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# Supplementary Materials for

## Ultrahigh areal number density solid-state on-chip microsupercapacitors via electrohydrodynamic jet printing

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**The PDF file includes:**



**Other Supplementary Material for this manuscript includes the following:**

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**Supplementary Materials**

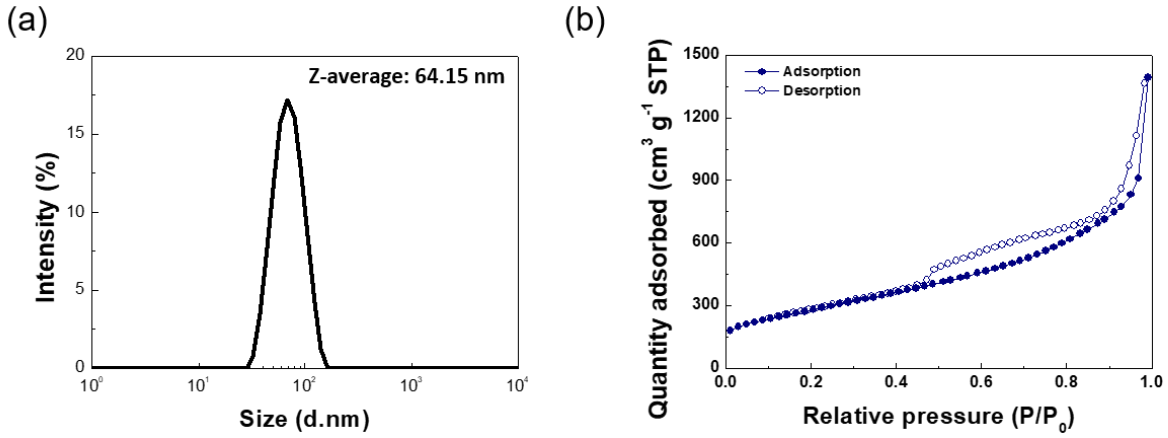
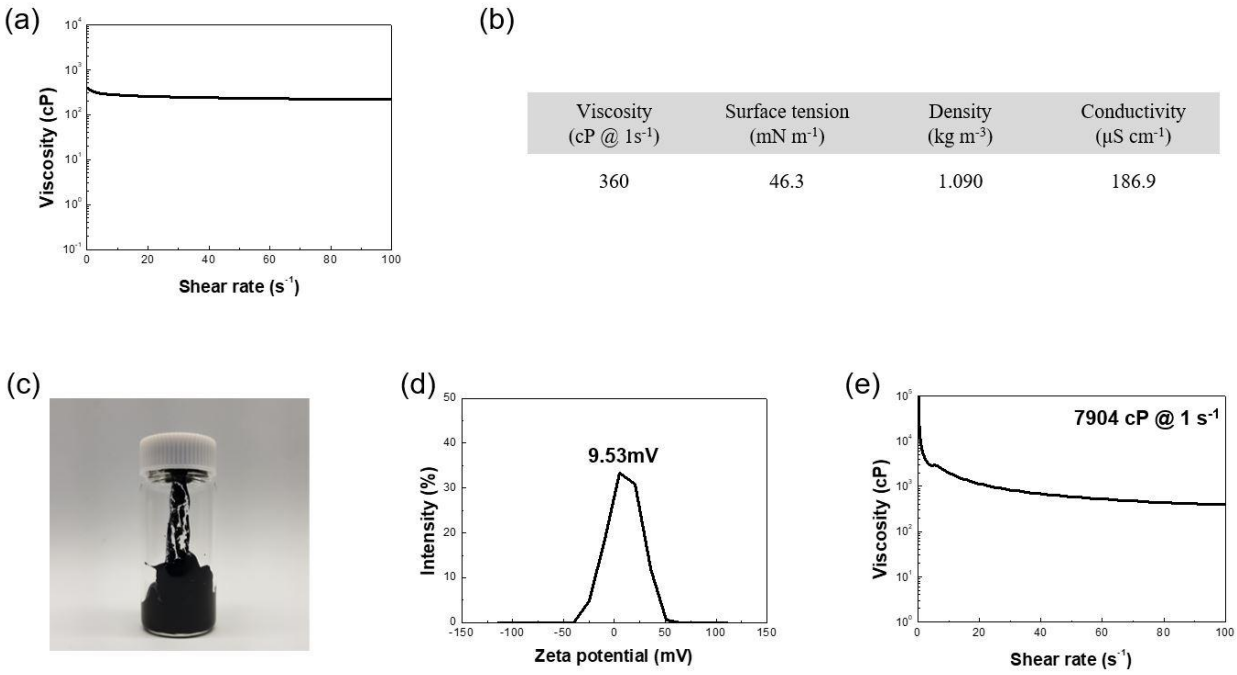


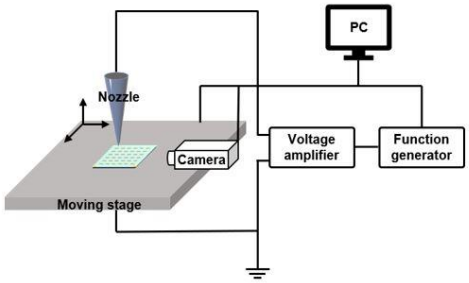

**Fig. S1. Basic characteristics of nAC.** (**a**) Particle size distribution. (**b**) Nitrogen gas adsorption-desorption isotherms at 77K.



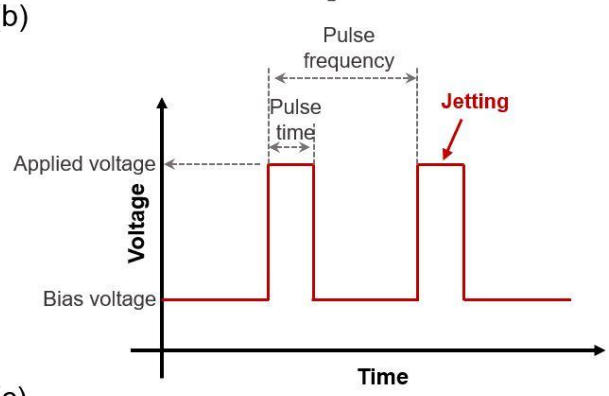

| Viscosity (cP @ 1s$^{-1}$) | Surface tension (mN m$^{-1}$) | Density (kg m$^{-3}$) | Conductivity (μS cm$^{-1}$) |
|---|---|---|---|
| 360 | 46.3 | 1.090 | 186.9 |

**Fig. S2. Characteristics of the electrode ink with (a,b) and without (c-e) CMC binder.** (a) Viscosity as a function of shear rate. (b) Summary of other major physical properties. (c) Photograph. (d) Zeta potential profile. (e) Viscosity as a function of shear rate. Photo credits: (c) Kwon-Hyung Lee, Ulsan National Institute of Science and Technology.

(a)




(b)




(c)

| Parameter | Value |
|---|---|
| Applied voltage (kV) | 0.42 ~ 0.44 |
| Bias voltage (kV) | 0.1 |
| Pulse time (ms) | 0.2 |
| Pulse frequency (Hz) | 20 |
| Printing speed (mm/s) | 0.06 ~ 0.1 |
| Distance between nozzle and substrate (μm) | 15 |

**Fig. S3. Process description of the EHD jet printing for the fabrication of interdigitated electrodes.** (**a**) Schematic representation of the EHD jet-printing equipment. (**b**) The pulse DC signal as a function of time. (b) Summary of the optimized process parameters for the EHD jet-printed electrode.

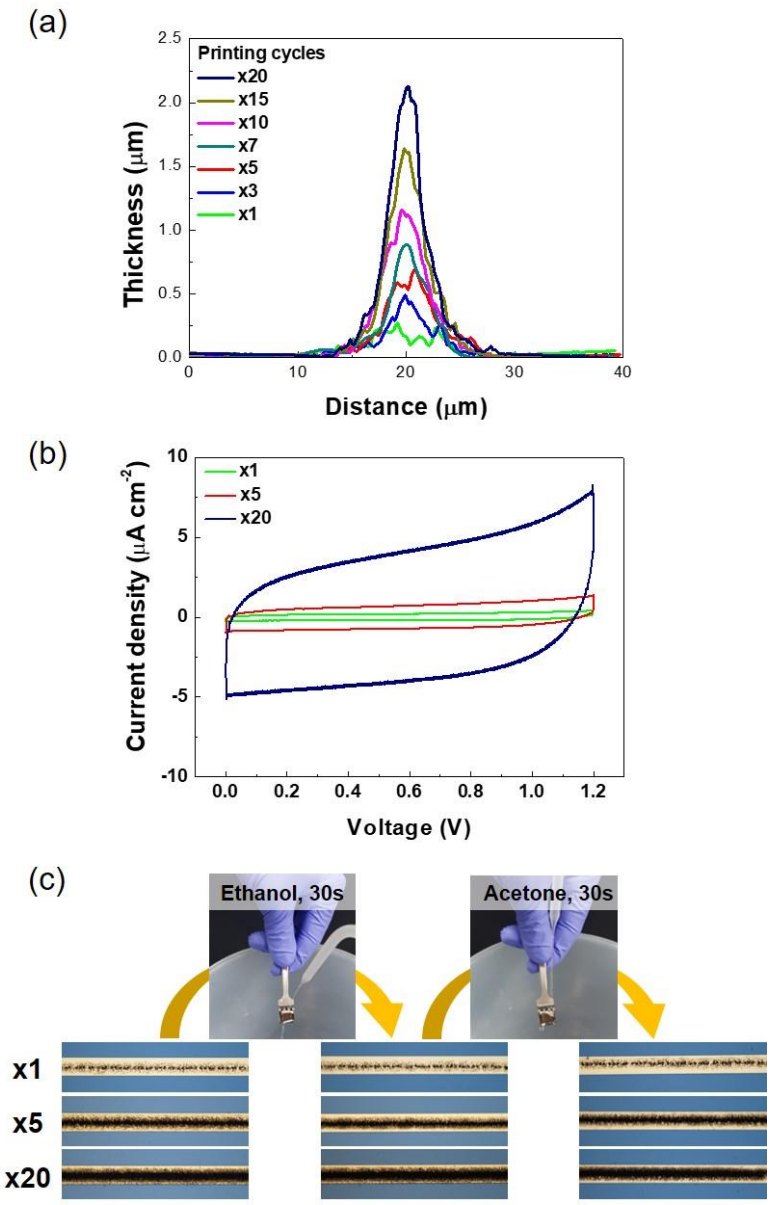
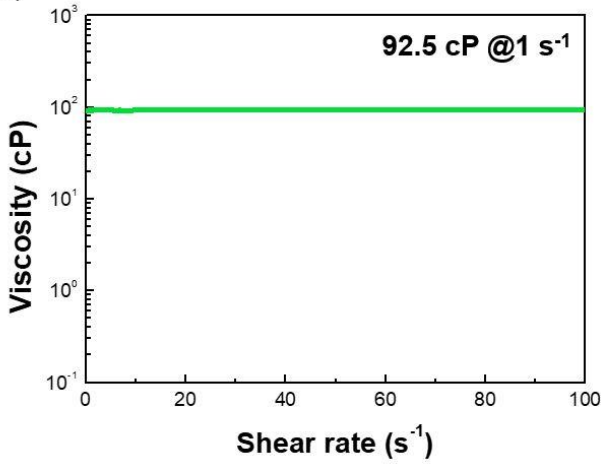



**Fig. S4. Characterization of the EHD jet-printed electrodes as a function of printing cycles.** (**a**) Atomic force microscopy (AFM) depth profiles. (**b**) CV profiles (scan rate: 50 mV $s^{-1}$) of the SS–MSC. (**c**) Optical microscopy images after the washing test (ethanol followed by acetone, exposure time = 30 s, respectively). Photo credits: (c) Kwon-Hyung Lee, Ulsan National Institute of Science and Technology.

(a)




(b)

| Parameter | Value |
|---|---|
| Applied voltage (kV) | 0.75 ~ 1.2 |
| Bias voltage (kV) | 0.5 ~ 0.8 |
| Pulse time (ms) | 1 |
| Pulse frequency (Hz) | 333 |
| Printing speed (mm/s) | 0.2 ~ 0.3 |
| Distance between nozzle and substrate (μm) | 100~50 |
| Pump speed (μl min$^{-1}$) | 0.5 ~ 1 |

**Fig. S5. EHD jet printing of the electrolyte ink.** (**a**) Viscosity of the electrolyte ink (*i.e.*, before UV curing) as a function of shear rate. (**b**) Summary of the optimized process parameters for the electrolyte preparation.

(a)

(b)

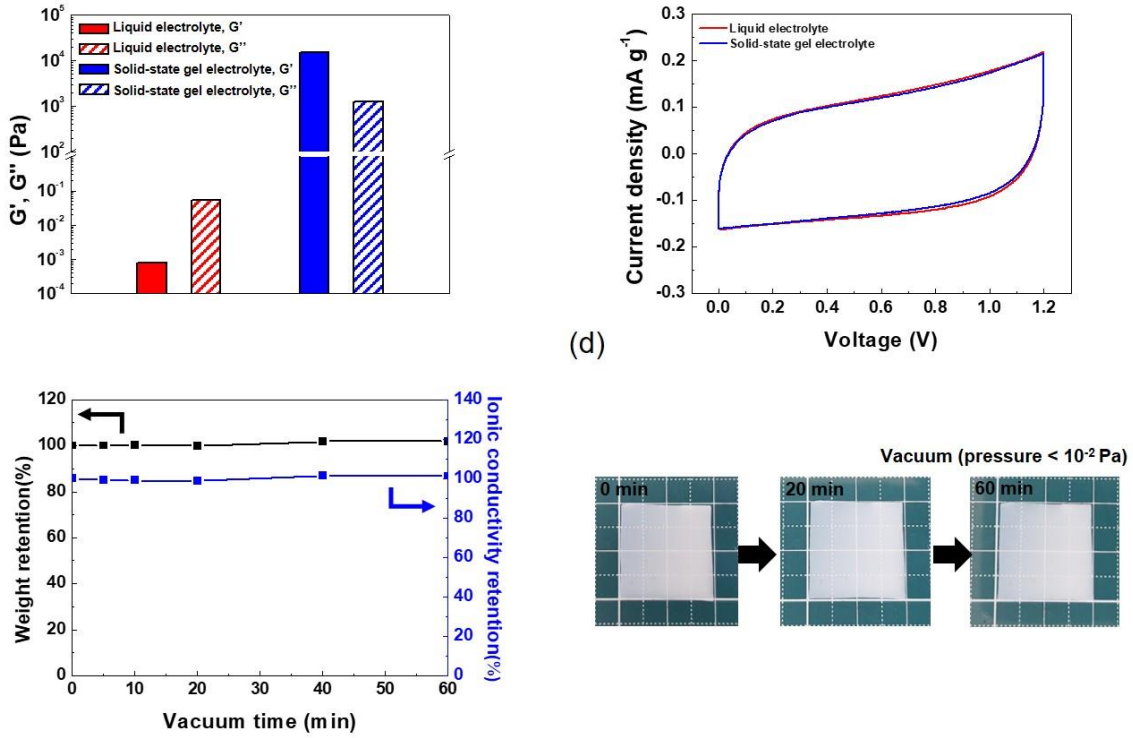



**Fig. S6. Characteristics of the solid-state gel electrolytes.** (**a**) Viscoelastic properties (G' and G'') of the electrolyte ink (*i.e.*, before UV irradiation) and solid-state gel electrolyte (after UV irradiation) at an oscillation frequency 0.1 Hz. (**b**) CV profiles (scan rate: 20 mV $s^{-1}$) of the model cells: solid-state gel electrolyte vs. liquid electrolyte ([EMIM][TFSI] alone). (**c**) Change in the weight and ionic conductivity and (**d**) photographs upon exposure to vacuum state (pressure < $10^{-2}$ Pa) as a function of vacuum time. Photo credits: (d) Kwon-Hyung Lee, Ulsan National Institute of Science and Technology.

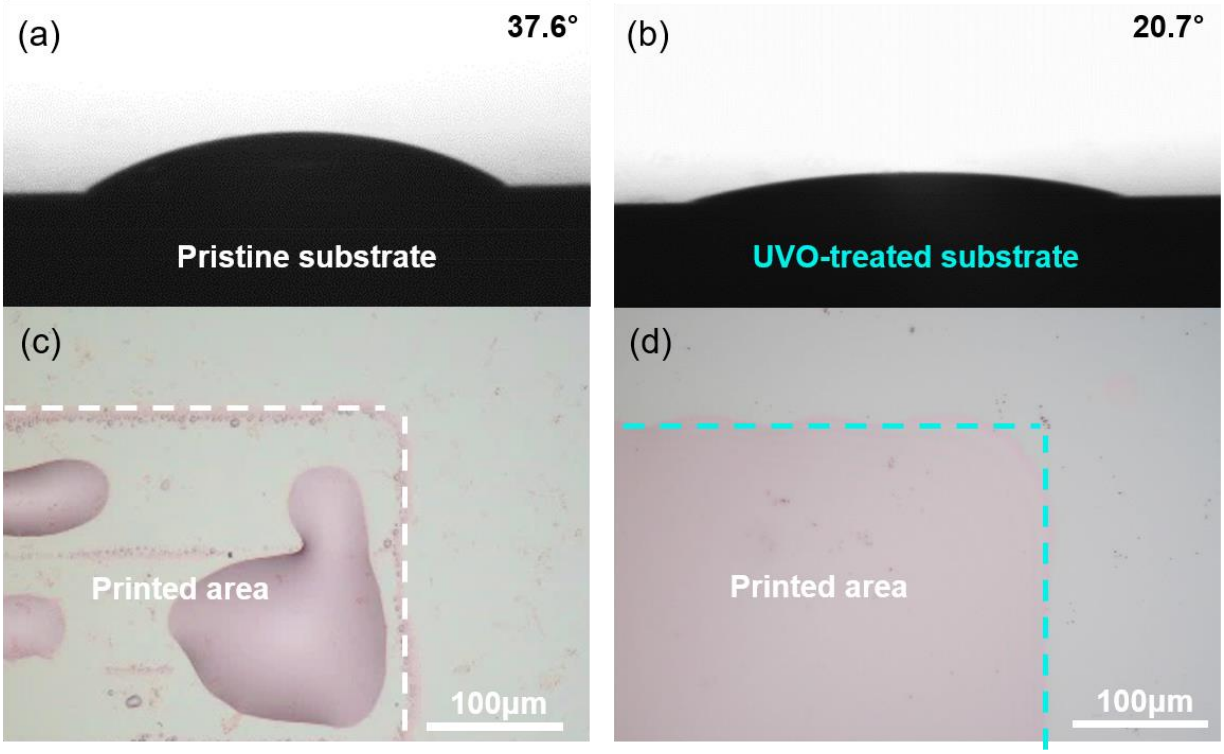
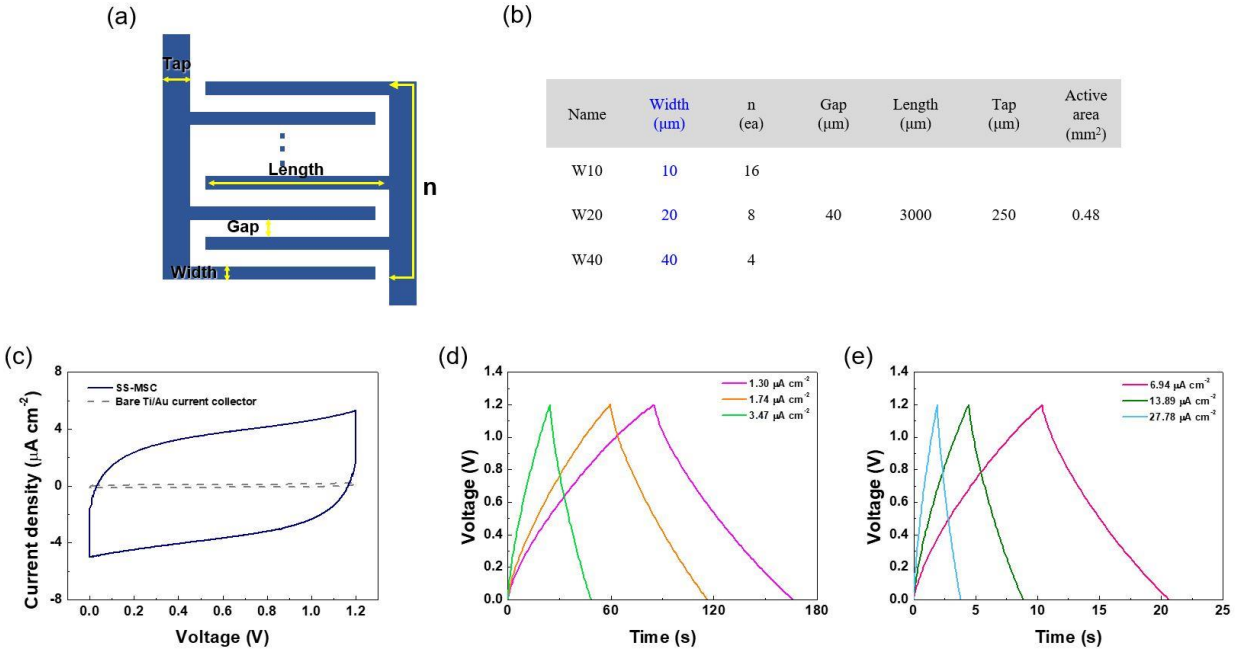



**Fig. S7. Photographs showing contact angle change and effect on the affinity between the electrolyte ink and $SiO_2$/Si chip substrate.** (**a**,**c**) Pristine $SiO_2$/Si chip substrate. (**b**,**d**) UVO-treated $SiO_2$/Si chip substrate.




| Name | Width (μm) | n (ea) | Gap (μm) | Length (μm) | Tap (μm) | Active area ($mm^2$) |
|---|---|---|---|---|---|---|
| W10 | 10 | 16 | | | | |
| W20 | 20 | 8 | 40 | 3000 | 250 | 0.48 |
| W40 | 40 | 4 | | | | |

**Fig. S8. Configuration (a,b) and electrochemical performance (c-e) of the SS–MSC.** (a) Schematic representation. (b) Dimension values as a function of electrode width. (c) Comparison of the CV profiles (scan rate: 50 mV $s^{-1}$): SS–MSC vs. bare Ti/Au current collector. (d–e) GCD profiles of the SS–MSC at varied current densities of 1.30 ~ 27.78 μA $cm^{-2}$.

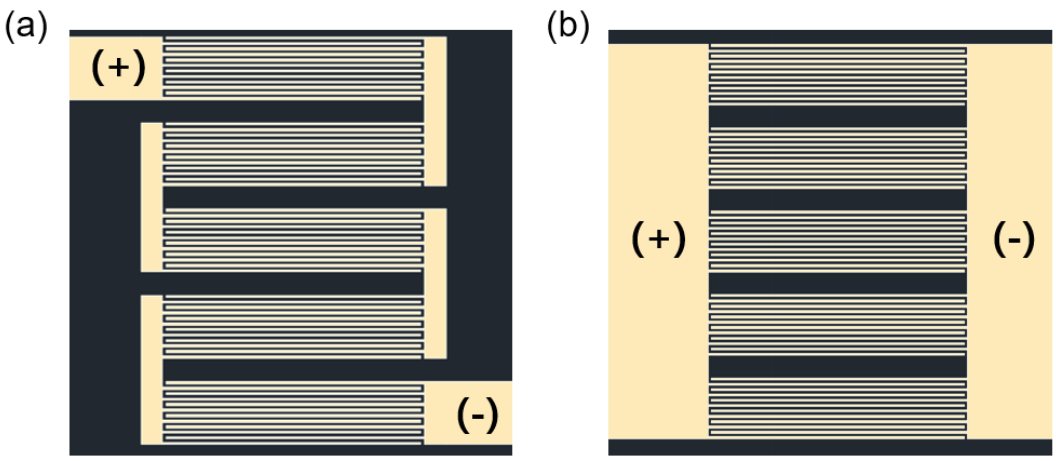



**Fig. S9. Schematic representation of the five SS–MSCs.** (**a**) In-series connection. (**b**) In-parallel connection.

**Table S1. Comparison in the major characteristics between the on-chip UHD SS–MSCs (this study) and the previously reported MSCs fabricated by printing techniques.**

| Fabrication technique | Feature size (μm) | # of cells (n) | Unit cell area ($cm^2$) | Cell-to-cell distance (mm) | Cell voltage (V) | Areal-number-density of cells (cells $cm^{-2}$) | Areal-operating-voltage (V $cm^{-2}$) | Reference |
|---|---|---|---|---|---|---|---|---|
| ***EHD printing*** | ***10*** | ***36*** | ***0.0127*** | ***0.22*** | ***43.2*** | ***54.9*** | ***65.9*** | ***This study*** |
| Inkjet printing/ electrodeposition | 35 | 3 | N/A | N/A | 3.2 | 0.84* | 0.89* | [16] |
| Inkjet printing | 50 | 2 | 2* | N/A | 1.6 | 1** | 0.8** | [18] |
| Inkjet printing | 300 | 2 | 1.2 | 12* | 1.8 | 0.51* | 0.46 | [20] |
| Inkjet printing | 300* | 5 | 0.86* | 0.5* | 10 | 1.26* | 2.7* | [22] |
| Inkjet printing | 300 | 144 | 0.2* | 0.85* | 12 | 3.16* | 0.40* | [37] |
| Inkjet printing | 1000 | 4 | 3 | N/A | 3.2 | 0.33** | 0.26** | [17] |
| 3D printing | 250 | 1 | N/A | N/A | 1.0 | N/A | N/A | [38] |
| 3D printing | 250 | 1 | N/A | N/A | 0.8 | N/A | N/A | [39] |
| Screen printing | 600 | 130 | 29.1 | 7.5* | 104 | 1.08* | 0.87* | [40] |

N/A: Not available.
* These values were obtained using an image analysis technique.
** Normalized values based on unit cell area.

**Movie S1. Video clip showing the fabrication of interdigitated electrodes on top of the $SiO_2$/Si chip substrate through the EHD jet printing.**

**Movie S2. Video clip showing the fabrication of solid-state gel electrolytes on top of interdigitated electrodes-deposited $SiO_2$/Si chip substrate through the EHD jet printing.**

*Note that all movies are playing four times faster than real-time for readers' better understanding.