	Dimatix DMP-2800, ambient conditions	Glass substrates	None specified	Areal capacitance $2.0 \pm 0.3 \text{ nF cm}^{-2}$ ($\sim 3 \text{ }\mu\text{m hBN}$); Dielectric constant 6.1 ± 1.7 ; Breakdown field $1.9 \pm 0.3 \text{ MV cm}^{-1}$
Solid-state flexible supercapacitors using CQDs/GO hybrid ink [141]	Water and ethanol mixture	Carbon quantum dots (CQDs) as nano-spacers	Not specified	HP Deskjet 1110, thermal annealing at $100\text{--}300 \text{ }^{\circ}\text{C}$	A4 printing paper, weighing paper	Thermal annealing (optimized at $200 \text{ }^{\circ}\text{C}$), PVA/ H_2SO_4 gel electrolyte	Areal capacitance 4.2 mF cm^{-2} at 1 mV s^{-1} ; Energy density $0.078 \text{ mWh cm}^{-3}$ at 0.28 mW cm^{-3} ; 10,000 cycles with 83% retention
All-inkjet-printed solid-state flexible supercapacitors on paper [148]	Water (SWNT/AC), Water/IPA (1:1, Ag NW)	SDBS (1.0 wt%), CNF primer layer	SWNT/AC: $\sim 20 \text{ cP}$, Ag NW: $\sim 18 \text{ cP}$, CNF: $\sim 18 \text{ cP}$	HP Deskjet 1010, $60 \text{ }^{\circ}\text{C}$ platen, UV curing for electrolyte	A4 paper with CNF nanomat primer layer	UV-curing of [BMIM][BF ₄]/ETPTA electrolyte	0.2 mA cm^{-2} ; $\sim 100 \text{ mF cm}^{-2}$ for $>10,000$ cycles; Coulombic efficiency $\sim 100\%$
All-solid-state micro-supercapacitor (GO + pen ink hybrid) [149]	Water, ethylene glycol	Commercial pen ink (graphite carbon nanoparticles)	Not specified	Dimatix DMP-2800, ambient, 5 passes	PET	HI vapor reduction, $150 \text{ }^{\circ}\text{C}/2 \text{ h}$	$0.1 \text{ }\mu\text{A}$ (GCD); Areal capacitance $19.18 \text{ }\mu\text{F cm}^{-2}$; $\sim 100\%$ retention after 10,000 cycles
All-solid-state asymmetric micro-supercapacitor ($\text{K}_2\text{Co}_3(\text{P}_2\text{O}_7)_2 \cdot 2\text{H}_2\text{O}$ // graphene) [150]	Ethanol	PTFE (10 wt%), acetylene black	Not specified	Fujifilm Dimatix 3000, vacuum oven $100 \text{ }^{\circ}\text{C}$, 3 layers	PET with inkjet-printed Ag current collectors	KOH/PVA gel electrolyte, dried in air	10 mA cm^{-3} ; 6.0 F cm^{-3} ; 5000 cycles with 94.4% retention; Energy density 0.96 mWh cm^{-3} ; Power density 54.5 mW cm^{-3}

Table 3. Cont.

Device Type	Solvent	Additives	Viscosity	Printing Conditions	Substrate	Post-Treatment	Performance Results
MWCNT/Ag nanoparticle based asymmetric supercapacitors [151]	Water	Sodium dodecylbenzene sulfonate (SDBS), MnO ₂ nanoparticles	Not specified	HP Deskjet 1010, ambient conditions	Paper substrate	4 M LiCl electrolyte assembly	1.8 V window; Energy density 1.28 mWh cm ⁻³ at 96 mW cm ⁻³ ; 96.9% retention after 3000 cycles
Supercapacitor electrodes with graphene/PANI [139]	Water	SDBS surfactant	~1 mPa·s	Inkjet printing at room temp, 86 V, pulse 40 ms	Carbon fabric	Drying at 80 °C for 2 h	20 mV s ⁻¹ ; Specific capacitance 82 F g ⁻¹ ; Energy density 2.4 Wh kg ⁻¹ at 124 kW kg ⁻¹ ; 1000 cycles (stable)
Carbon-based micro-supercapacitors with interdigital configuration [138]	Ethylene glycol	PTFE polymer binder (5 wt%), Triton X100 surfactant	Not specified	AltaDrop equipment, 140 °C substrate temperature, annealing 240 °C	Silicon with 150 nm SiO ₂ , Ti/Au electrodes (150 Å Ti, 300 nm Au)	Thermal annealing at 240 °C	1 mV s ⁻¹ ; Areal capacitance 2.1 mF cm ⁻² (2.5 V); Areal energy 0.00183 mWh cm ⁻² (6.6 mJ cm ⁻²); Volumetric energy ≈2.34 mWh cm ⁻³ ; Areal power 44.9 mW cm ⁻²
SWNT/RuO ₂ nanowire supercapacitors on cloth and flexible substrates [137]	Water with SDS	Sodium dodecyl sulfate (SDS, 1 wt%)	~20 cP after centrifugation	Epson Artisan 50 piezoelectric printer, 1440 × 1440 dpi	PET sheets, cloth fabrics, SiO ₂ /Si substrates	PVA/H3PO4 gel electrolyte preparation	8 A g ⁻¹ ; Specific capacitance 138 F g ⁻¹ ; Coulombic efficiency >99%; Energy density 18.8 Wh kg ⁻¹ ; Power density 96 kW kg ⁻¹

Critically, each of these advancements embodied specific trade-offs in material design and processing. Liu's CQD-based approach excelled in mechanical robustness and stability but diluted active material density, thus limiting volumetric capacitance. In contrast, Zhang's MXene formulations prioritized maximum energy density and conductivity, though the simplified processing limited the functionalization of surface groups and long-term environmental stability. Diao's method represented an intermediate balance, integrating covalent anchoring to enhance performance while accepting the complexity of chemical synthesis.

Overall, these studies illustrated a systematic progression where each technological breakthrough strategically addressed previous limitations. The evolution from carbonaceous modifications toward inherently active two-dimensional materials progressively eliminated surfactants and simplified post-processing, setting a foundation for scalable manufacturing. The significant advances in volumetric capacitance from approximately 100 F cm^{-3} in early graphene-based materials to over 500 F cm^{-3} in MXenes highlighted the substantial potential of advanced two-dimensional materials for next-generation supercapacitor technologies.

The period from 2022 to 2023 marked a phase in the technological maturation of inkjet-printed supercapacitors, characterized by systematic optimization in chemical doping, multilayer architectures, and asymmetric geometries. Researchers transitioned from proof-of-concept validations toward textile and wearable applications, standardizing printing parameters and refining post-treatment strategies.

Sun et al. [23] emphasized antioxidation and cyclic stability by introducing N,S co-doping into MXenes, as reported in 2022, achieving a 94.6% capacitance retention after 10,000 cycles. Their approach employed a dual-step treatment involving high-temperature annealing under nitrogen followed by solvothermal processing with thiourea, allowing precise integration of nitrogen and sulfur atoms (Figure 7d). This significantly enhanced environmental oxidation resistance and electronic conductivity, although the complex synthetic conditions posed challenges for scalable production.

Building upon these insights, Sun et al. [135], expanded the energy density and operating voltage window through an innovative asymmetric electrode design comprising porous NiS/N-MXene and reduced graphene oxide (rGO). This architecture increased the operational voltage window dramatically from the conventional 0.6 V to 1.5 V. Critical to this design was the solvothermal synthesis under protective hydrazine conditions, effectively mitigating oxidation during electrode preparation. Despite the increased complexity required to balance electrolyte compatibility and electrode mass ratios, their configuration achieved an impressive energy density of 33.5 mWh cm^{-3} , representing an improvement over previous designs.

Simultaneously, Gibertini et al. [41] integrated supercapacitors directly onto textile substrates, employing a UV-curable polymer gel electrolyte. Utilizing a cotton substrate coated with thermoplastic polyurethane (TPU) and optimizing the polymer electrolyte formulation (LiCl/EG/AM/MBA), they successfully printed fully integrated textile supercapacitors (Figure 7e). The UV-curing process significantly simplified manufacturing by eliminating aggressive thermal treatments incompatible with textiles. While demonstrating commendable mechanical compatibility, the lower ionic conductivity (0.60 mS/cm) and moderate cyclic stability (61.7% retention after extensive mechanical flexing) highlighted inherent trade-offs between flexibility, electrochemical performance, and durability.

Printing and post-treatment parameters were progressively standardized across these studies, notably optimizing substrate temperature (45°C to 50°C) to enhance solvent evaporation and ink adhesion. The number of printed layers also increased systematically to balance resistance and flexibility, while post-treatments diversified according to spe-

cific applications from thermal gelation for enhanced capacitance to UV-curing processes tailored for textile compatibility.

Quantitative performance metrics across these efforts revealed clear trade-offs. Capacitance and energy density improved markedly through advanced doping and asymmetric electrode designs, yet cyclic stability showed gradual declines under increasingly demanding mechanical conditions. Gibertini's textile-integrated approach, although practical for wearable applications, faced significant limitations including lower areal capacitances and mechanical durability challenges under repeated flexure.

Critical analysis highlighted the persistent challenge of electrolyte sealing, with gradual water evaporation and structural degradation limiting practical lifetimes. Additionally, textile adhesion under variable environmental conditions required further validation, alongside systematic toxicological evaluation of MXene-polymer formulations for wearable use. Scalability also emerged as a persistent barrier, necessitating improved quality control in multilayer composite synthesis and reproducibility in post-print treatments.

The period from 2024 to 2025 reflects a shift toward intelligent multifunctional devices, moving beyond traditional energy storage paradigms. This cutting-edge phase is defined by integrating photochemical, electrochemical, and electrochromic functionalities into flexible, scalable systems, enabling potential applications in smart windows, wearable electronics, and integrated energy systems.

In 2024, Qiu et al. [142] expanded the functionality into smart window applications by developing supercapacitors with electrochromic capabilities based on heterogeneous $W_{17}O_{47}/Na_{0.1}WO_3$ nanowires. This novel integration achieved optical modulation of 69.13% and rapid color-switching kinetics in approximately 4–5 s, coupled with stable electrochemical performance of 70 mAh cm^{-2} over 1000 cycles. The carefully optimized $Zn(ClO_4)_2$ electrolyte and three-dimensional MXene substrates laid a solid foundation for practical architectural applications (Figure 7f). However, despite these promising results, the limited cycle stability and the need for specialized equipment present barriers to widespread commercial adoption.

A bifunctional device combining energy storage and photochemical reactions was developed by Sangiorgi et al. [42], utilizing WO_3 electrodes capable of simultaneous solar absorption and charge storage. Their hybrid screen-printing and inkjet decoration approach yielded a significant increase in areal capacitance under illumination (1.6 mF cm^{-2} versus 0.7 mF cm^{-2} in darkness). The symmetrical device configurations reached 7.9 mF cm^{-2} at optimal light intensity tested at 0.3 mA cm^{-2} (500 W m^{-2}), signifying a step toward fully integrated photo-rechargeable systems. Despite the innovative nature of this approach, it still faces limitations such as restricted spectral absorption due to the WO_3 bandgap (2.4–2.7 eV) and lower ionic conductivity of the agar-agar gel electrolyte compared to conventional aqueous formulations.

Parallely, Ji et al. [22], introduced practical flexible electrodes using V_2CT_x MXene inkjet-printed onto PET substrates, emphasizing low-cost and straightforward fabrication compatible with standard commercial printers. Additionally, they used N-methyl-2-pyrrolidone (NMP) as the solvent to minimize the “coffee-ring” effect, exploiting the lower molecular weight of V_2CT_x relative to other MXenes. The resulting electrodes demonstrated stable performance, achieving areal capacitances of $531.3\text{--}5787.0 \text{ }\mu\text{F cm}^{-2}$, high optical transparency (42–87% at 550 nm), and capacitance retention of 83% after 7000 cycles. Despite these advancements, challenges persist regarding solvent environmental impact, complex MXene synthesis involving hazardous chemicals, and protection requirements against environmental oxidation.

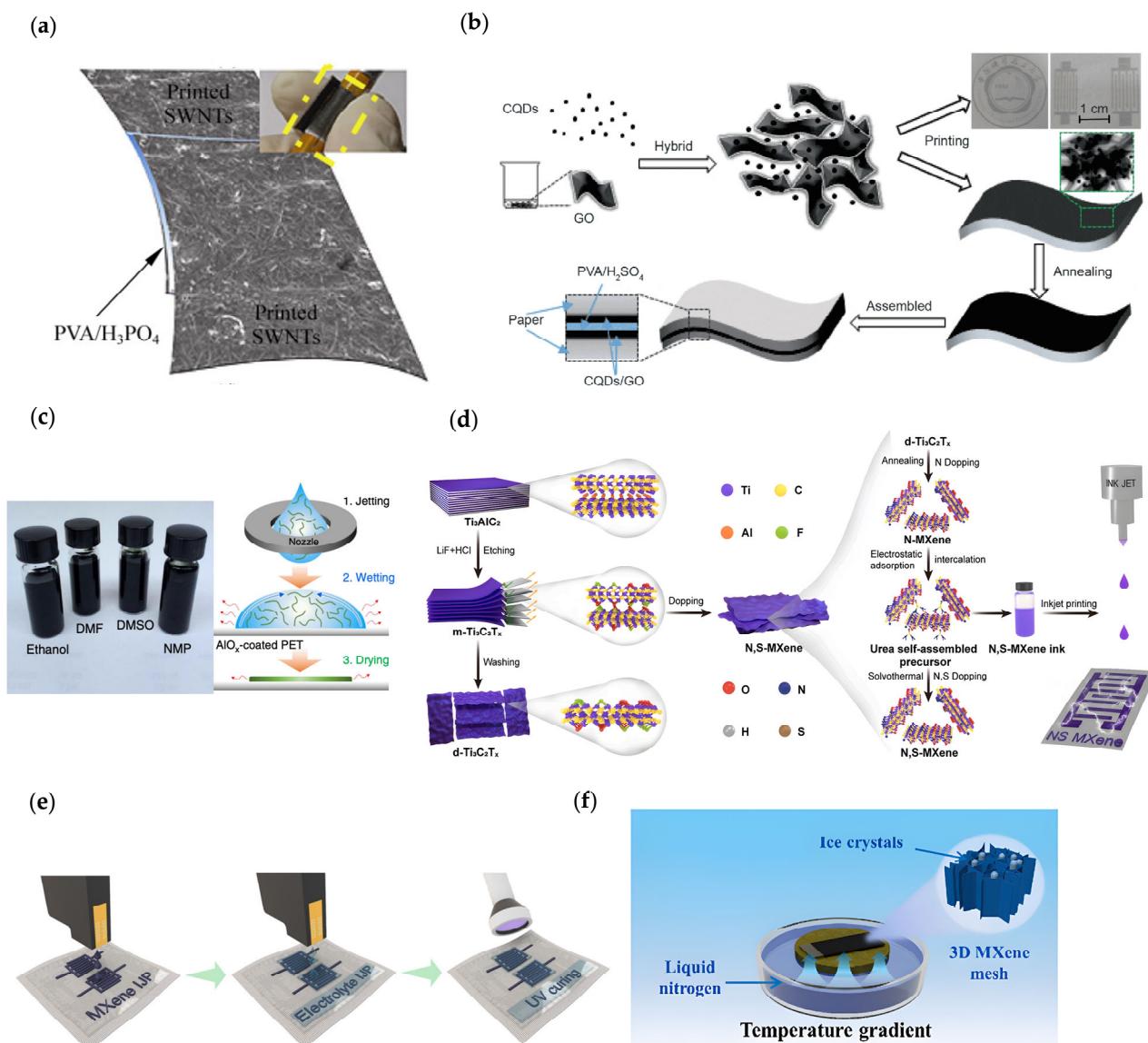


Figure 7. (a) Schematic of an inkjet-printed single-walled carbon-nanotube (SWNT) supercapacitor on cotton fabric with a PVA/ H_3PO_4 gel electrolyte; photograph of the flexible device wrapped around a pencil [137]. (b) Schematic illustration of the inkjet-printing process of carbon quantum dots (CQDs)/graphene oxide (GO) hybrid ink on paper substrate for flexible supercapacitors [141]. (c) Photographs of additive-free organic and aqueous $\text{Ti}_3\text{C}_2\text{T}_x$ MXene inks (left) and schematic illustration of the inkjet and extrusion printing process on flexible substrates (Right) [31]. (d) Schematic illustration of the synthesis and inkjet printing process of nitrogen and sulfur co-doped $\text{Ti}_3\text{C}_2\text{T}_x$ MXene inks for fabrication of micro-supercapacitors [23]. (e) Schematic representation of the all-inkjet-printed Ti_3C_2 MXene symmetrical capacitor fabrication process on TPU-coated cotton textile substrate [41]. (f) Schematic illustration of the 3D interconnected MXene conductive network deposited on fluorine-doped tin oxide (FTO) glass substrate, designed to enhance charge transfer and ion transport in electrochromic devices [142].

The operational parameters critical to these technologies varied significantly, with device-specific voltage windows requiring careful optimization for optimal performance. While electrochromic devices operated efficiently within narrow voltage ranges, V_2CT_x -based supercapacitors utilized broader operational windows, highlighting the importance of tailored electrical control schemes. Additionally, mechanical adhesion between printed layers emerged as a critical challenge, particularly for flexible devices subject to repeated mechanical deformation.

Despite significant advancements in material design and multifunctionality, inkjet-printed supercapacitors remain far from industrial adoption. Persistent obstacles, including mechanical delamination at printed interfaces, electrolyte evaporation under realistic operational conditions, and the intricate balance between advanced chemistries (such as carbon quantum dots or MXenes) and scalable roll-to-roll manufacturing, undermine many proof-of-concept achievements. Additionally, reliance on toxic or energy-intensive solvents and specialized laboratory printers raises critical concerns regarding environmental impact, reproducibility, and economic feasibility.

To advance beyond academic demonstrations, the field must first comprehensively elucidate fundamental degradation mechanisms experienced by devices under mechanical and chemical stresses. Researchers should move beyond merely reporting capacity retention and instead correlate electrochemical degradation with physical damage. For instance, conducting *in situ* scanning electron microscopy (SEM) during repeated mechanical bending cycles (e.g., 1000 cycles at a 5 mm radius) or aging cells under controlled environmental conditions (85% relative humidity and 60 °C) would enable precise tracking of impedance changes. This approach could effectively identify specific degradation pathways, such as MXene oxidation, binder decomposition, or delamination, facilitating targeted interventions.

Moreover, it is crucial to harmonize fabrication standards across research platforms. Currently, groups utilize different equipment, such as the Dimatix DMP-2800 (FUJIFILM Dimatix, Santa Clara, CA, USA; operating at 140 V with 70 μ s pulses and a substrate temperature of 50 °C) versus the HP Deskjet 2132 (HP Inc., Palo Alto, CA, USA; operating at 40 V under ambient conditions) yet publish results without fully disclosing printing protocols. Establishing a standardized “printing profile”—clearly specifying ink viscosity (e.g., 2.7 ± 0.1 mPa·s), pulse voltage (30–40 V), droplet spacing (20–50 μ m), and post-treatment procedures (e.g., annealing at 200 °C for 30 min under argon) would greatly enhance comparability and reproducibility across different laboratories.

Finally, the research community must actively transition toward greener solvent systems and adopt continuous processing technologies to ensure environmental sustainability and industrial scalability. Strategies such as replacing hazardous solvents like N-methyl-2-pyrrolidone (NMP) with environmentally benign water/ethanol mixtures (70:30 ratio) or biodegradable ester-based solvents, incorporating natural dispersants such as functionalized lignin, and validating scalable slot-die roll-to-roll coating methods at production speeds (e.g., 1 m/min) are essential for demonstrating real-world practicality. Additionally, implementing solvent recovery loops and comprehensive life-cycle assessments would ensure eco-friendly formulations perform reliably without imposing hidden environmental costs.

Only by addressing these interconnected challenges, clarifying degradation mechanisms, standardizing manufacturing protocols, and adopting sustainable production practices, inkjet-printed supercapacitors can transition from laboratory-scale studies to reliable, cost-effective components ready for integration into future energy storage solutions.

8. Conclusions

Over the past two decades, inkjet printing has emerged as a promising technique for fabricating energy storage devices such as batteries and supercapacitors. This review has surveyed the field from colloidal fundamentals and rheological control to multilayer and interdigital electrode architectures. Inkjet printing offers contactless deposition, digital design freedom, and compatibility with fragile substrates; however, achieving high performance requires careful tuning of particle size, solid loading, viscosity, and dynamic surface

tension. These parameters govern droplet formation and spreading, which ultimately determine electrode morphology and electrochemical behavior.

For lithium-ion systems, inkjet-printed LiFePO_4 and LiCoO_2 cathodes typically deliver initial capacities around $130\text{--}150\text{ mAh g}^{-1}$ when formulated with carbon black and polymeric binders. Multilayer approaches have increased electrode thickness and areal capacity without compromising structural integrity, while modified current collectors such as MXenes provide flexibility and improved conductivity. Fully printed aqueous zinc–manganese dioxide batteries on paper exemplify the technology’s potential for wearable electronics, delivering 300 mAh g^{-1} at 200 mA g^{-1} and energy densities of 330 Wh kg^{-1} with retention at high power.

For supercapacitors, concentrated graphene or MXene inks yield printed micro-devices with volumetric capacitances typically in the mid-hundreds ($\approx 500\text{--}700\text{ F cm}^{-3}$) and cycle stability exceeding 97% after 10,000 cycles, while advanced MXene composites achieve energy densities from 9.8 to 33.5 mWh cm^{-3} . By consolidating reported performance data and elucidating the interplay between ink formulation and printability, this review highlights inkjet printing as a pathway to sustainable, low-waste manufacturing of energy storage components. The importance of balancing rheological stability against nozzle clogging and integrating conductive binders or surface treatments to enhance electrode integrity is underscored. Such insights should inform process optimization and facilitate the translation of laboratory demonstrations to practical devices.

Despite notable progress, challenges remain. Low solids loading, nozzle clogging, and the need for multiple printing passes limit areal capacity and throughput. Mechanical durability under bending and long-term electrochemical stability also require further study. Future research should explore greener solvent systems, oxidation-resistant metallic inks, and multi-material architectures, alongside improved voltage-waveform design and in situ monitoring to control droplet dynamics. Hybrid additive manufacturing strategies and roll-to-roll processing could unlock higher throughput, while extending inkjet techniques to sodium-ion, zinc-ion, and solid-state chemistries may broaden their applicability. Addressing these gaps will be crucial for realizing the full potential of inkjet-printed energy storage devices.

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Abbreviations

The following abbreviations are used in this manuscript:

Abbreviation	Definition	Abbreviation	Definition
AC	Activated Carbon	LTO	Lithium Titanate Oxide
AM	Acrylamide	Li-PAA	Lithium Polyacrylic Acid
Ag NW	Silver Nanowires	LiAlO ₂	Lithium Aluminate
AlPO ₄	Aluminum Phosphate	LiCl	Lithium Chloride
Al ₂ O ₃	Aluminum Oxide	LiFePO ₄	Lithium Iron Phosphate
CAGR	Compound Annual Growth Rate	LNMO	Lithium Nickel Manganese Oxide
CB	Carbon Black	mAh g ^{−1}	milliampere hours per gram
CHP	Cyclohexylpyrrolidone	mAh cm ^{−2}	milliampere hours per square centimeter
CIJ	Continuous Inkjet	mF cm ^{−2}	millifarads per square centimeter
CMC	Carboxymethyl Cellulose	mPa·s	millipascal seconds
CNF	Carbon Nanofiber	mV s ^{−1}	millivolts per second
CNT	Carbon Nanotube	mW cm ^{−3}	milliwatts per cubic centimeter
CQDs	Carbon Quantum Dots	mWh cm ^{−3}	milliwatt hours per cubic centimeter
DI	Deionized	MWCNT	Multi-Walled Carbon Nanotube
DLS	Dynamic Light Scattering	MXene	M stands for early transition metal, X for C or N
DMF	Dimethylformamide	NMP	N-methyl-2-pyrrolidone
DMSO	Dimethyl Sulfoxide	PAA	Poly(acrylic acid)
		PANI	Polyaniline
DoD	Drop-on-Demand	PET	Polyethylene Terephthalate
dpi	dots per inch	PIJ	Piezoelectric Inkjet
EDC/NHS	1-Ethyl-3-(3-dimethylaminopropyl) carbodiimide/N-Hydroxy succinimide	PLAL	Pulsed Laser Ablation in Liquid
EDX	Energy-Dispersive X-ray Spectroscopy	PTFE	Polytetrafluoroethylene
EG	Ethylene Glycol	PVA	Polyvinyl Alcohol
EHD	Electrohydrodynamic	PVB	Polyvinyl Butyral
ETPTA	Ethoxylated Trimethylolpropane Triacrylate	PVDF	Polyvinylidene Fluoride
F cm ^{−3}	farads per cubic centimeter	PVP	Polyvinylpyrrolidone
F g ^{−1}	farads per gram	rGO	reduced Graphene Oxide
FTO	Fluorine-doped Tin Oxide	RFID	Radio Frequency Identification
GNP	Graphene Nanoplatelets	RPM	revolutions per minute
GO	Graphene Oxide	SBR	Styrene-Butadiene Rubber
GSM	grams per square meter	SDBS	Sodium Dodecyl Benzene Sulfonate
hBN	hexagonal Boron Nitride	SDS	Sodium Dodecyl Sulfate
HP	Hewlett-Packard	SEM	Scanning Electron Microscopy
HPA	Hypo Phosphorous Acid	SIJ	Super-fine Inkjet
H ₂ SO ₄	Sulfuric Acid	SWNT	Single-Walled Carbon Nanotube
H ₃ PO ₄	Phosphoric Acid	TIJ	Thermal Inkjet
IJP	Inkjet Printing	TMOS	Tetramethyl Orthosilicate
IPA	Isopropanol	TPU	Thermoplastic Polyurethane
IoT	Internet of Things	μF cm ^{−2}	microfarads per square centimeter
KOH	Potassium Hydroxide	μm	micrometers
kW kg ^{−1}	kilowatts per kilogram	μWh cm ^{−2}	microwatt hours per square centimeter
		μW cm ^{−2}	microwatts per square centimeter
LDH	Layered Double Hydroxide	V	Volts
LDS	Lithium Dodecyl Sulfate	W kg ^{−1}	watts per kilogram
LFP	Lithium Iron Phosphate	Wh kg ^{−1}	watt hours per kilogram
LIB	Lithium-Ion Battery	Wh L ^{−1}	watt hours per liter

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