Dry Printing Fully Functional Eco-Friendly Transient Papertronics

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Abstract:

The demand for flexible printed electronics is growing fast, especially with the move toward the Internet of Things (IoT). These printed electrons are usually designed for short-term use, after which they are disposed of. The polymeric substrates used in printed electronics comprise the biggest portion of their non-biodegradable E-waste after their disposal. This paper demonstrates the feasibility of printing fully functional transient electronics on flexible, water-soluble, and biodegradable paper substrates using the dry printing approach. The in-situ generation and real-time sintering of silver nanoparticles at room temperature enables the fabrication of complex circuits on such water-soluble papers. A layout similar to an Arduino pro mini board is printed on both sides of a paper substrate with electrical interconnects. Various electrical components are then directly mounted to fabricate a complete, working paper Arduino circuit. Cyclic bending tests demonstrate the mechanical durability and reliability of printed paper circuits under repeated bending stress. The process uniquely achieves robust and complex printed electronics without thermal damage, and the water solubility tests successfully show rapid dissolution of the paper devices in water. Furthermore, the components detached during dissolution are collected and reused, demonstrating the recyclability of the process. Overall, this transformative manufacturing method establishes key technical capabilities to produce next-generation sustainable, green electronics and sensors using renewable materials.

Keywords: Papertronics, transient electronics, printed electronics, additive nanomanufacturing, dry printing, biodegradable electronics.

Introduction

Additive manufacturing (AM) has seen rapid advancements in recent years, with important implications for electronics and the Internet of Things (IoT) [1-3]. As more things and devices become interconnected, there is growing interest in techniques that enable flexible and customized fabrication directly onto various surfaces [4]. However, conventional electronics manufacturing is wasteful, using harsh chemicals as well as generating substantial non-biodegradable e-waste that persists in landfills [5-9]. This highlights the need for more sustainable approaches in the manufacturing of electronics in the future, especially for applications where the electronics are designed to be short-lived. Printing functional circuits on water-soluble and biodegradable paper substrates is a promising solution, allowing electronics and sensors to be safely disposed of after use [10]. Such water-soluble papertronics could have diverse applications ranging from disposable biosensors [11-14] to transient electronics for security, military, and aerospace systems [15, 16]. The ability to directly print conductive traces onto these substrates using dry additive processes eliminates issues with ink compatibility and enables precise, layer-by-layer patterning at atmospheric conditions [17].

Various printing techniques have been explored for fabricating electronics on paper substrates [18-22], including inkjet printing (IJP) [23-25], aerosol jet printing (AJP) [26, 27], and screen printing [28-31]. However, these liquid-based approaches face considerable challenges when printing on porous, water-soluble papers [16]. For instance, Balliu et al. [23] discussed issues with ink compatibility and paper burning during laser sintering. The capillary wicking of papers also causes inks to spread uncontrollably [32], making precise patterning difficult. Surfactants and additives are required in inks to control surface tension and viscosity, but these contaminate papers [33]. Furthermore, post-printing thermal annealing is often necessary to remove solvents and enhance conductivity[34], but temperatures above 100-150°C cause irreparable damage to most papers [28]. To make the paper compatible with such ink-based printing, people have tried using waxed papers or laminated papers, both of which significantly undermine the biodegradability advantages of papers. These challenges have greatly hindered the development of functional circuits on water-soluble substrates [32]. Achieving reliable electrical performance under folding and humidity without proper encapsulation has proven problematic as well [33].

Recently, dry additive nanomanufacturing (Dry-ANM) has introduced an entirely new approach to printing functional electronics on various substrates [35]. This technique relies on the in-situ and on-demand generation of pure nanoparticles inside the printer head using laser ablation of solid targets. These nanoparticles are then directed onto the surface of the substrates through a nozzle and simultaneously sintered by a secondary laser in real time [36]. This process allows liquid and contaminant-free printing at atmospheric conditions directly onto porous untreated papers [16]. Unlike liquid-based methods, this dry printing approach offers exceptional control over the electrical and mechanical properties of printed conductors by tuning the laser sintering parameters [16, 37]. Such tuning eliminates the need for post-processing steps like thermal annealing. Furthermore, the dry nanoparticles are compatible with water-soluble substrates, as verified by the rapid disposability of printed traces [16]. By eliminating process contaminants and the constraints around ink development/optimization, this dry printing technique provides a versatile platform for next-generation green electronics and sensors [16].

Table 1 compares the dry printing method with other printing techniques, highlighting the unique advantages of this approach for fabricating transient papertronics. Unlike inkjet printing (IJP) and aerosol jet printing (AJP) methods, which require the use of solvents or additives in the ink formulation, the Dry-ANM technique utilizes solid bulk silver as the feedstock, eliminating the need for ink preparation. This not only simplifies the printing process but also reduces the environmental impact associated with the ink manufacturing processes, such as the use of chemicals and solvents. Furthermore, the Dry-ANM method enables direct printing on untreated water-soluble paper substrates, which is challenging for liquid-based printing techniques. The real-time laser sintering of the deposited silver nanoparticles eliminates the need for post-processing steps such as thermal sintering or chemical treatment, making the process more environmentally friendly and compatible with temperature-sensitive substrates.

Table 1. Comparison of various printing techniques for fabricating conductive silver traces on paper substrates.

Substrate	Input materials	solvent/additive	Printing method	Post-processing method	Ability to print on water-soluble paper	Resistivity ($\Omega\cdot$m)	Ref.
Glossy photo paper	Silver nitrate	Polyacrylic acid, triethanolamine, ethylene glycol, deionized water	IJP	Thermal sintering (60 ° C)	No	9×10^{-8}	[38]
Glossy photo paper	Silver nitrate	Polyacrylic acid, ethylene glycol, 1-propanol, water	IJP	Thermal sintering (50 ° C)	No	8.9×10^{-8}	[39]
Cellulose fiber paper	Silver flake ink	Not mentioned	AJP	Hot-air sintering (80 ° C)	No	2.4×10^{-7}	[40]
Nanofibrillated cellulose (NFC) paper	Silver nitrate	Sodium nitrate, trisodium citrate, water	optical printing	Rinsing with DI water, NaCl solution, drying with a nitrogen gun	No	9.7×10^{-8}	[18]
Regular copy paper	Solid bulk silver	Not needed	Dry-ANM	Not needed	Yes	2×10^{-7}	This study

This paper demonstrates the feasibility of developing eco-friendly transient electronic devices using biodegradable and water-soluble materials. The ability to print on water-soluble paper and the compatibility with transient electronics applications highlight the unique advantages of the Dry-ANM technique for green and sustainable electronics manufacturing. The optimized low-temperature nanoparticle sintering enables mechanically robust flexible papertronics without thermal degradation. We further show the performance, disposability, and recyclability of the printed papertronics using water-soluble substrates.

Methods

In this study, we have utilized the additive nanomanufacturing method recently developed by the authors [16, 35-37]. Briefly, this dry printing process comprises a nanoparticle generation chamber, a nozzle, a target, a pulsed laser for nanoparticle generation, a gas flow system, and a secondary laser for nanoparticle sintering onto substrates (**Figure 1a**). Commercially available, high-purity silver target was ablated using a pulsed laser (1064 nm wavelength) to generate a plume of silver nanoparticles in background argon gas. These nanoparticles exited through the nozzle by the argon carrier gas flow and were directed toward the paper substrate mounted on an XY stage. A separate nanosecond laser simultaneously sintered the arrived nanoparticles along pre-programmed paths to print desired silver trace patterns. To demonstrate the applicability of the approach with printing complex functional electronics circuits and functionality, we chose an Arduino (**Figure 1b)** circuit for demonstration.

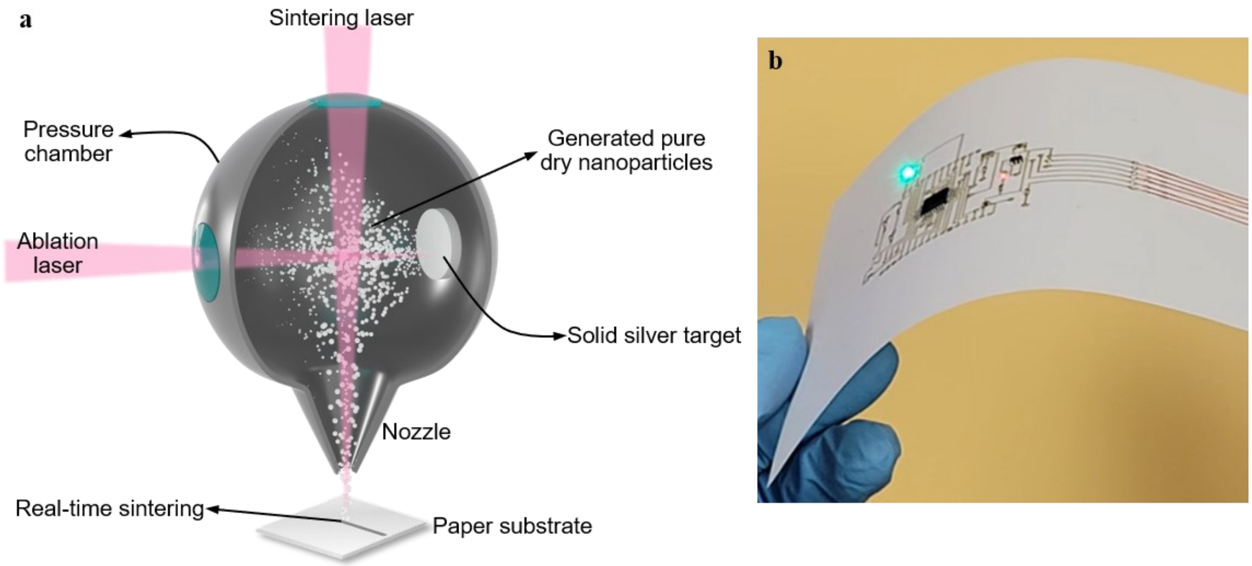

Figure 1. (**a**) Schematic illustration of the Dry-ANM system. (**b**) Demonstration of working paper Arduino.

Results and Discussions

The Arduino circuit was designed to demonstrate the compatibility and reliability of our approach with printing functional electronic devices on biodegradable and water-soluble paper substrates. The design comprised printed silver traces in a layout similar to a commercial Arduino Pro mini board, which were printed on both the back and front sides of the paper. Various electrical components, including a microcontroller (ATmega328P), crystal oscillator, LEDs, resistors, and capacitors, were then directly mounted onto the printed traces.

The fabrication steps and characterization of the printed papertronics are illustrated in **Figure 2**. Silver traces were first printed on both sides of the paper (**Figure 2a** and **2b**). Next, via holes were created using a 400 μm needle to connect the front and back traces (**Figure 2c**). Silver epoxy was used to interconnect both sides through the holes (**Figure 2d**) as well as attach the components to the printed traces (**Figure 2e**). The epoxy was allowed to cure at room temperature for 1 hour to achieve mechanical and electrical connectivity. In addition to the epoxy, a small drop of super glue helped reinforce the components in position.

Key printing process parameters for silver nanoparticle generation included a laser pulse energy of 1 mJ with a pulse repetition rate of 30 kHz and a chamber argon pressure of 5 PSIG. The secondary sintering laser power density was ~0.7 MW/m^2, and the printing speed was 4 mm/s, using a nozzle diameter of 260 microns. The electrical performance of the printed lines was evaluated by measuring their resistance over multiple pass counts, as shown in **Figure 2f**. The conductivity increased with the number of printed layers, indicating the formation of more robust and interconnected sintered silver nanoparticle networks. The corresponding optical microscopy image is shown in the inset of **Figure 2f**. Based on the optimum resistance results, the number of printed layers was adjusted to 20 passes as no significant change of resistance was observed for passes more than 20.

3D profilometry was employed further to characterize the thickness of the printed silver lines. **Figure 2g** shows a 3D scan of a printed silver line, revealing its surface topography and uniformity. The cross-sectional height profile extracted from the profilometry data (**Figure 2h**) provides a rough estimate of the thickness and surface roughness of the printed trace. These characterizations help optimize the printing parameters and understand the relationship between the printed structure and its electrical performance. Authors previously reported detailed cross-sectional characterization and density studies of printed lines on paper [16]. By using these characterizations, the resistivity of the silver lines for 20 passes iteration was calculated to be 1.5×10^{-7} Ω·m in this work, about an order of magnitude higher than the bulk silver.

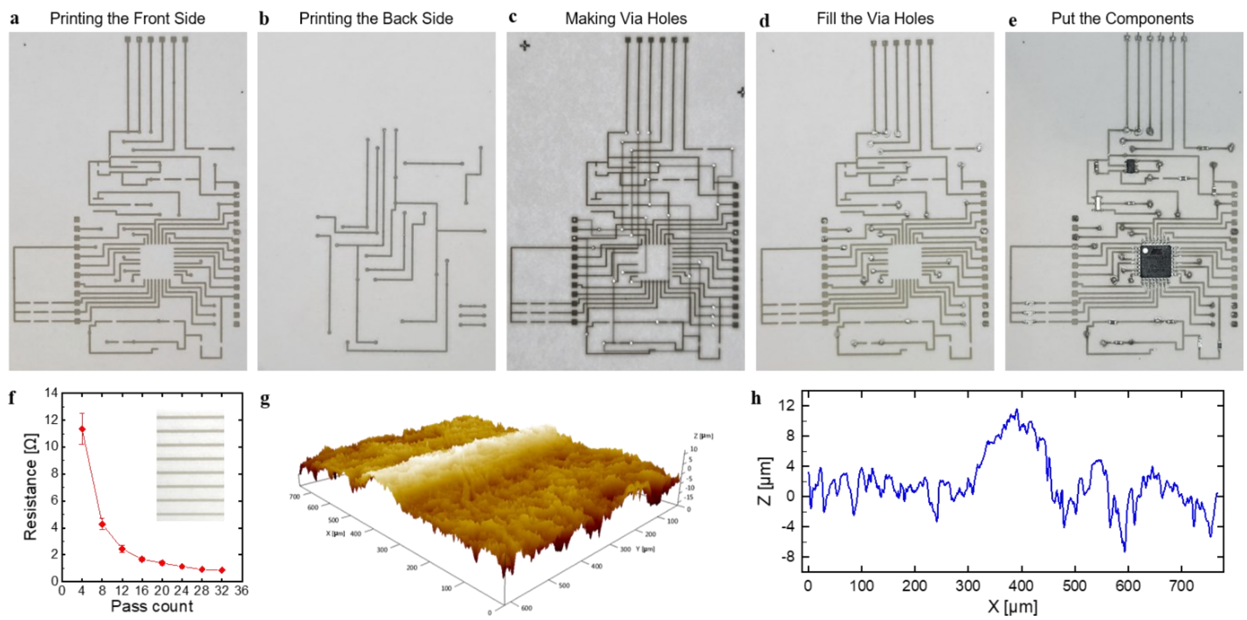

Figure 2. Fabrication process and characterization of the printed papertronics. (**a**) Initial printed circuit layout on the front side; (**b**) Initial printed circuit layout on the back side; (**c**) Created via holes using a needle (the picture was taken with backlight on to visualize both sides of the paper simultaneously); (**d**) the via holes are filled with silver epoxy; (**e**) Mounted components; (**f**) Resistance performance graph demonstrating the conductivity of printed lines over multiple printed pass counts (i.e., layers). The inset is the corresponding optical microscopy image of the printed silver traces with different pass counts; (**g**) a 3D profilometry scan of a printed line; and (h) a cross-sectional height profile from the profilometry data.

The multiscale optical and scanning electron microscopy (SEM) images of the dry printing and via formation/filling processes for fabricating the circuits on paper substrates are presented in **Figure 3**. The top row (**Figure 3a-3d**) shows optical microscope images of the key creation steps via interconnects. First, a silver (Ag) line is printed on the paper surface (**Figure 3a**), followed by the formation of a circular Ag pad (**Figure 3b**). Next, a via hole is precisely created through the paper on the pad using a needle (**Figure 3c**), and then filled with Ag epoxy (**Figure 3d**) to establish electrical and mechanical connections between the top and bottom printed traces. The middle row (**Figure 3e-3h**) displays SEM images of the corresponding steps, revealing the morphological changes of the printed features. The SEM images in the bottom row (**Figure 3i-3k**) provide a closer look at the structure of the sintered Ag traces and the integration with the Ag epoxy.

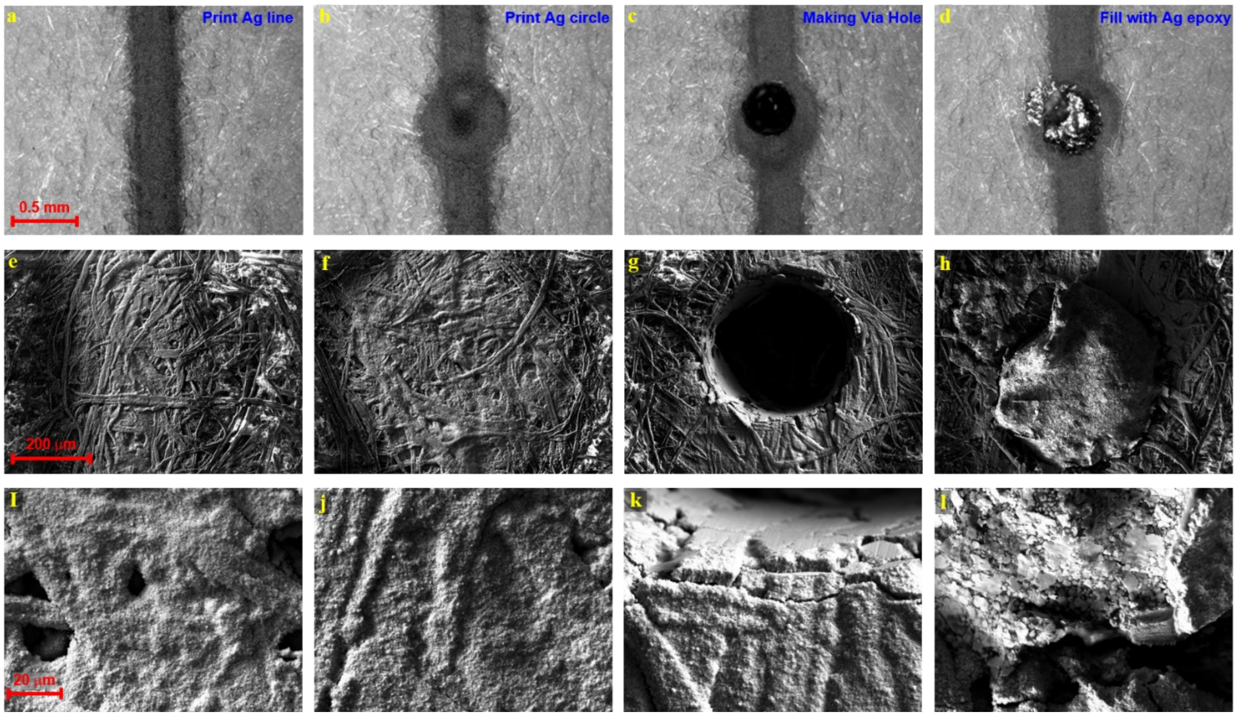

Figure 3. Multiscale imaging of the ANM process for electronic circuit fabrication on paper substrates. The top row (**a-d**) displays the optical images at various stages of the process: (**a**) a line of silver (Ag) is printed; (**b**) a silver circle is formed; (**c**) a via hole is created; (**d**) the hole is filled with silver epoxy. The second row (**e-h**) presents scanning electron microscopy (SEM) images highlighting the surface morphology during the steps in (**a-d**). The bottom row (**i-k**) shows SEM images at higher magnifications during the steps in (**a-d**). The scale bar of images in the same row is equal.

To demonstrate the full functionality of the printed paper Arduino circuit, a traffic light function was printed and programmed into the design. The ATmega328P microcontroller was first bootloaded externally using an Arduino nano board as an In-System Programmer (ISP). This allowed the microcontroller to communicate with Arduino IDE. The successful functioning of the complete paper Arduino board was validated by connecting it to a PC via a universal serial bus (USB) port using an FT232RL mini-USB to TTL serial converter module (see **Movie S1** in the Supporting Information). This allowed uploading the test code on the microcontroller for a three LED traffic light sequence to demonstrate the paper circuit's capabilities. The functioning was verified by the sequential cycling of red (**Figure 4a**), yellow (**Figure 4b),** and green (**Figure 4c)** LEDs (see **Movie S2** in the Supporting Information).

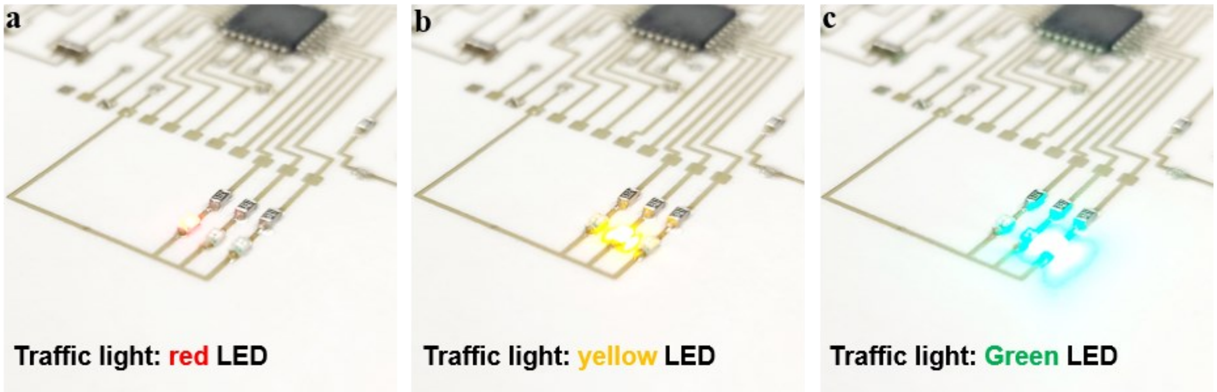

Figure 4. Demonstration of a fully functioning paper Arduino traffic light sequence: The optical images when the (**a**) red, (**b**) yellow, and (**c**) green LEDs are on (see **Movie S2** in the Supporting Information).

To assess the mechanical durability and reliability of the printed paper circuits, a bending cycle test was performed on a simple circuit consisting of an IC, resistors, conductive silver traces (on both sides of the paper), vias, and an LED. The simple circuit was subjected to 20,000 bending cycles with a bending radius of 75 mm. During the test, the LED remained continuously on, powered by the IC, demonstrating the circuit's stable performance without any interruptions or failures. The successful completion of the bending cycle test highlights the reliability of the printed paper circuit, as the conductive silver traces, vias, and components withstood the repeated bending stress without experiencing any mechanical failures, which is crucial for the practical application of transient paper-based electronics. The video demonstrating the bending cycle test and the continuous operation of the LED is provided in the Supplementary Information (**Movie S3**), supporting the mechanical durability and reliability of the printed paper circuits fabricated using the Dry-ANM technique.

Testing Water Solubility and Recyclability of the Components

In the pursuit of developing transient and green electronics, the water solubility and recyclability of the components of paper-based circuits present a critical evaluation. **Figure 5** illustrates the dissolution process and the recovery of electronic components. The fully assembled electronic circuit (**Figure 5a**) was immersed in water, where it began to dissolve (**Figure 5b**). The water-soluble paper fibers disintegrated, leaving behind the intact electronic components, which were then collected from the bath (**Figure 5c**).

The recovered components were dried and cleaned (**Figure 5d**), preparing them for potential reuse in new applications. **Figure 5e** shows the clean and dried electronic components that are ready for recycling after collection. To demonstrate the feasibility of component reusability, a new circuit was printed, and the recovered components were reintegrated into a new circuit (**Figure 5f**). This new device using the recycled components was fully functional, highlighting the possibility of reusing components from transient paper electronics in future applications. This proof-of-concept experiment showcases the potential for reducing electronic waste by designing circuits with recyclability in mind from the initial stages of development, especially for devices with short mission durations.

Our methodology enabled a quick and efficient disassembly process, where the paper substrate dissolved, allowing for the easy separation and recovery of the electronic components (see **Movie S4** in the Supporting Information). Focusing on the recyclability and reusability of the components could potentially transform the way electronics are manufactured and disposed of, leading to a significant reduction in electronic waste. The results emphasize the importance of considering the end-of-life management of electronic devices during the design phase, promoting a more sustainable and environmentally friendly approach to electronics manufacturing.

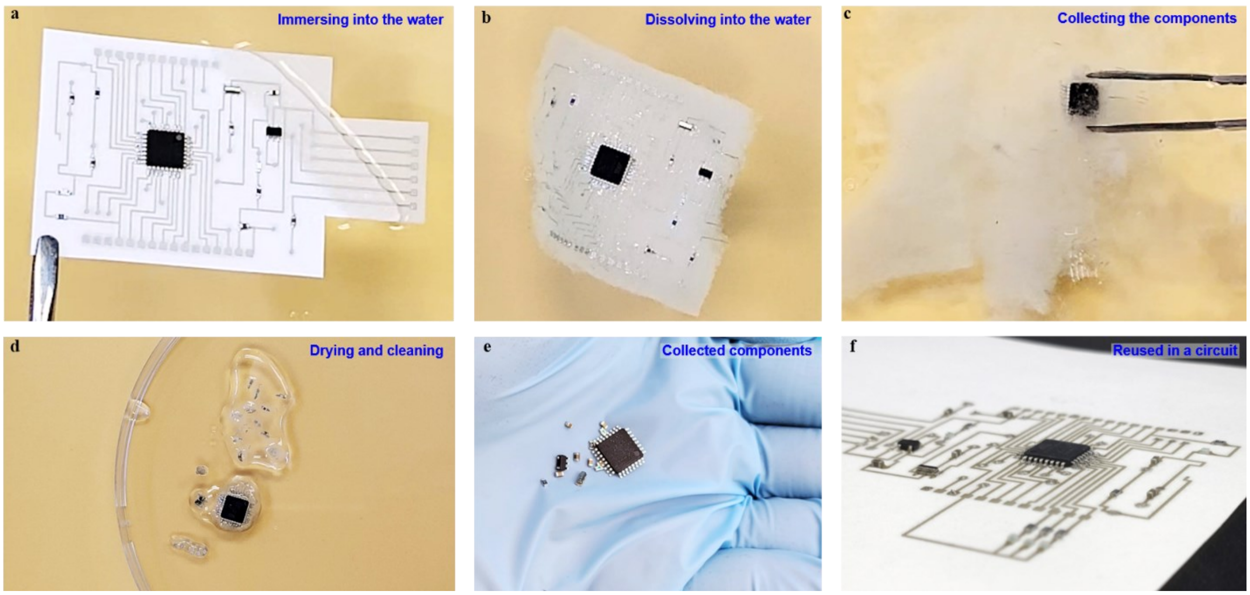

Figure 5. Demonstrating the water solubility and recyclability of the printed paper circuit. (**a**) The fully assembled printed papertronics prior to immersion in water; (**b**) The circuit begins to dissolve when immersed; (**c**) Electronic components are collected from the dissolution bath; (**d**) The components are then dried and cleaned, preparing them for recycling; (**e**) The clean and dried electronic components after collection, ready for reuse; (**f**) Reintegrated components on a new circuit, illustrating the potential for component reusability in new electronic applications. (see **Movie S4** in the Supporting Information).

Testing Recyclability of the Silver Trace

The water-solubility and recyclability of printed silver nanostructures on paper substrates were investigated to explore the potential for recovering valuable materials from electronic waste. **Figure 6** illustrates the process of dissolving the paper substrate and recycling the printed silver grid lines. Initially, a grid pattern of silver lines was printed on water-soluble paper (**Figure 6a**). The sample was then immersed in water (**Figure 6b**), gradually dissolving the paper substrate. As the cellulose fibers dissolved, the printed silver grid remained (**Figure 6c**) and eventually detached from the disintegrating paper (**Figure 6d**).

The floating silver grid structure retained its original shape and was easily collected (**Figure 6e**). **Figure 6f** shows the collected silver grid lines demonstrating the feasibility of recovering the printed silver nanostructures for potential recycling and therefore additional printing. This eco-friendly process showcases the potential for recovering valuable materials such as silver content from electronic waste using a simple, water-based approach.

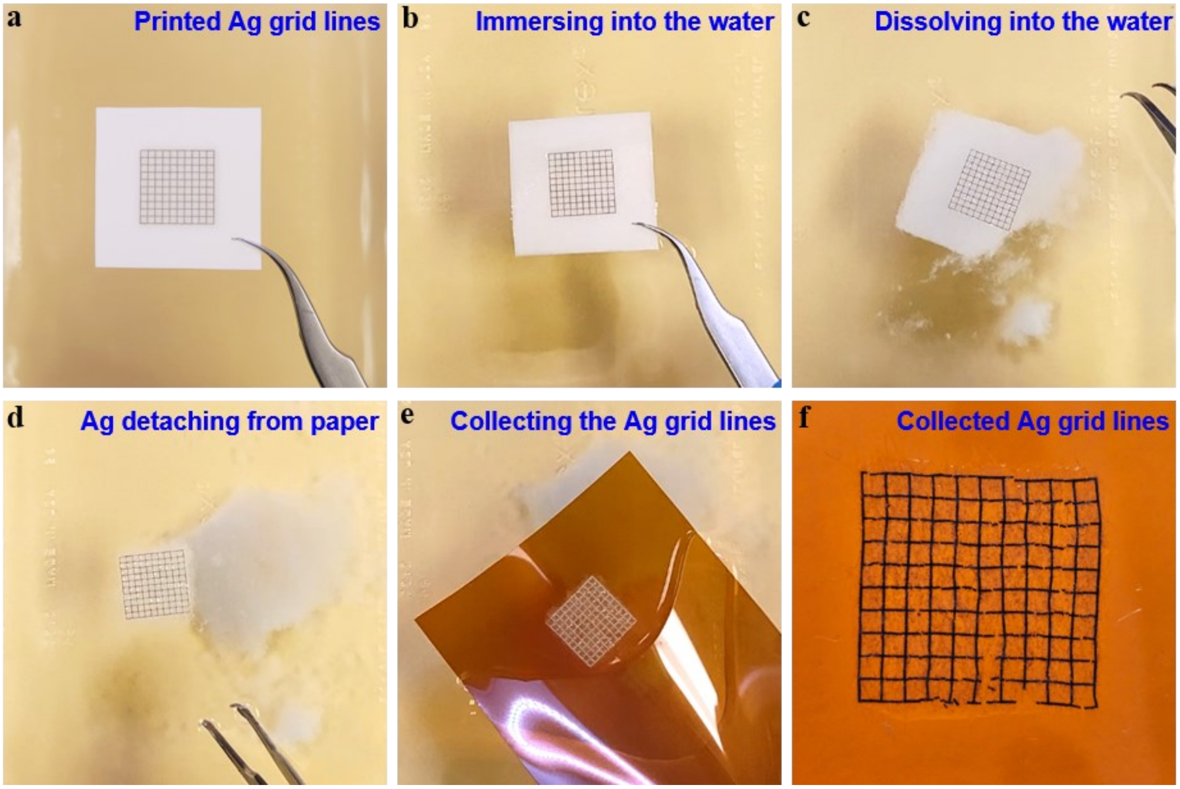

Figure 6. Illustration of the water-solubility and recyclability of printed silver grid lines on paper substrate. (**a**) A printed grid of silver lines before the dissolution process. (**b**) The substrate begins immersion into water. (**c**) Paper substrate visibly dissolving in water (**d**) Detachment of the silver grid from the dissolving paper. (**e**) Collection of the intact silver grid post-dissolution with a polyimide sheet. (**f**) The silver grid lines successfully collected for recycling purposes.

Conclusion

This paper has successfully demonstrated fully functional transient papertronics on biodegradable and water-soluble paper substrates, leading to greener, recyclable, and reduced e-waste electronics manufacturing. The non-contaminating, dry generation and real-time laser sintering of silver nanoparticles enabled precise printing of complex layouts with 3D interconnects on porous water-soluble papers without thermal degradation. A paper Arduino board was fabricated with double-sided printed traces and vias for component attachment. Programming the mounted microcontroller generated a traffic light sequence to validate the printing feasibility of entire electronic systems. Cyclic bending tests showed the mechanical durability and reliability of the printed paper circuits under repeated bending stress, which is crucial for practical applications in flexible and disposable electronics. Dissolving the paper device showed rapid water solubility and allowed the collection and reuse of the components and printed silver materials, proving recyclability. This study highlights the recyclability of printed materials and components, eliminates non-biodegradable polymer-based substrates, and drives technological innovations to reclaim valuable resources from electronic waste in an eco-friendly manner, establishing a sustainable lifecycle for transient electronics. Future work could aim to explore the fabrication of monolithic via structures using the Dry-ANM method by incorporating advanced automation tools, as well as printing diverse functional materials beyond silver, to enable the creation of fully integrated transient paper-based electronics and expand applicability.

Methods and Characterizations

The experiments employed a 99.99% pure, 1-inch diameter solid silver target from Kurt J Lesker, a substrate of regular 32 lb Hammermill Premium laser print copy paper (slow water soluble), MG Chemicals 8331D silver epoxy, and argon as the inert gas environment. Some experiments also utilized 5 pt fast water-soluble paper purchased from SmartSolve.

Fiber lasers (1064 nm wavelength) were employed for both the ablation and sintering processes in the in situ dry nanoparticle generation. The ablation laser parameters were set to 22 W power, 30 kHz frequency, while the chamber pressure was maintained at 5 PSIG.

To optimize printing and sintering, the following parameters were employed: a sintering laser power density of 0.7 MW/m^2, a nozzle diameter of 260 µm, a nozzle-substrate gap of 1.5 mm, an X–Y stage (printing) speed of 4 mm/s, and a total of 20 printing passes.

The SEM images were taken using a ZEISS SEM EVO-10 instrument.

To determine the resistivity, a four-point probe technique was employed, utilizing a Keithley 2400 SourceMeter to measure the resistance of the samples.

The thickness and surface topography of the printed traces were characterized using a KLA Tencor MicroXAM-800 optical profilometer. Scans were performed over an area of 600 µm × 750 µm, with a vertical (z-axis) resolution of 1 µm.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

A.T. designed and performed the experimental setup, printing experiments, materials characterization, data acquisition and analysis, and manuscript writing. A.P. participated in the printing and characterization steps. M.M.S. led the project and participated in experimental design, data analysis, discussions, and manuscript preparation. All of the authors participated in the manuscript preparation and revision processes.

Supporting Information

Supporting information is available online.

- Communication of paper Arduino to a PC (MP4)
- Fully functioning paper Arduino (MP4)
- Bending cycle test demonstrating the mechanical durability and reliability of the printed paper circuit (MP4)
- Water solubility of the circuit and recyclability of the components (MP4)

Data availability statement

Any data that support the findings of this study are included within the article.

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