

Evaluation of commercially-available conductive filaments for 3D printing flexible circuits on paper

Aditya R. Jangid¹, E. Brandon Strong¹, Jacqueline Chuang²,
Andres W. Martinez³ and Nathaniel W. Martinez¹

¹ Department of Biological Sciences, California Polytechnic State University-San Luis Obispo, San Luis Obispo, California, United States

² Department of Biomedical Engineering, California Polytechnic State University-San Luis Obispo, San Luis Obispo, California, United States

³ Department of Chemistry and Biochemistry, California Polytechnic State University-San Luis Obispo, San Luis Obispo, California, United States

ABSTRACT

Three commercially-available conductive filaments were evaluated for 3D printing flexible circuits on paper. While all three filaments were printed successfully, the resulting conductive traces were found to have significantly different impedances when characterized by electrochemical impedance spectroscopy. Using a graphite-doped polylactic acid filament, the flexibility of paper-based conductive traces was evaluated, methods of integrating common electrical and electronic components with the conductive traces were demonstrated, and the resistive heating of the traces was characterized. The ability to 3D print conductive traces on paper using commercially available materials opens many opportunities for rapid prototyping of flexible electronics and for integrating electronic circuits with paper-based microfluidic devices.

Subjects Materials Science (other), Porous Materials, Additive Manufacturing

Keywords Additive manufacturing, Paper, Flexible circuits, Conductive traces

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Corresponding author

Nathaniel W. Martinez,
nmarti32@calpoly.edu

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Additional Information and
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INTRODUCTION

Flexible electronics have many advantages over their ridged counterparts for applications that require moving parts, light weight, irregular or curved shapes, or constrained spaces (Palm *et al.*, 2001; Tsutsui & Fujita, 2002; Nathan *et al.*, 2012). Flexible circuits are typically fabricated by printing conductive inks onto plastic films (Harris, Elias & Chung, 2016; Zardetto *et al.*, 2011), but cellulose-based paper has also been shown to be a suitable substrate with many inherent advantages for the production of flexible electronics (Tobjörk & Österbacka, 2011; Eder *et al.*, 2004; Siegel *et al.*, 2010). Paper is inexpensive (~\$10/m²), widely accessible, thin (<200 µm), lightweight (~80 g/m²) and made from renewable materials (Tobjörk & Österbacka, 2011; Harper, 2000; Russo *et al.*, 2011). Several techniques for fabricating circuits on paper have been developed including metal evaporation or sputtering (Siegel *et al.*, 2010; Kim & Steckl, 2010; Carvalhal *et al.*, 2010), photolithography (Eder *et al.*, 2004), screen printing (Kit-Anan *et al.*, 2012; Tan, Ge & Wang, 2010; Nie *et al.*, 2010; Dungchai, Chailapakul & Henry, 2009), inkjet printing

(Tobjörk & Österbacka, 2011; Mannerbro et al., 2008; da Costa et al., 2015; Bihar et al., 2017; Yang et al., 2007; Yoon et al., 2011; Kawahara et al., 2013), drawing (Russo et al., 2011; Santhiago & Kubota, 2013; Santhiago, Henry & Kubota, 2014; Dossi et al., 2014; Gao, Li & Liu, 2012), and 3D printing (Zhang et al., 2016). Among these techniques, 3D printing of conductive filaments stands out for its speed, ease of use, precision, and relatively low set up and operation costs. To date, most publications describing 3D printed flexible circuits have focused on the preparation of novel conductive filaments. In this work we evaluate commercially-available conductive filaments for 3D printing flexible circuits on paper and demonstrate some unique advantages and applications of this approach.

Additive manufacturing, otherwise known as 3D printing, has revolutionized prototyping in several fields such as construction (Holt et al., 2019), energy (Gebler, Schoot Uiterkamp & Visser, 2014), biotechnology (Gross et al., 2014; Schubert, Langeveld & Donoso, 2014), and healthcare (Yang et al., 2018; Klein, Lu & Wang, 2013; Ventola, 2014). While there are many types of 3D printing (e.g., material extrusion, vat photopolymerization, powder bed fusion, material jetting), this work focused on extrusion-based 3D printing (also known as fused deposition modeling or FDM) due to its ease of use, low cost (a basic 3D printer can be purchased for ~\$200), commercial-availability, and ability to print directly on paper. These characteristics make this technique broadly accessible to users who may not have formal training in conventional fabrication techniques. Two current limitations of extrusion-based 3D printing for the production of flexible circuits are the relatively low resolution and conductance of the printed traces. However, continued research in print settings, hardware, and filament formulation will likely overcome these limitations so that 3D printed flexible circuits could eventually match the quality associated with more advanced manufacturing techniques.

Conductive filaments for 3D printing are commonly made from thermoplastics, such as polylactic acid (PLA) or acrylonitrile butadiene styrene (ABS), doped with a conductive material, such as graphite or copper (Table S1). The three commercially-available conductive filaments selected for this study represent these typical types of filament and were Protopasta, a graphite-doped PLA filament (~\$0.10/g); BuMat, a graphite-doped ABS filament (~\$0.05/g); and Electrifi, a copper infused polyester filament (~\$1.8/g) (Protopasta, <https://www.proto-pasta.com/>; BuMat, <https://www.bumatusa.com/>; MULTI3D, <https://www.multi3dlc.com/>). Although conductive filaments have been demonstrated previously for the fabrication of electronic devices (MacDonald et al., 2014; Duarte et al., 2017; Leigh et al., 2012; Rymansaib et al., 2016; Flowers et al., 2017; Palenzuela et al., 2018), these filaments have not been evaluated for printing flexible circuits on paper. The results of this investigation provide a foundation for users who may be interested in prototyping flexible electronics or integrating electronics with paper-based microfluidic devices using accessible and relatively inexpensive instrumentation and materials.

MATERIALS AND METHODS

Three commercially-available conductive filaments were evaluated in this study: 2.85-mm graphite-doped Protopasta PLA filament (0.5-kg spool; Protoplant, Vancouver, WA,

USA), BuMat Elite Dreamer 1.75-mm conductive ABS filament (1.5-lb spool; BuMat, Lonay, Switzerland), and 2.85-mm Multi-3D Electrifi copper-infused conductive filament (0.1-kg spool; Multi3D, Middlesex, NC, USA). The Protopasta and Electrifi filaments were 3D printed using a BCN3D-Sigma R17 dual extrusion printer (2.85-mm filament compatible; BCN3D Technologies, Barcelona, Spain), and the BuMat filament was 3D printed using a Creality Ender-3 printer (1.75-mm filament compatible; Shenzhen Creality 3D Technology Co., Shenzhen, China). A non-conductive PLA filament (Back-to-basics PLA; Protoplant, Vancouver, WA, USA), was also used. Three types of paper were tested as substrates: Whatman Grade 3 Chr cellulose chromatography paper (Cytiva, Marlborough, MA, USA), Whatman Grade 1 Chr cellulose chromatography paper (Cytiva, Marlborough, MA, USA), and 20 lb. copy paper (Hammermill, Erie, PA, USA). The printed traces were evaluated using an electrochemical workstation (CH Instruments, Bee Cave, TX, USA), a digital multimeter (Dr.meter), a single-board microcontroller (Arduino Uno R3) equipped with an 8-channel 5V solid state relay (SainSmart), and a thermal camera (FLIR C2).

Circuit designs were created in SolidWorks, imported to BCN3D Cura as STL files for printer parameter optimization, and translated to GCODE for interpretation by the printer. A sheet of paper was then secured onto the build plate of the printer using masking tape. The temperature of the build plate was set to 60 °C. For the BCN3D-Sigma printer, an automated build plate calibration was carried out before each print to prevent bed misalignments and x-y axis changes. The build plate's z-magnitude did not require recalibration to account for the additional height of the paper (~200 µm). The Creality printer was calibrated manually. After calibration, the designs were printed on the paper substrates. To print conductive traces on both sides of a piece of paper, a trace was printed on one side of the paper as described, then the paper was flipped over, reattached to the build plate, and a second trace was printed on the other side of the paper.

The electrical properties of traces printed with the three conductive filaments on Whatman Grade 3 Chr paper were characterized *via* electrochemical impedance spectroscopy (EIS) using an electrochemical workstation. The standard traces used for this study were 30-mm long, 3-mm wide, and 0.6-mm high. Lengths of the traces were varied from 10 mm to 100 mm in 10-mm increments, widths were varied from 1 mm to 5 mm in 1-mm increments, and heights were varied from 0.2 mm to 1 mm in 0.2-mm increments. The resolution of the printer was set to 0.2 mm for printing all the traces. EIS was performed on each trace over the frequency range of 100 Hz–10 kHz. When each dimension was varied and tested, the other two dimensions were held constant. To assess the reproducibility of the traces, the resistivities of three generations of traces (30 mm × 3 mm × 0.6 mm, 10 traces per generation) printed using Protopasta were determined by measuring the resistance of each trace using a multimeter. Statistical analysis of the results was performed in JMP (v12.1). A one-way ANOVA was used to determine if there were any significant differences in trace resistivity across the three generations of traces.

To evaluate the flexibility of the conductive traces printed on paper, traces were bent or rolled manually. In one experiment, traces were bent manually to a 90° angle, and the

resistance of the traces was measured before and after bending using multimeter connected to the trace using alligator clips. In a second experiment, traces were subjected to manual bending cycles until they fractured.

Hydrophobic patterns were incorporated into the paper substrates by wax printing (Fig. S1). Patterns were designed in Adobe Illustrator (CS5 v15.1.0) and printed on one side of Whatman Grade 1 Chr paper using a solid ink printer (Phaser 8650; Xerox, Rochester, NY, USA). Traces were then 3D printed on the opposite side of the paper so that the wax would not prevent adhesion of the filament. After 3D printing, the circuits were placed circuit side up in a digital convection oven set to 195 °C for 1 min to melt the wax into the paper.

To characterize resistive heating, traces printed using Protopasta on Whatman Grade 3 Chr paper were connected to a solid state relay with a load voltage of 120 VAC. The amount of power transferred from the relay to the trace was controlled by pulse width modulation, which was manipulated by changing the relay's duty cycle using a single-board microcontroller (Fig. S2). Duty cycles in the range of 0.5% to 21% were tested. The temperature of the traces was monitored continuously using a thermal camera and was recorded once it equilibrated. The traces were suspended in air throughout the experiments.

RESULTS AND DISCUSSION

All three conductive filaments were successfully 3D printed onto paper substrates to produce flexible circuits (Fig. 1). The first time a filament was printed on a paper substrate, extensive optimization of print settings was required to obtain reproducible prints. While the settings will vary depending on the printer, materials, downstream use, and print complexity, we found that an infill density in the range of 75–90%, a retraction distance in the range of 0.5–1.0 mm, a print speed in the range of 50–70 mm/s, and a print acceleration of 4,000 mm/s² worked best for our applications. There are several open-source platforms that can assist with print quality optimization such as Simplify3D and MatterHackers (SIMPLIFY3D, <https://www.simplify3d.com/>; MatterHackers, <https://www.matterhackers.com/>). The BCN3D printer, which is more sophisticated and expensive (~\$3,000) than the Creality printer, was more convenient to work with. Nevertheless, the Creality printer produced adequate traces, demonstrating that even an inexpensive 3D printer (~\$200) can be used for making flexible circuits on paper.

The strongest adhesion between filament and paper, for all three filaments, was observed for Whatman Grade 3 Chr paper, so this paper was used for the characterization experiments. We hypothesize that the higher surface roughness of Whatman Grade 3 Chr paper compared to the other two types of paper was the main reason for the stronger adhesion of the filament. Traces printed on Whatman Grade 1 Chr paper and copy paper detached frequently when the paper was bent or folded. Copy paper was also torn frequently by the print head as it moved across the surface of the paper during printing.

Comparing the three filaments, we found that BuMat's print quality was adequate (*i.e.*, defined edges, consistent infill pattern, and slight variability in topography along the top of the trace) but the filament was not as conductive as Protopasta or Electrifi (Fig. 1B).

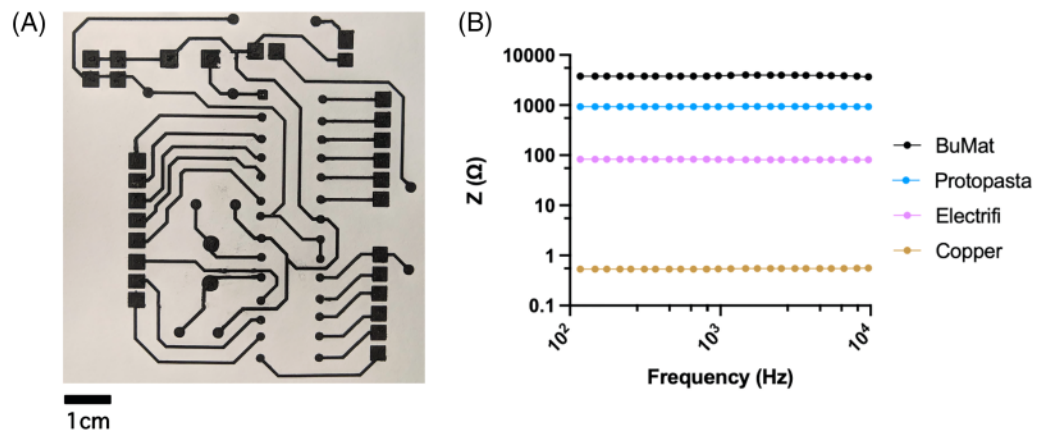


Figure 1 Conductive filaments printed on paper. (A) Example of a flexible circuit 3D printed on paper using Protopasta filament. (B) Impedance vs frequency plots for traces printed with the three filaments. All traces were 30-mm long, 3-mm wide and 0.6-mm high. A solid copper wire (22 AWG, 30-mm long) was also tested for comparison. Full-size [DOI: 10.7717/peerj-matsci.21/fig-1](https://doi.org/10.7717/peerj-matsci.21/fig-1)

The Electrifi filament was the most conductive, but it was challenging to work with because of the extensive print optimization that was required and the raw filament brittleness, which led to many mechanical difficulties (e.g., print-head clogging, motor jam, snapped filament in Bowden tube, etc.). The challenges with the raw filament may be due to the filament's preference for a direct-drive extrusion printer rather than the Bowden tube feeding delivery system we used with the BCN3D printer (Flowers et al., 2017; Roy et al., 2017). Protopasta was the most convenient filament to work with since the traces were reproducible, well-defined, and moderately conductive. It should be noted that commercially-available filaments can vary from batch to batch. Therefore, print settings may need to be optimized for each new roll of filament, and the electrical properties of conductive traces may change when a new roll of filament is used.

When characterizing the electrical properties of traces printed on paper, we observed the trends that would be expected (Fig. 2 and Fig. S3). Impedance increased with increasing trace length and decreased with increasing trace height and width (Figs. 2A–2C). By changing the width and height of the traces, it should be possible to control the impedance of traces with different lengths. There was no significant difference in resistivity across three different generations of traces printed with Protopasta ($F = 0.256$, $df = 2.27$, $p = 0.776$), which suggests that it should be possible to print reproducible circuits using this filament (Fig. 2D).

3D printed circuits on paper are flexible, but the flexibility of the circuits decreases as the height of the printed trace increases (Fig. 3A). We observed minimal changes in resistance when traces were bent or rolled (Figs. 3B–3E). When traces were bent to a 90° angle, the resistance increased momentarily but then equilibrated back to within 0–0.2 kΩ of the original resistance (Fig. S4). We also found that traces printed on paper using Protopasta could be bent multiple times (≥ 20 bending cycles for traces with a height of 0.2 mm) before fracturing, although traces with a height of 1 mm fractured after only two bending cycles. For geometrically constrained systems, the flexible circuits could

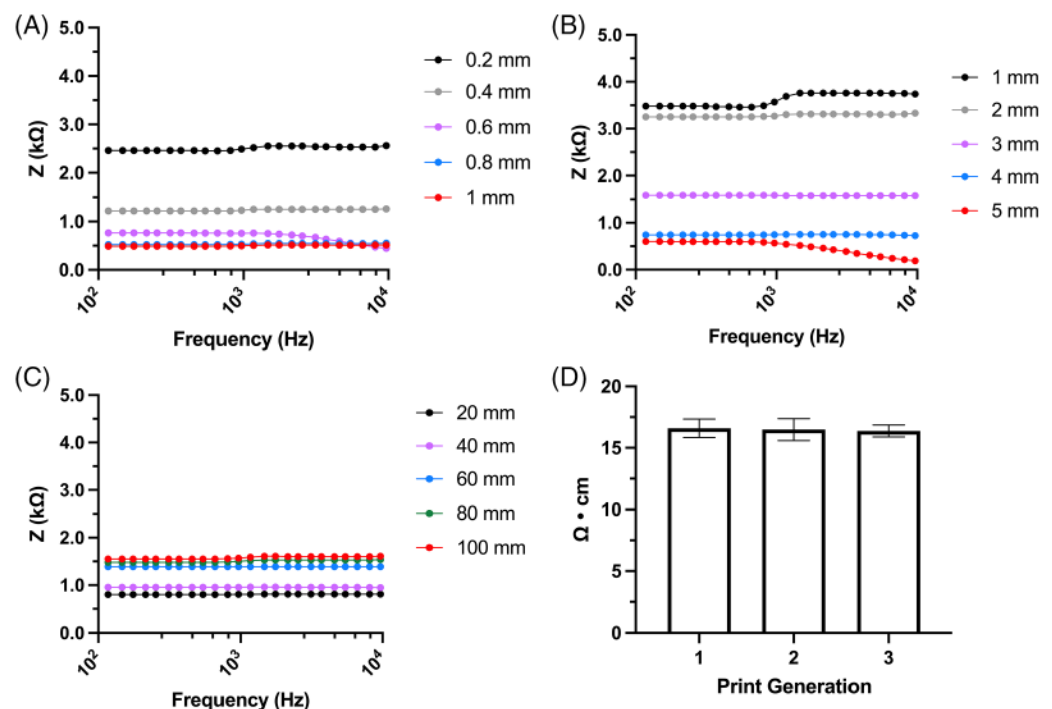


Figure 2 Characterization of Protopasta traces printed on Whatman Grade 3 Chr paper. (A) Effect of trace height on impedance. All traces were 30-mm long and 3-mm wide. (B) Effect of trace width on impedance. All traces were 30-mm long and 0.6-mm high. (C) Effect of trace length on impedance. All traces were 3-mm wide and 0.6-mm high. (D) Reproducibility of printed traces across three generations (10 traces/generation). All traces were 30-mm long, 3-mm wide and 0.6-mm high. No significant differences in resistivity were observed ($F = 0.256$, $df = 2.27$, $p = 0.776$).

Full-size [DOI: 10.7717/peerj-matsci.21/fig-2](https://doi.org/10.7717/peerj-matsci.21/fig-2)

be rolled up or folded. In addition, printing on both sides of the paper substrate allows for the production of multi-sided flexible electronics (Figs. 3D and 3E). The paper acts as an insulator, so conductive traces printed on opposite sides of a piece of paper are electrically isolated from each other.

A unique capability of 3D printing is the ability to fabricate conductive structures in various shapes and integrate them with other conductive or non-conductive structures (Fig. 4 and Fig. S5). A simple demonstration of this capability is the production of overlapping circuits. Using the BCN3D-Sigma printer's dual extrusion system, we printed conductive traces going under and over slabs of non-conductive PLA, which served as an insulating layer and electrically isolated the conductive traces (Figs. 4A and 4B). These layered structures may be useful in the production of complex circuit networks. Additionally, it is possible to print features that facilitate interfacing the paper-based circuits with other electrical components. For example, nodules printed at the beginning of conductive traces can serve as simple connection points for alligator clips (Fig. 4C). A sufficiently thick trace of conductive PLA (typically ≥ 1 mm) can also be used as a hot melt adhesive to glue common electrical components such as LEDs, resistors or metal wires directly to the printed circuit (Figs. 4D and 4E). Heating the printed circuit on a hot-plate to 60–65 °C softens the conductive PLA so that electrical components can be inserted

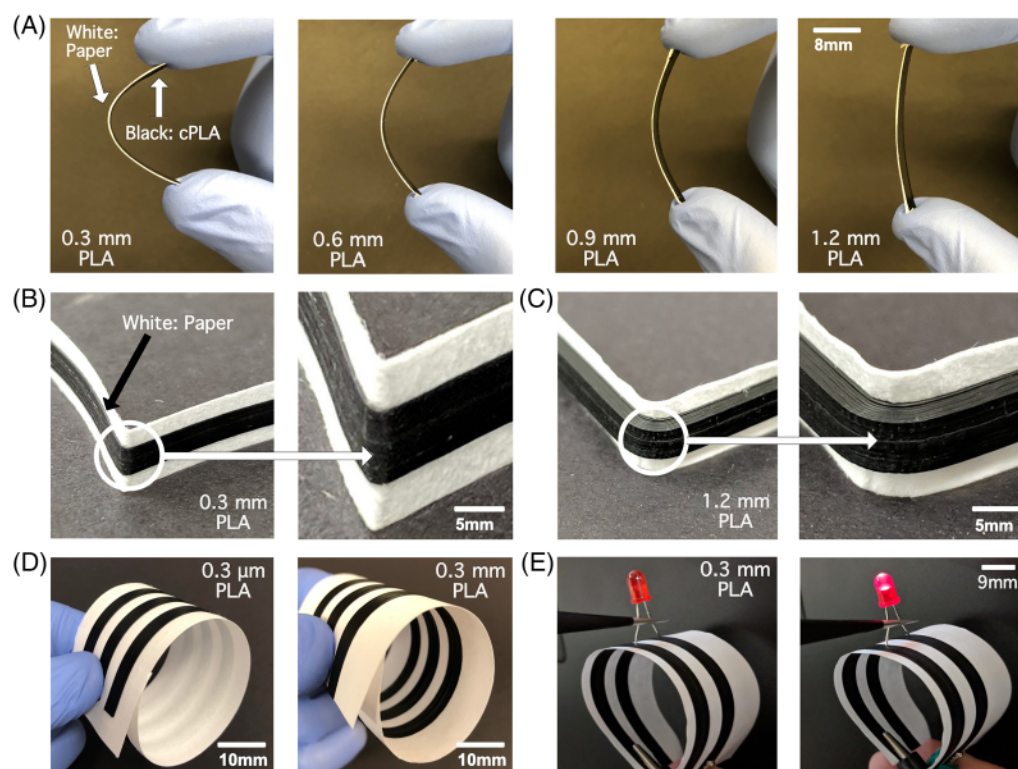


Figure 3 Flexibility of 3D printed traces on paper. (A) Protospasta (cPLA) printed with various heights and flexed. (B) and (C) Printed electrodes could be bent at both the smallest (B) and largest (C) heights tested without any significant change in resistance. (D) Rolled one-sided and two-sided circuits. (E) Rolled two-sided circuit powering an LED.

Full-size [DOI: 10.7717/peerj-matsci.21/fig-3](https://doi.org/10.7717/peerj-matsci.21/fig-3)

without cracking or damaging it. Upon cooling to room temperature, the electrical components become fixed in place. Since this process is reversible, the electrical components can be easily recovered by reheating the circuit. Finally, it is also possible to print more than one type of conductive filament to harness the capabilities of the different filaments in one circuit. We demonstrated a combination of Electrifi and Protospasta, where the Protospasta elements could function as resistors (Fig. S6).

An inherent limitation of paper as a substrate for flexible circuits is its hydrophilicity. If an aqueous solution came into contact with a paper-based circuit, capillary forces would spread the solution across the entire circuit, which could damage the structural integrity of paper, weaken the adhesion between the conductive trace and the paper, or short the circuit. An obvious solution to this problem would be to make the paper hydrophobic prior to 3D printing the circuit, however, we found that the conductive filaments adhered poorly to paper that had been made hydrophobic using wax. We overcame this problem by applying a technique known as wax printing that is used to fabricate paper-based microfluidic devices (Carrilho, Martinez & Whitesides, 2009). In wax printing, wax is printed onto one side of a sheet of paper, and then the paper is heated to reflow the wax so that it penetrates through the thickness of the paper creating patterns of hydrophobic barriers and hydrophilic channels. We found that it was possible

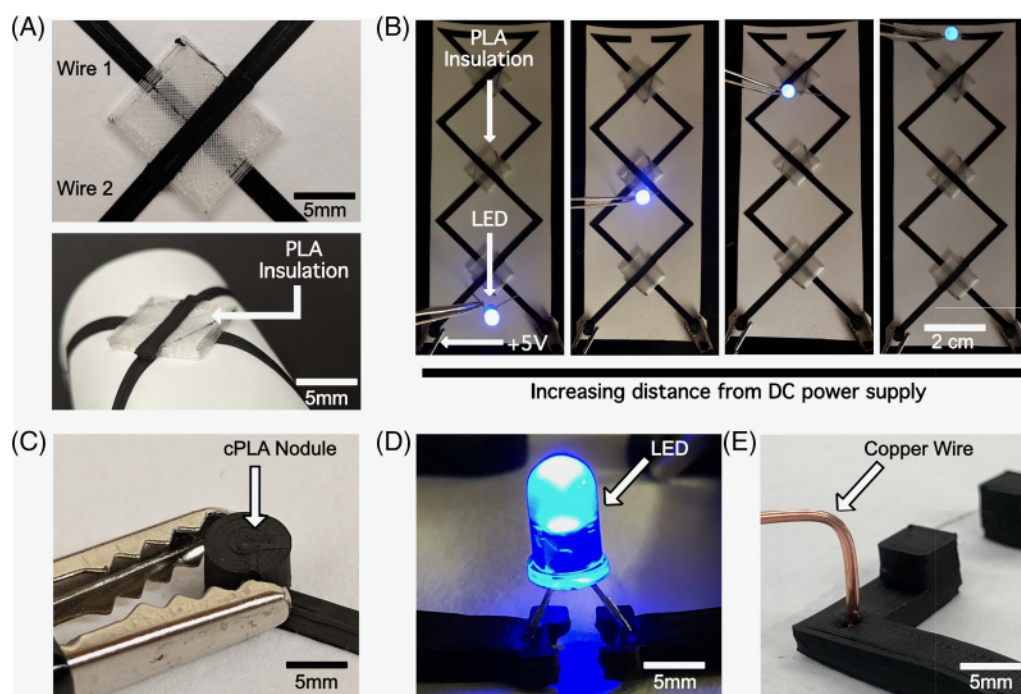


Figure 4 Applications of 3D printed circuits. (A) Crossing conductive traces with a non-conductive PLA insulation layer between them. (B) Criss-crossing conductive traces powering an LED at various points in the circuit. LED brightness visually decreases with increasing distance from the +5V power supply due to the resistance of the traces. (C) Conductive nodule for interfacing with external electronic components such as alligator clips. (D) Direct integration of an LED into a Protopasta circuit. (E) Direct integration of copper wire into a Protopasta circuit. Full-size DOI: [10.7717/peerj-matsci.21/fig-4](https://doi.org/10.7717/peerj-matsci.21/fig-4)

to create hydrophobic paper-based circuits by printing wax on one face of the paper, then 3D printing the circuit on the other face of the paper, and finally heating the paper in an oven to reflow the wax (Fig. S1). This process maintained good adhesion between the filament and the paper, and, in principle, could also be used to integrate 3D printed circuits with paper-based microfluidic devices.

Another potential concern with using 3D printed circuits on paper is that, for applications requiring high current flow, the circuits could generate enough heat to damage the circuit. As would be expected, running current through Protopasta traces does lead to resistive heating, but we found that the paper substrate provides structural stability to the circuit allowing the traces to reach higher temperatures before deforming compared to freestanding traces (Fig. S7). Traces printed on paper could withstand temperatures up to 78 °C without warping or detaching from the paper, whereas freestanding traces started to warp at temperatures around 60 °C. We also found that the temperature of a given trace could be controlled by changing either the duty cycle or the dimensions of the trace (Fig. 5). These results suggest 3D printed traces could be used as resistive heaters for applications requiring controlled temperatures such as isothermal DNA amplification (Chang *et al.*, 2012). Since paper is a relatively good insulator, paper-based 3D printed traces allow for localized and targeted heating (Uetani & Hatori, 2017).

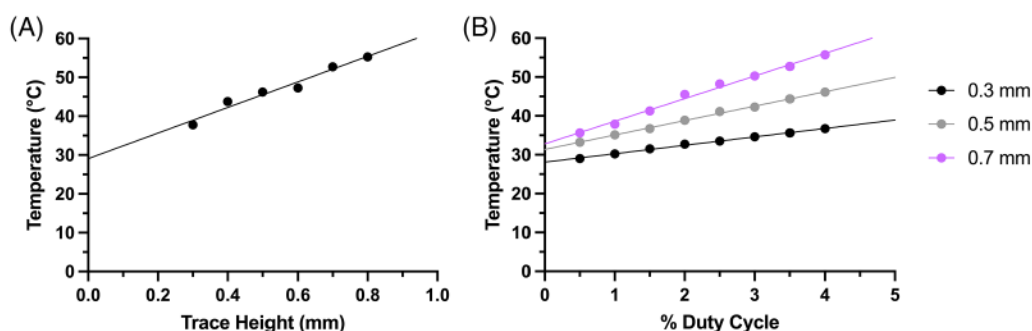


Figure 5 Characterization of Protopasta traces as resistive heaters. (A) Plot of temperature of the trace as a function of its height. All traces were 40-mm long and 5-mm wide, and the microcontroller was programmed to a 5% duty cycle. The data were fit with a linear trendline (Temperature = 32.94 (Height) + 29.06, $R^2 = 0.965$). (B) Plot of temperature of traces with various heights as a function of % duty cycle. All traces were 30-mm long and 3-mm wide. The data were fit with linear trendlines (0.3 mm: Temperature = 2.17 (% Duty Cycle) + 28.10, $R^2 = 0.99$. 0.5 mm: Temperature = 3.71 (% Duty Cycle) + 31.38, $R^2 = 0.99$. 0.7 mm: Temperature = 5.82 (% Duty Cycle) + 32.81, $R^2 = 0.99$). Actual temperatures may vary with different batches of filament, so additional calibration may be required for applications requiring specific temperatures.

Full-size [DOI: 10.7717/peerj-matsci.21/fig-5](https://doi.org/10.7717/peerj-matsci.21/fig-5)

CONCLUSIONS

We characterized three commercially-available conductive filaments for 3D printing flexible circuits on paper. The conductive PLA filament Protopasta gave the most reproducible traces and was the easiest to work with. Using this filament, we were able to fabricate a variety of flexible circuits and demonstrate simple ways of integrating common electrical components into these circuits. Conductive filaments can also be used to 3D print resistive heaters on paper that can achieve stable temperatures in the range of 30 to 78 °C and could be integrated with paper-based microfluidic devices for applications that require heating. Electronic circuits are an integral part of modern technologies, and the ability to make prototypes of electronic circuits rapidly, cost-effectively, and using commercially-available materials will expand the range of users who can develop and benefit from these technologies.

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ADDITIONAL INFORMATION AND DECLARATIONS

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Competing Interests

Andres W. Martinez is an Academic Editor for PeerJ. The authors declare that they have no other competing interests.

Author Contributions

- Aditya R. Jangid conceived and designed the experiments, performed the experiments, analyzed the data, performed the computation work, prepared figures and/or tables, authored or reviewed drafts of the paper, and approved the final draft.
- E. Brandon Strong conceived and designed the experiments, performed the experiments, analyzed the data, performed the computation work, prepared figures and/or tables, authored or reviewed drafts of the paper, and approved the final draft.
- Jacqueline Chuang conceived and designed the experiments, performed the experiments, authored or reviewed drafts of the paper, and approved the final draft.
- Andres W. Martinez conceived and designed the experiments, analyzed the data, prepared figures and/or tables, authored or reviewed drafts of the paper, and approved the final draft.
- Nathaniel W. Martinez conceived and designed the experiments, analyzed the data, prepared figures and/or tables, authored or reviewed drafts of the paper, and approved the final draft.

Data Availability

The following information was supplied regarding data availability:

The raw data for the characterization of the conductive traces printed on paper is available in the [Supplemental Files](#).

Supplemental Information

Supplemental information for this article can be found online at <http://dx.doi.org/10.7717/peerj-matsci.21#supplemental-information>.

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