

Abstract

Combining metal nanoparticle printing with polymer additive manufacturing has the potential to enable the manufacturing of devices with embedded electronic elements. State-of-the-art manufacturing processes offer a trade off between circuit resistivity or polymer damage. The goal of this work is to significantly decrease interconnect resistivity without damaging the substrate. In order to accomplish this goal we explore the mechanisms of Fused Filament Fabrication (FFF) of Acrylonitrile Butadiene Styrene (ABS), printing of silver NPs and out-of-chamber Intense Pulsed Light (IPL) sintering of the printed circuits. While attempting to achieve appropriate levels of circuit resistance, IPL of only-nanosphere based circuits lead to thermal damage of the polymer. However, a combination of both Ag nanospheres and nanowires achieves a resistivity several times lower than state-of-the-art (13.1 $\mu\Omega\text{-cm}$ or 8 x bulk silver) without any polymer damage. This circuit was sintered within 0.75 s of IPL

Experimental

The polymer substrate base was fabricated using a CraftBot Plus FFF printer (Craftunique), the build plate was then removed from the printer and the Ag NP ink was then deposited onto the substrate via an Aerosol Jet Printing (AJP) head with the use of laser-cut polypropylene masks, the build plate is then moved to the IPL Sinteron 3000 system (Xenon Corporation) where the silver NP's are sintered together. The build plate is then moved back to the Craftbot printer where additional material is deposited on top of the printed circuit (Fig. 1). Lengths of copper tape are attached to the ends of the deposited circuit to measure electrical resistance values.

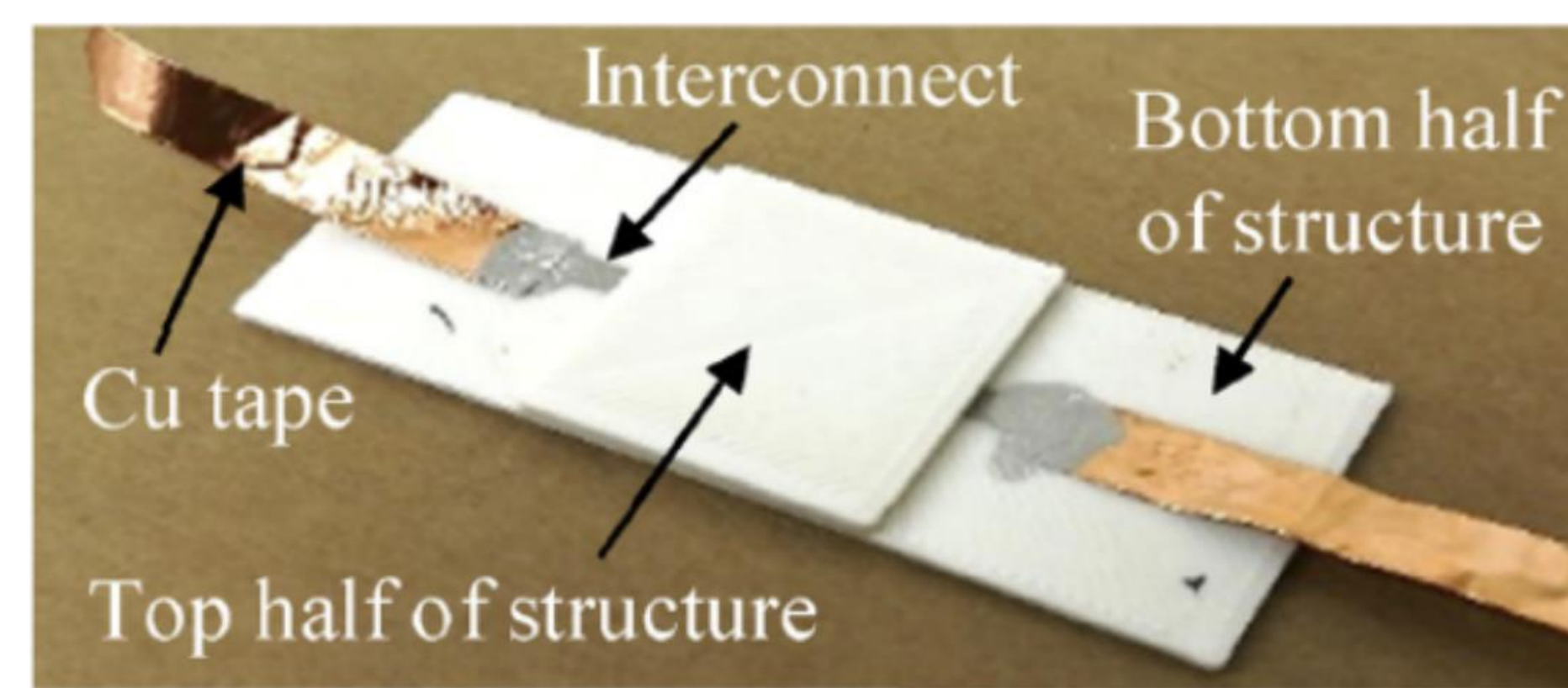


Fig. 2: Completed Sample with Embedded Circuit

IPL parameters for sintering of the deposited inks were varied to achieve the lowest resistance possible, changing both fluence J/cm^2 and number of pulses applied. Increasing the fluence or the number of pulses would cause a reduction in resistance up until a certain point after which IPL damages or evaporates a portion of the circuit causing a spike in resistance. In addition to changing IPL parameters, ink formulae were changed to find the optimal NW:NS combination for the lowest resistance possible.

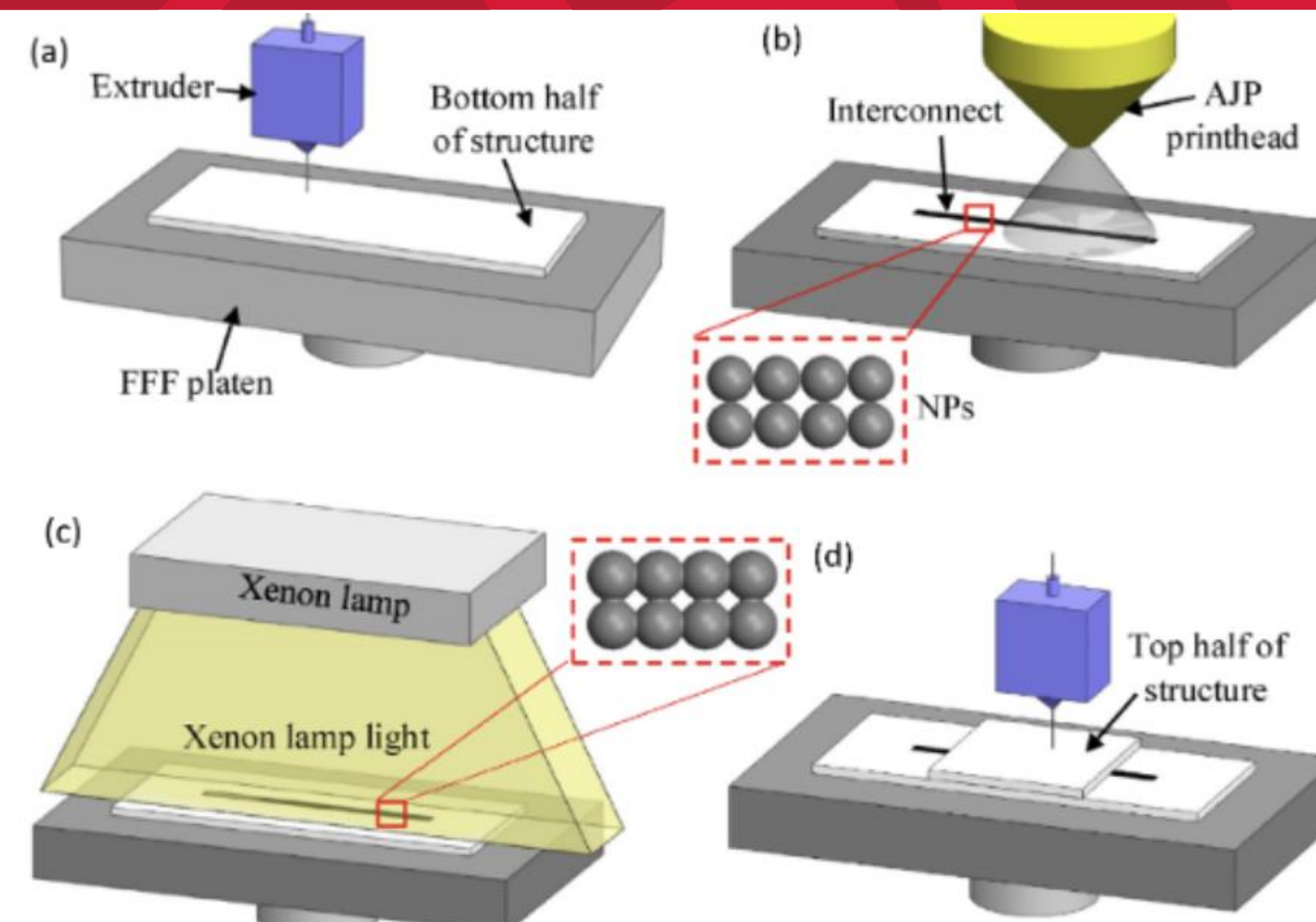


Fig. 1: Schematic of Printing Process: (a) FFF of bottom structure (b) AJP of interconnect (c) IPL (d) Overprinting of top structure

Results and Discussion

The highest as-printed resistances were seen with interconnects that had a NW:NS ratio of 0:100, this resistance was on the order of $\text{M}\Omega$. When attempting to find optimal IPL parameters for the 0:100 NW:NS ratio the ABS substrate is significantly damaged before any appreciable reduction in resistivity is observed.

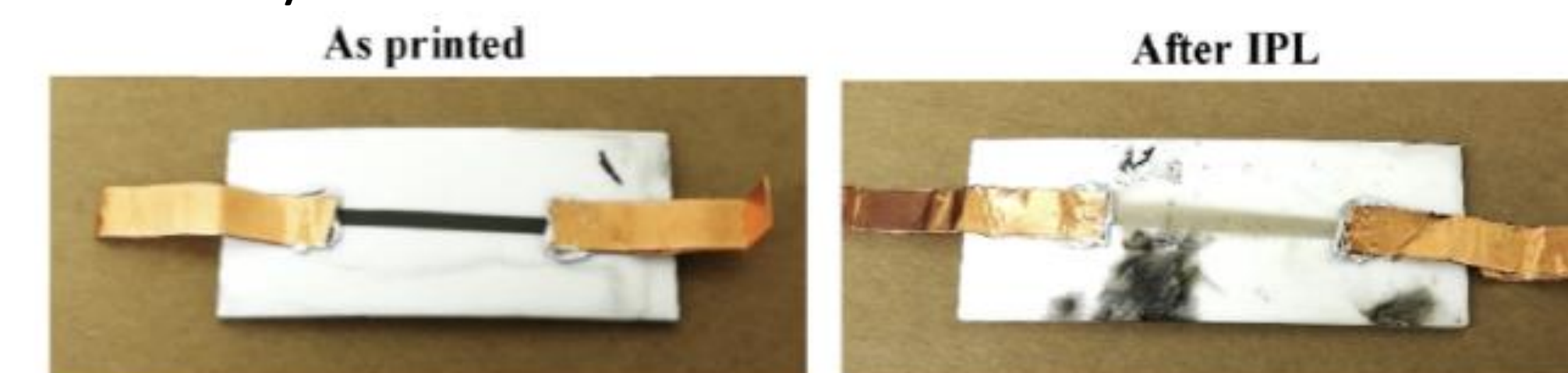


Fig. 3: Damage on ABS substrate with 0:100 NW:NS Ratio Interconnect

Interconnects comprised of both NW and NS had significantly lower as-printed resistances, and had notable decreases in resistance without any damage occurring on ABS substrate. As-printed resistivity decreases with increasing NW content, lowest as-printed resistivity is seen with 100:0 NW:NS (22 $\mu\Omega\text{-cm}$). Additionally, interconnects with greater NW content also see greater reductions in post-IPL resistivity, the lowest value achieved was 13.1 $\mu\Omega\text{-cm}$ for a NW:NS ratio of 100:0. This resistivity is 3.8 times lower than state-of-the-art damage-free oven annealing.

Table 1: Optimal Fluence and Pulses for various NW:NS Ratios

NW: NS	Fluence (J/cm^2)	No. of pulses	Total time (s)
25:75	2.5	3	1.3
50:50	3	3	1.5
100:0	4.5	1	0.75

Dynamic changes in resistivity were measured during the overprinting process (Fig 4). The vertical lines of the graph indicate the completion of an ABS layer deposition (layers 1–6).

Dynamic changes in resistivity were measured during the overprinting process (Fig 4). The vertical lines of the graph indicate the completion of an ABS layer deposition (layers 1–6). Initially there is a minor reduction in resistivity in stage A. After about 25% of the first layer is deposited there is a more significant increase in resistivity which lasts until the completion of layer 1 deposition. In stage C, deposition of layers 2 through 4, the resistivity remains constant or decreases slightly. In stage D, deposition of the 5th layer, there is a sharp decrease in resistivity, even after printing a 6th layer.

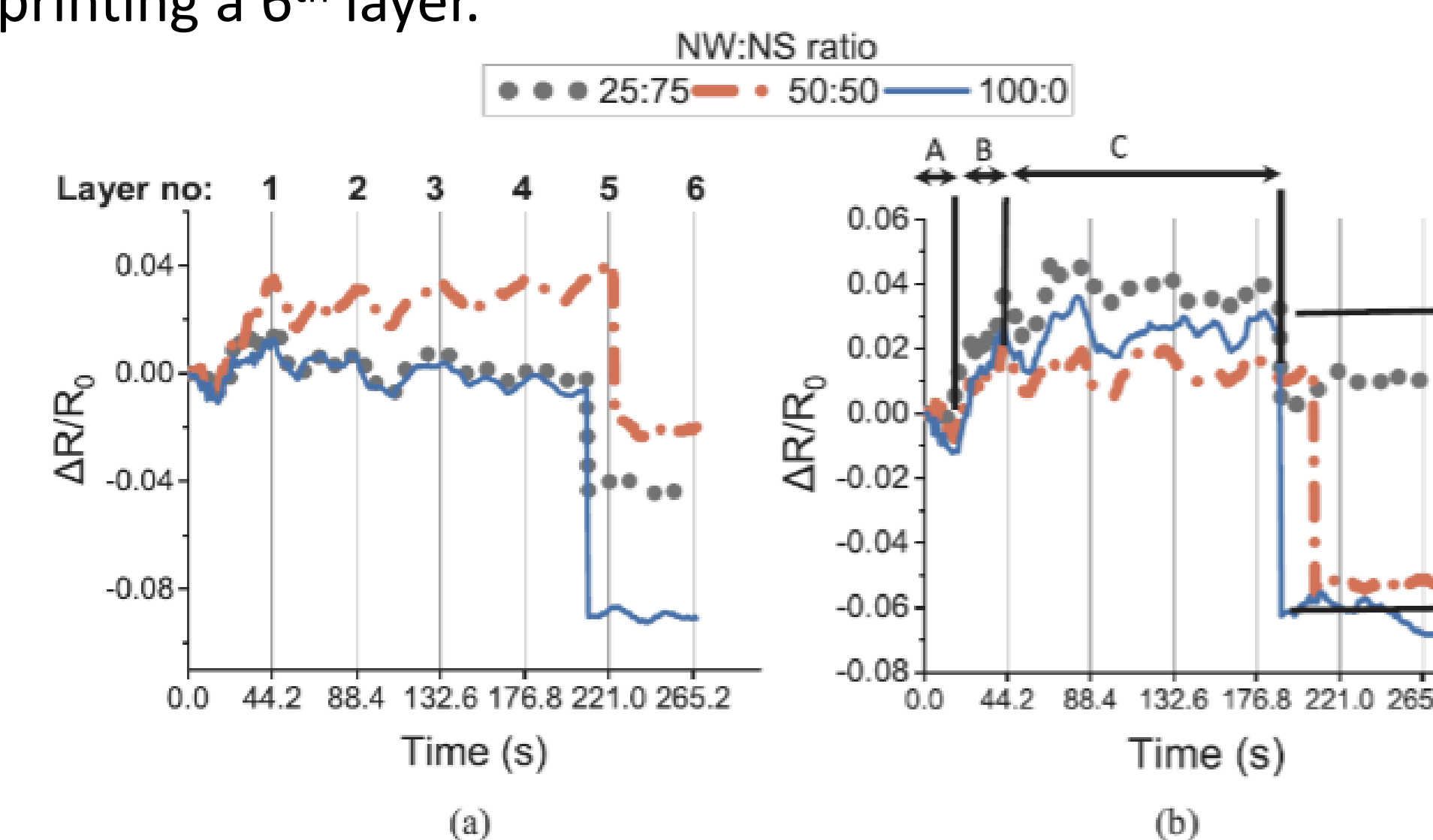


Fig 5. Dynamic resistivity during overprinting (a) unsintered samples (b) post-IPL samples.

The following mechanisms are responsible for the observed resistivity changes during overprinting. First is the temperature-induced increase in resistivity of metals due to heat transfer from deposited polymer. Second is thermomechanical stress, the tensile stress during expansion can break interparticle necks and increase resistivity and compressive stress during shrinkage can increase interparticle neck size to reduce resistivity. Final mechanism is the temperature-induced sintering which can reduce the resistivity during overprinting.

Future Direction

The future goal of this project is to construct a novel, hybrid 3D printer capable of combining all the manufacturing processes into one. This would enable the uninterrupted manufacturing of embedded structures with 3D patterned interconnects. Creating this device would also serve as a testament to the scalability of the manufacturing process as well as a significant increase in throughput capability.

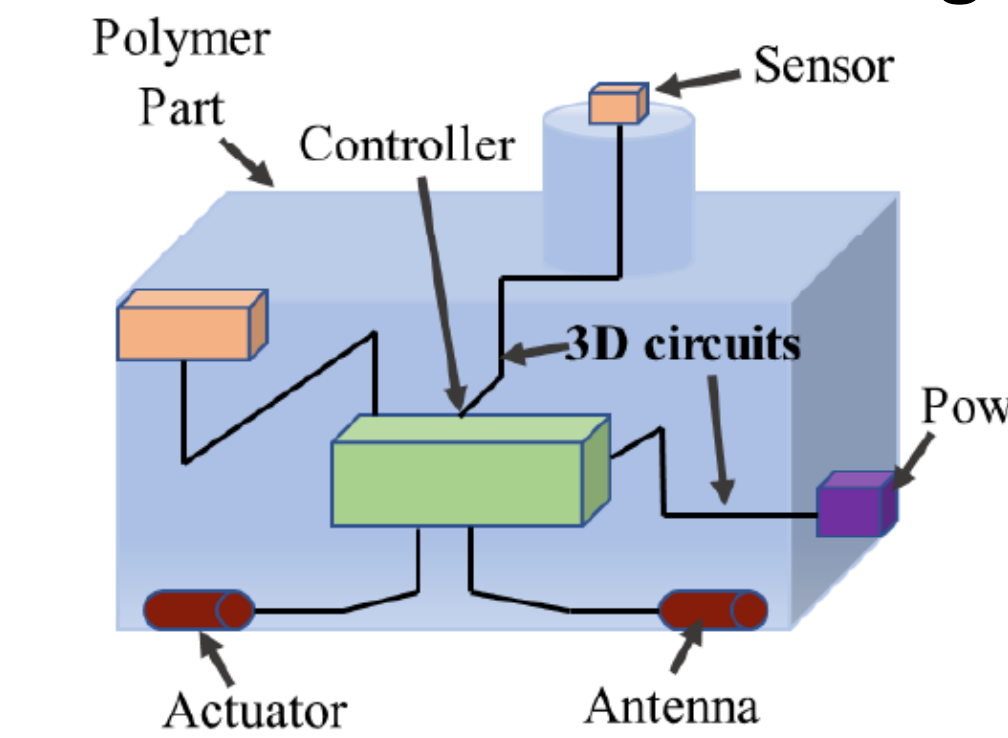


Fig. 6: 3D Embedded Electronic

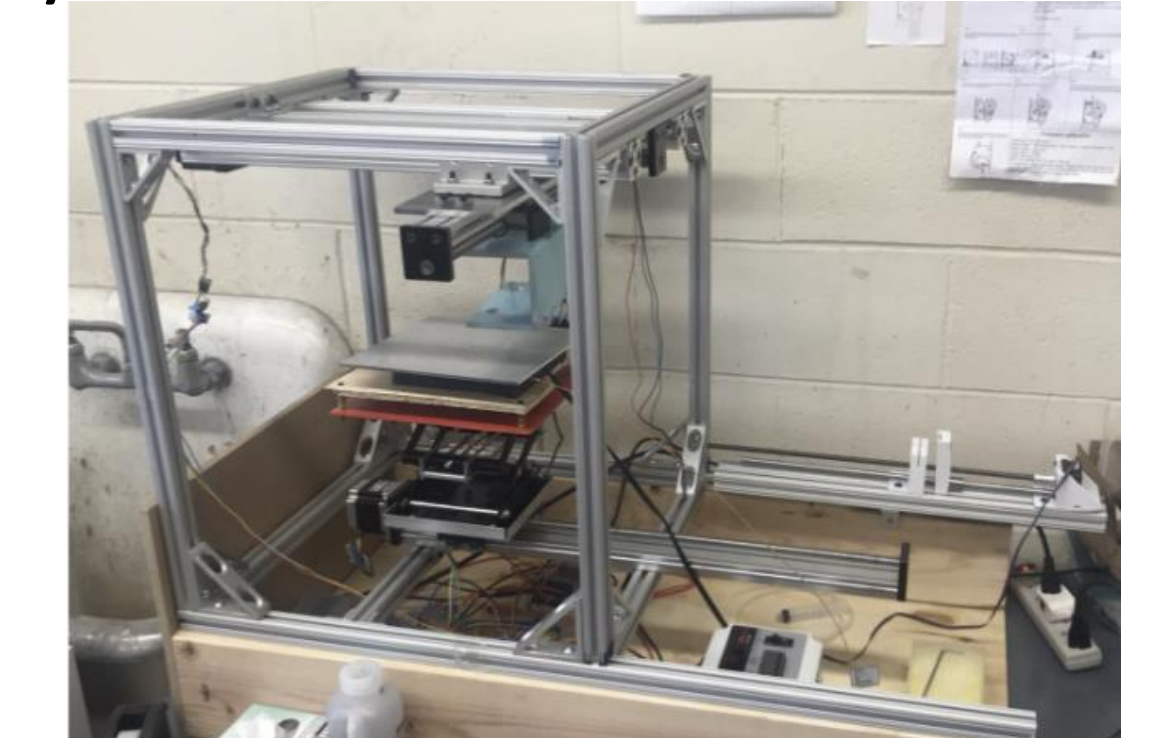


Fig. 7: Hybrid 3D Printer in Construction

Acknowledgements

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References

- [1] E. MacDonald, R. Wicker, Multiprocess 3D printing for increasing component functionality, *Science* 353 (6307) (2016) aaf2093.
- [2] N. Zhou, C. Liu, J.A. Lewis, D. Ham, Gigahertz electromagnetic structures via direct ink writing for radio-frequency oscillator and transmitter applications, *Adv. Mater.* 29 (15) (2017) 1605338.
- [3] Y. Jo, J.Y. Kim, S. Jung, B.Y. Ahn, J.A. Lewis, Y. Choi, S. Jeong, 3D polymer objects with electronic components interconnected via conformally printed electrodes, *Nanoscale* 9 (39) (2017) 14798–14805.
- [4] E. MacDonald, R. Salas, D. Espalín, M. Perez, E. Aguilera, D. Muse, R.B. Wicker, 3D printing for the rapid prototyping of structural electronics, *IEEE Access* 2 (2014) 234–242.
- [5] M. Navarrete, A. Lopes, J. Acuna, P. Estrada, E. MacDonald, J. Palmer, R. Wicker, Integrated Layered Manufacturing of a Novel Wireless Motion Sensor System With GPS, University of Texas El Paso, W.M. Keck Center for 3D Innovation, 2007.
- [6] C. Shemelya, F. Cedillos, E. Aguilera, D. Espalín, D. Muse, R. Wicker, E. MacDonald, Encapsulated copper wire and copper mesh capacitive sensing for 3-D printing applications, *IEEE Sens.* 15 (2) (2015) 1280–1286.
- [7] N. Lazarus, S.S. Bedair, G.L. Smith, Creating 3D printed magnetic devices with ferrofluids and liquid metals, *Addit. Manuf.* 26 (2019) 15–21.
- [8] D.-H. Kim, Y. Kim, J.-W. Kim, Transparent and flexible film for shielding electromagnetic interference, *Mater. Des.* 89 (2016) 703–707.
- [9] S. Yao, J. Cui, Z. Cui, Y. Zhu, Soft electrothermal actuators using silver nanowire heaters, *Nanoscale* 9 (11) (2017) 3797–3805.
- [10] D.J. Roach, X. Kuang, C. Yuan, K. Chen, H.-J. Qi, Novel ink for ambient condition printing of liquid crystal elastomers for 4D printing, *Smart Mater. Struct.* 27 (12) (2018) 125011.
- [11] P.F. Flowers, C. Reyes, S. Ye, M.J. Kim, B.J. Wiley, 3D printing electronic components and circuits with conductive thermoplastic filament, *Addit. Manuf.* 18 (2017) 156–163.
- [12] K.M.M. Billah, J.L. Coronel, M.C. Halbig, R.B. Wicker, D. Espalín, Electrical and thermal characterization of 3D printed thermoplastic parts with embedded wires for high current-carrying applications, *IEEE Access* 7 (2019) 18799–18816.
- [13] M. Liang, C. Shemelya, E. MacDonald, R. Wicker, H. Wu, 3D printed microwave patch Antenna via fused deposition method and ultrasonic wire mesh embedding technique, *IEEE Antenna Wirel. Propag.* 14 (2015) 1346–1349.
- [14] M. Doster, A. Pflau, Z. Gao, G.S. Herman, C.H. Chang, R. Malhotra, Modeling nanoscale temperature gradients and conductivity evolution in pulsed light sintering of silver nanowire networks, *Nanotechnology* 29 (50) (2018) 505205.
- [15] H.-J. Hwang, H. Devaraj, C. Yang, Z. Gao, C.-h. Chang, H. Lee, R. Malhotra, Rapid Pulsed Light Sintering of Silver Nanowires on Woven Polyester for personal thermal management with enhanced performance, durability and cost-effectiveness, *Sci. Rep.* 8 (1) (2018) 17159.
- [16] R.-Z. Li, A. Hu, T. Zhang, K.D. Oakes, Direct writing on paper of foldable capacitive touch pads with silver nanowire inks, *ACS Appl. Mater. Interfaces* 6 (23) (2014) 21721–21729.
- [17] S. Das, D. Cormier, S. Williams, Potential for multi-functional additive manufacturing using pulsed photonic sintering, *Procaccia Manuf.* 1 (2015) 366–377.
- [18] G.L. Fiore, F. Jing, J.K.C. Young, C.J. Cramer, M.A. Hillmyer, High T_g aliphatic polyesters by the polymerization of proxiolactide derivatives, *Polym. Chem.* 1 (6) (2010) 870–877.
- [19] D.J. Finn, M. Lotya, J.N. Coleman, Inkjet printing of silver nanowire networks, *ACS Appl. Mater. Interfaces* 7 (17) (2015) 9254–9261.
- [20] M.S. Saleh, J. Li, J. Park, R. Panat, 3D printed hierarchically porous microtactile electrode materials for exceptionally high specific capacity and areal capacity lithium ion batteries, *Addit. Manuf.* 23 (2018) 70–78.
- [21] K. Dharmadasa, B. Lavery, M. Dharmadasa, T. Duffell, Intense pulsed light treatment of cadmium telluride nanoparticle-based thin films, *ACS Appl. Mater. Interfaces* 6 (7) (2014) 5034–5040.
- [22] H.-S. Kim, S.R. Dhage, D.-E. Shim, H.T. Hahn, Intense pulsed light sintering of copper nanoinks for printed electronics, *Appl. Phys. A* 97 (4) (2009) 791–798.
- [23] J.S. Kang, J. Ryu, H.S. Kim, H.T. Hahn, Sintering of inkjet-printed silver nanoparticles at room temperature using intense pulsed light, *J. Electron. Mater.* 40 (11) (2011) 2268–2277.
- [24] S. Bansal, C.-H. Chang, R. Malhotra, The coupling between densification and optical heating in intense pulsed light sintering of silver nanoparticles, *American Society of Mechanical Engineers Manufacturing Science and Engineering Conference*, Blacksburg, Virginia, USA, 2016, p. 10.