#### **Supporting information**

#### Segregation-controlled Self-assembly of Silver Nanowire Networks using a

#### **Template-free Solution-based Process**

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#### **Results and discussion (Supplementary Material)**

**Morphology of SGAgNW-coated PET film.** Fig. S8 shows the snapshot images of the AgNW conducting thin films on PET that were connected with copper electrodes (Dimension: 7 cm × 9 cm) with a high optical transparency. The film transparency is slightly darker with higher AgNW content, whereas the university emblem located behind the film is clearly visible.

The coating thickness was estimated to be one or two wire sizes using cross-sectional TEM images and EDS mapping of Pt and Ag (Fig. S9). A total of 40–50 nm of AgNW was coated on the PET film—except for the Pt coating layer—for high-resolution TEM measurements; the nanowires overlapped.

**Heating performance: Temperature distribution.** Fig. S10 shows the time-dependent temperature profiles of the SGAgNWbased transparent thin-film heater on the PET film with respect to the applied voltage. A constant DC bias voltage was applied to the two ends of the electrode; the temperature distribution was investigated using temperature probes. The TE film heater exhibits a heating property proportional to the applied voltage. In addition, a fast response time of less than 3 min was observed to achieve the saturated temperature (0.8 Ts) under all conditions while maintaining a stable temperature. The heat generation reached 34.5 °C, 46.0 °C, and 64.3 °C under the application of 3 V, 5 V, and 7 V, respectively; This could then increase to 94.5 °C when a DC voltage of 10 V was applied. This verifies that the present film heater can achieve a well-controlled heating property controlled by the electrical voltage. According to Joule heating, the following equation can determine the electro-to-thermal power, where I denotes the electric current flowing through a conductor, *V* represents the applied voltage, and *R* represents the electrical resistance of the heater <sup>1, 2</sup>. As described in equation (3), a lower electrical resistance can generate a better heating performance.

$$P = IV = \frac{V^2}{R},\tag{3}$$

**Thermal stability: Heating and cooling cycles.** The reliability and thermal stability of the transparent thin-film heaters were evaluated using heating and cooling cycles. The temperature distribution was investigated under five repeated cycles for five times with an operating time of 20 min (Fig. S11). Considering the return time to reach the initial state, the cooling interval was set to 20 min at 5 V, and the turn-off time was set to 40 min at 7 V. The temperature profiles were maintained at a maximum steady-state temperature similar to the initial saturated temperature. The heating and cooling rates were measured by plotting the derivative of the temperature versus time. As shown in the slope graph, the heating performance appears identically during the test, which confirms that the AgNW-coated heaters on the PET film can repetitive operation with superior thermal stability at a low input voltage below 10 V.

#### Reference (Supplementary Material)

1. De, S.; Higgins, T. M.; Lyons, P. E.; Doherty, E. M.; Nirmalraj, P. N.; Blau, W. J.; Boland, J. J.; Coleman, J. N., Silver Nanowire Networks as Flexible, Transparent, Conducting Films: Extremely High DC to Optical Conductivity Ratios. *ACS nano* **2009**, *3* (7), 1767-1774.

2. Kim, D.-H.; Cho, K.-S.; Kim, H.-K., Thermally evaporated indium-free, transparent, flexible SnO2/AgPdCu/SnO2 electrodes for flexible and transparent thin film heaters. *Scientific Reports* **2017**, *7* (1), 2550.

# List of Supporting Figures and Tables

**Figure S1.** SEM images and diameter size distribution of silver NWs. (a-b) High aspect ratio and (c-d) low aspect ratio.

Figure S2. XRD patterns of silver NWs. (a) High aspect ratio and (b) low aspect ratio.

Figure S3. Contact angle of the surface on the substrates (before and after hydrophilic treatment).

**Figure S4.** OM images of the self-assembled AgNWs for low aspect ratio of  $(a_1) 0.25 \text{ wt\%}$ ,  $(a_2) 0.5 \text{ wt\%}$ ,  $(a_3) 1.0 \text{ wt\%}$ ,  $(a_4) 2.0 \text{ wt\%}$ ,  $(a_5) 3.0 \text{ wt\%}$  and high aspect ratio of  $(b_1) 0.25 \text{ wt\%}$ ,  $(b_2) 0.5 \text{ wt\%}$ ,  $(b_3) 1.0 \text{ wt\%}$ ,  $(b_4) 2.0 \text{ wt\%}$ ,  $(b_5) 3.0 \text{ wt\%}$ .

**Figure S5.** SEM images of the self-assembled AgNWs for the low aspect ratio of  $(a_1) 2.0$  wt%.  $(a_2) 3.0$  wt%.

Figure S6. Haze values of the self-assembled AgNWs. (a) Low aspect ratio and (b) high aspect ratio.

Figure S7. OM images of the damaged PET films (Inset: snap shot of damaged film).

**Figure S8.** Optical images of the SGAgNW transparent thin film heaters; (a) 0.1 and (b) 0.3 wt.% of AgNW.

**Figure S9.** (a) Cross-sectional TEM image, (b) STEM image, and (c) EDS mapping of Pt sputtered SGAgNWs-coated PET transparent thin film heater after FIB milling.

**Figure S10.** Temperature piles of the SGAgNW-based transparent thin film heaters with respect to the applied voltages (3, 5, 7, and 10 V).

**Figure S11.** Thermal stability for transparent thin film heater (a) at the applied voltage of 7 V and (b) plot of the derivative of temperature versus time.

#### **List of Supporting Tables**

**Table S1.** Size distribution, optical, and electrical properties of the self-assembled AgNWs with different aspect ratios.

**Table S2.** Comparison of different coating methods to produce the conductive patterns based on conducting nanomaterial inks for transparent electrodes.

**Table S3.** Properties of the organic solvents used in the silver ink solution.



5

Figure S2.



Figure S3.



### Figure S4.







Figure S6.



Figure S7.



## Figure S8.



# Figure S9.



# Figure S10.



Figure S11.



| Tabl | e S1. |
|------|-------|
|------|-------|

|                            | Ag (wt %) | Cell size (µm)  | Cell-to-cell<br>distance (µm) | Transmittance<br>at 550 nm (%) | Haze (%)     | R <sub>s</sub> (Ω/□ ) |
|----------------------------|-----------|-----------------|-------------------------------|--------------------------------|--------------|-----------------------|
| a. Low<br>aspect<br>ratio  | 0.25      | 200.32 ± 38.17  | 15.81 ± 4.25                  | 95.03                          | 2.44 ± 0.34  | 1951.98±840.26        |
|                            | 0.5       | 145.38 ± 27.31  | 22.37 ± 9.39                  | 87.08                          | 3.71 ± 0.48  | $21.36 \pm 3.24$      |
|                            | 1.0       | 126.08 ± 21.96  | 70.85 ± 36.17                 | 81.13                          | 6.61 ± 0.57  | $20.26 \pm 8.06$      |
|                            | 2.0       | 3.89 ± 1.00     | 22.55 ± 6.93                  | 74.83                          | 8.03 ± 0.43  | $7.70 \pm 0.18$       |
|                            | 3.0       | 1.27 ± 0.34     | 25.32 ± 7.76                  | 54.83                          | 14.44 ± 0.18 | $2.95 \pm 0.12$       |
| b. High<br>aspect<br>ratio | 0.25      | 344.68 ± 159.98 | 15.12 ± 6.39                  | 98.68                          | 0.60 ± 0.21  | -                     |
|                            | 0.5       | 121.90 ± 32.65  | 13.95 ± 6.53                  | 96.46                          | 1.21 ± 0.22  | $2165.28 \pm 1425.87$ |
|                            | 1.0       | 78.96 ± 20.22   | 20.17 ± 8.87                  | 89.56                          | 5.13 ± 0.61  | $1506.76 \pm 540.40$  |
|                            | 2.0       | 84.41 ± 34.76   | 36.56 ± 13.70                 | 88.42                          | 6.40 ± 0.53  | $25.44 \pm 6.15$      |
|                            | 3.0       | 106.09 ± 39.02  | 43.88 ± 19.20                 | 82.96                          | 7.31 ± 0.26  | $9.71 \pm 0.16$       |

#### Table S2.

| Coating method               | Material | %T<br>(%) | R <sub>s</sub><br>(ohm/sq) | Figure<br>of merit | Sintering condition                        | Haze<br>(%) | Reference<br>[year] |
|------------------------------|----------|-----------|----------------------------|--------------------|--|-------------|---------------------|
| Nancimprinting               | ANNIN    | 68        | 29                         | 30.6               | Plasma sintering                           | 2.7         | Ref. 10<br>[2019]   |
| Nanoimprinting               | Aunw     | 92        | 227                        | 19.5               | $(5\% H_2 \text{ in Ar}, 15 \text{ min.})$ | 1.6         |                     |
| Recrystallized               |          | $\sim 78$ | 18.8                       | 75.8               | 80 °C/20                                   |             | Ref. 22<br>[2018]   |
| ice crystal                  | Aginw    | ~ 88      | 192.4                      | 14.8               | 80 C/30 mm.                                |             |                     |
| Air-water<br>interface       | AuNW     | ~ 92      | 130.1                      | 34.0               | Plasma treatment<br>(Ar, 20 min.)          |             | Ref. 24<br>[2013]   |
| Nanosecond<br>laser ablation | Cu       | 83        | 17.48                      | 110.4              |  |             | Ref. 25<br>[2009]   |
| Random crack                 | AgNW     | 85        | 92                         | 24.2               |  |             | Ref. 27<br>[2017]   |
| PDMS template                | CNT      | 78        | 264                        | 5.4                | 50 °C/4 h.                                 |             | Ref. 28<br>[2016]   |
| Laser sintering              | AgNP     | 85        | 30                         | 74.2               |  |             | Ref. 29<br>[2016]   |
| Template-free                | AgNW     | 87.08     | 21.36                      | 123                | IPL sintering                              | 3.71        | This work           |
| self-assembly                |          | 88.42     | 25.44                      | 117                | (1000 kW, 2000 µs)                         | 6.40        |                     |

#### Table S3.

|                                | α-terpineol | Water | Methanol            |
|--------------------------------|-------------|-------|---------------------|
| Molecular structure            |             | HH    | H <sub>3</sub> C—OH |
| Viscosity (at 20°C) [mPa.s]    | 67          | 1.001 | 0.594               |
| Solubility in water [g/L]      | 2.42        | -     | miscible            |
| Boiling point [°C]             | 220         | 99.98 | 64.7                |
| Vapor pressure (at 25°C) [kPa] | 0.0037      | 3.169 | 13.02               |