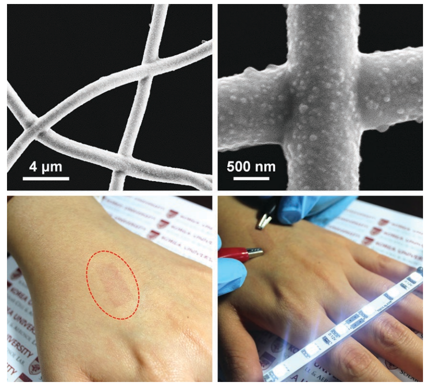
# COMMUNICATIONS

## Thin Films

S. An, H. S. Jo, D.-Y. Kim, H. J. Lee, B.-K. Ju, S. S. Al-Deyab, J.-H. Ahn, Y. Qin, M. T. Swihart, A. L. Yarin,* S. S. Yoon* .................................... X–XX

**Self-Junctioned Copper Nanofiber Transparent Flexible Conducting Film via Electrospinning and Electroplating**




**Self-junctioned copper nanofiber transparent flexible films** are produced using electrospinning and electroplating processes that provide high performances of $T = 97\%$ and $R_s = 0.42\ \Omega\ sq^{-1}$ by eliminating junction resistance at wire intersections. The film remains conductive after being stretched by up to 770% (films with $T = 76\%$) and after 1000 cycles of bending to a 5 mm radius.

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# Self-Junctioned Copper Nanofiber Transparent Flexible Conducting Film via Electrospinning and Electroplating

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Numerous existing and emerging devices, including light-emitting diodes (LEDs),[1] organic LEDs,[2] displays,[3] touch screens,[4] solar cells,[5] smart windows,[6] and interactive electronics[7,8] require transparent conducting electrodes (TCEs) that combine high transparency ($T$) with low sheet resistance ($R_s$). These requirements are mutually incompatible, as low $R_s$ relies on high mobility charge carriers that inevitably interact with light and reduce $T$. Indium-doped tin oxide (ITO), a mechanically stable and reliable material that provides a reasonable tradeoff between $R_s$ and $T$, dominates commercial TCE applications. However, the rising cost and diminishing supply of indium, along with the rigidity and brittleness of ITO, limit its use in low-cost flexible devices such as wearable electronics, flexible solar cells, roll-up displays, and electronic skins.[9]

Nanostructured materials including carbon nanotubes (CNTs),[3,10,11] graphene,[4,12–14] metal nanofibers,[9,15–17] and conductive polymers[18] have been proposed as ITO replacements. For CNT films, $R_s$ is typically an order of magnitude higher than that of ITO.[19,20] Chemical vapor deposition (CVD) grown graphene can outperform ITO,[4] but requires a multi-step growth-and-transfer fabrication process that limits its applicability. Metal nanowires (NWs) show great promise, achieving high transparency using nanostructures smaller than the wavelengths of visible light, while metallic conductivity allows low $R_s$ with sparse NW coverage. Metal NW films have achieved $R_s = 10\ \Omega\ \text{sq}^{-1}$ at $T \geq 90\%$.[21,22] However, further improvement has been limited by high contact resistance at NW intersections. This junction resistance is the central challenge for all 1D percolative materials, including those based on CNTs. Eliminating it requires producing a continuous mesh without contact resistance at wire intersections. Previous studies have applied thermal post-processing or mechanical pressing to merge these junctions.[23,24] However, these methods can damage the NWs and are incompatible with many low-cost, flexible substrates.

Wu et al. introduced copper nanofibers whose junctions were interconnected by thermally evaporated copper,[17] and were the first to achieve $R_s < 10\ \Omega\ \text{sq}^{-1}$ with $T > 90\%$ using metal nanowires. Hsu et al. achieved $R_s = 0.36\ \Omega\ \text{sq}^{-1}$ at $T = 92\%$ by combining mesoscale nanofibers with nanoscale metal wires.[16] However, the thermal evaporation used to link the nanowires with nanofibers is a high-vacuum process that may not be practical for low-cost production. Moreover, thermal post-treatment precludes the use of flexible low-cost polymer substrates for flexible, stretchable, and rollable optoelectronic devices. Here we break through these limitations with an ultra-fast process for fabricating TCEs with a world record combination of low $R_s$ and high $T$. This is achieved through electrospinning and electroplating for only a few seconds each, combining two high-throughput commercially viable processes. When transferred to an Eco-flex substrate, these TCEs can be stretched by 580% with less than a factor of 5 increase in $R_s$, and can withstand more than 1000 cycles of bending to a 5 mm radius without appreciable increase in $R_s$. The performance demonstrated here exceeds that achieved by Hsu et al., while also eliminating the use of silver and maintaining compatibility with virtually any substrate, including thermally sensitive polymers. The new approach demonstrated here could readily be incorporated into a continuous, roll-to-roll manufacturing sequence for low-cost, large-scale fabrication, because it can be implemented using only low-temperature, nonvacuum processing. Furthermore,

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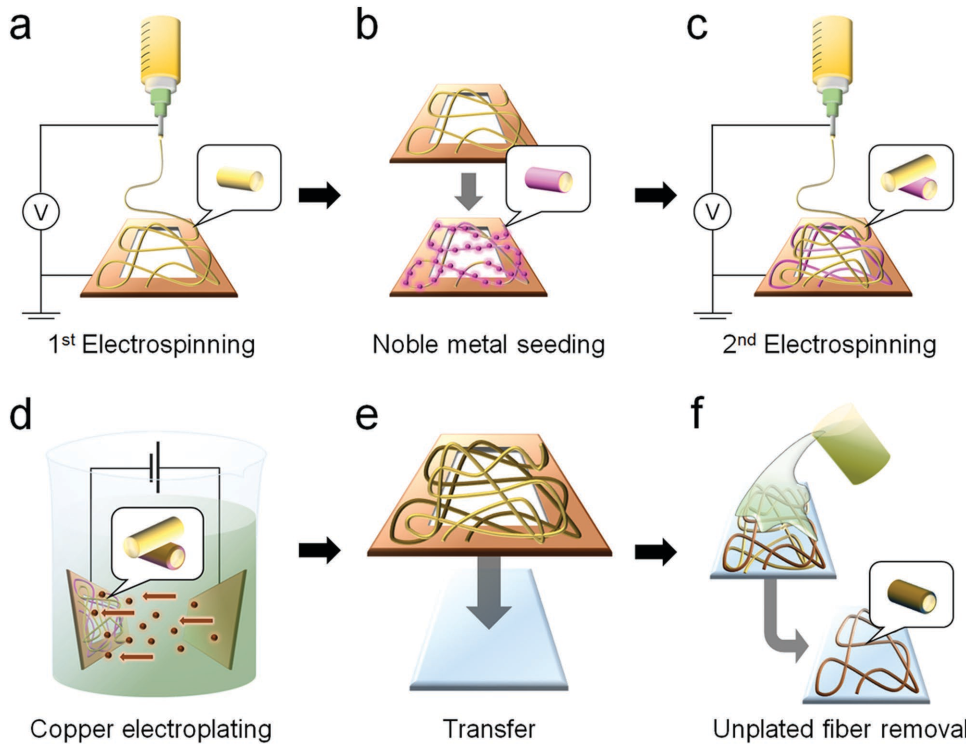


**Figure 1.** Schematic of the fabrication process for CuEW-TCEs. a) The first electrospun polymer nanofibers are deposited onto a copper frame. b) The nanofibers are made conductive by noble metal seeding. c) The second layer of electrospun polymer nanofibers is deposited above the platinum-seeded nanofibers. d) Only the metal-seeded nanofibers are electroplated by copper. (The second layer of electrospun nanofibers is not electroplated, because without metal seeding they are nonconductive). e) The electroplated and nonelectroplated nanofibers are transferred to a transparent substrate. f) The nonelectroplated nanofibers are removed by dissolution.

we introduce a new model based on the renormalization technique of percolation theory, which relates the electrical conductivity and optical transmittance for the nanowire network.

Production of copper electroplated nanowire meshes for transparent conducting electrodes (CuEW-TCEs) is illustrated in **Figure 1**. In electrospinning (Figure 1a,c), most of the solvent (dimethylformamide, DMF) is evaporated in flight and the solidified polymer (polyacrylonitrile, PAN) forms a self-intersecting mat from a single continuous nanofiber.[25] In the process of electroplating (Figure 1d), fiber junctions are bonded via metal deposition on the outer surface of the platinum-seeded (Figure 1b) nanofibers. The electroplated copper fills the space around the crossed nanofibers (**Figure 2**a), which dramatically reduces contact resistance at the junctions. Figure S2 (Supporting Information) shows cross-sections of the electroplated copper within a junction, demonstrating that the intersection is completely filled with dense electrodeposited copper, except for small pores corresponding to the original nanofibers. Drying in an inert atmosphere after electroplating is essential to the performance of the CuEW-TCEs. As shown in Figure S3 (Supporting Information), air drying produced substantial oxidation of the NWs which substantially reduced conductivity. However, once the NWs have been dried in an inert atmosphere, they are stable in air and their performance does not degrade upon air exposure. For these reasons, excellent electrical performance of CuEW-TCEs can be maintained even at locations such as human skin or a leaf (Figure 2b, Movie S1, Supporting Information). We measured single CuEW resistivity by transferring a CuEW to a bare substrate and contacting its ends with silver paint (Figure S4, Supporting Information). Measurement on a single wire of ≈1.8 µm diameter and ≈250 µm length gave a resistivity of $6.7 \times 10^{-8}$ Ω m. This is within a factor of four of the resistivity of bulk copper, and is quite reasonable given the small diameter, rough surface, nanoscale grain size, possible surface oxidation, and presence of pores due to the electrospun fiber template within the CuEWs. This measurement provides a lower limit of the conductivity, because it is a two-point measurement that does not account for contact resistance between the wire and the silver paint electrodes or leads.

**Figure 3** shows $T$ versus $R_s$ for the CuEW-TCEs on a polydimethylsiloxane (PDMS) substrate. $T$ decreases from 97% to 41% as the first electrospinning time ($t_{1st}$) increases from 1 to 180 s (SEM images are shown in Figure S5, Supporting Information), while $R_s$ only decreases slightly, from 0.42 to 0.31 Ω sq$^{-1}$. Note that here the $t_{1st}$ was varied at fixed electroplating time (3 s) and voltage (3 V). Thus, the total amount of copper deposited does not increase in proportion to $t_{1st}$. Even though only a few CuEWs are deposited for $t_{1st} = 1$ s, $R_s$ was already quite low, an outcome we attribute to the low junction resistance. In fact, $R_s$ can be decreased to 0.138 Ω sq$^{-1}$ when the electroplating time is increased to 7 s. However, $T$ decreases as the electroplating time increases (Figure S6, Supporting Information). Conversely, electroplating times below 3 s were insufficient to achieve complete electroplating. These results are superior to



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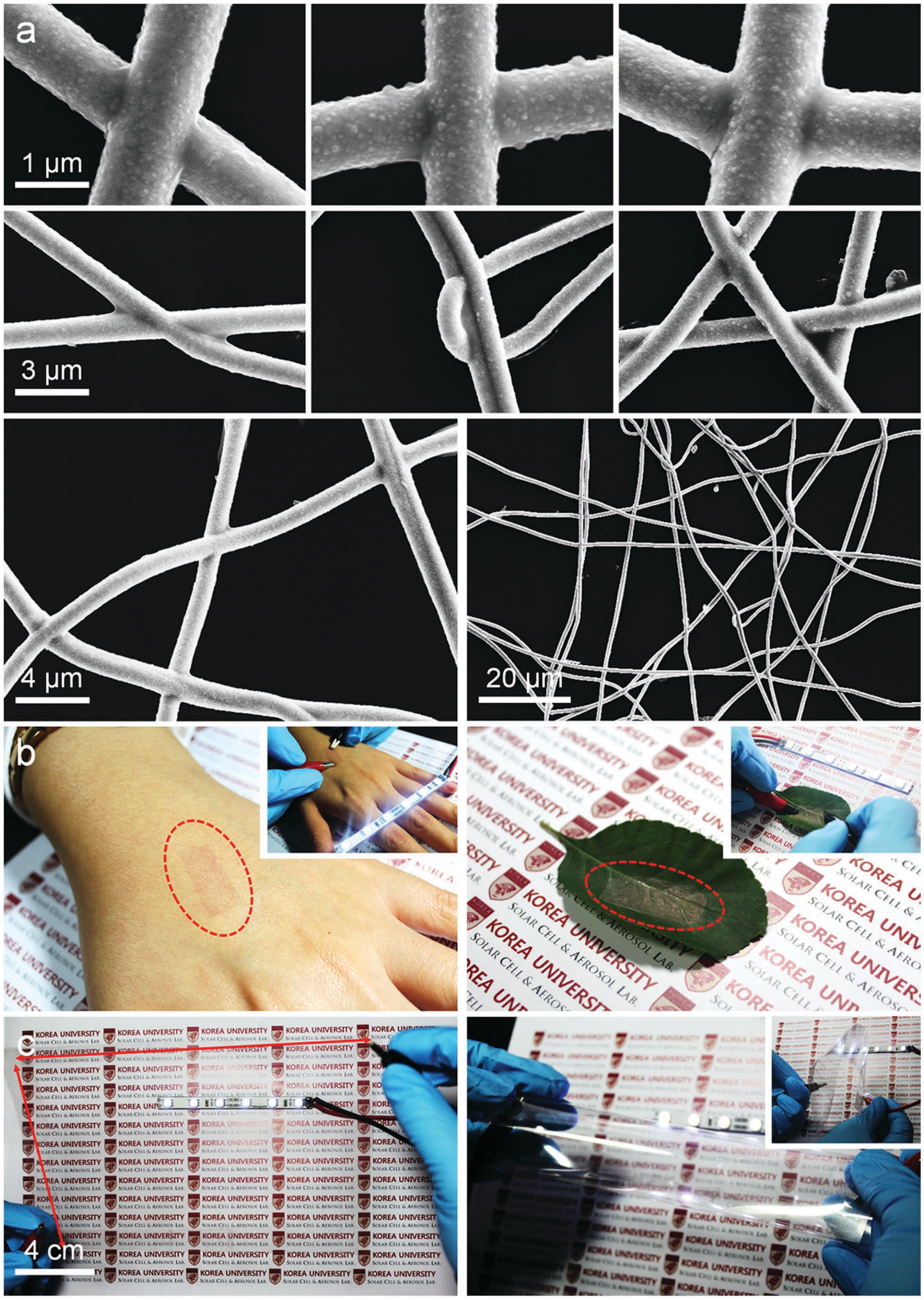


**Figure 2.** Copper electroplated wire (CuEW) for transparent conducting electrodes (TCEs). a) SEM images of CuEW-TCEs on PDMS. b) Images of CuEW-TCEs (duration of the first electrospinning stage $t_{1st} = 30$ s) transferred to a human hand and a leaf (LED operation is shown in Movie S1, Supporting Information). c) Images of large-scale CuEW-TCEs on PET ($t_{1st} = 10$ s).

those reported in prior studies of Cu nanotroughs,[17] AuNFs,[9] graphene,[4] PEDOT:PSS,[22] SWNT,[26] and AgNW[27] as shown in Figure 3a. The removal of unplated support fibers (Figure 1f) is essential for achieving these high values of *T*. Although these nanofibers play an important role in protecting the metal-seeded nanofibers during electroplating, they scatter a significant amount of light; removing them significantly increases the transmittance (by 30%, Figure S7, Supporting Information).

Figure 3e shows the transmittance spectra of all of the CuEW-TCEs (on PDMS substrates) after support fiber removal, showing uniform *T* over a wide range of wavelengths. This is enabled not only by the low coverage of CuEW, but also by the large size of the openings in the network (tens of µm, Figure 2a) relative to the wavelengths of visible light (hundreds of nm).[17] This flat transmittance spectrum is advantageous in many devices, including solar cells and photocatalytic water-splitting devices.

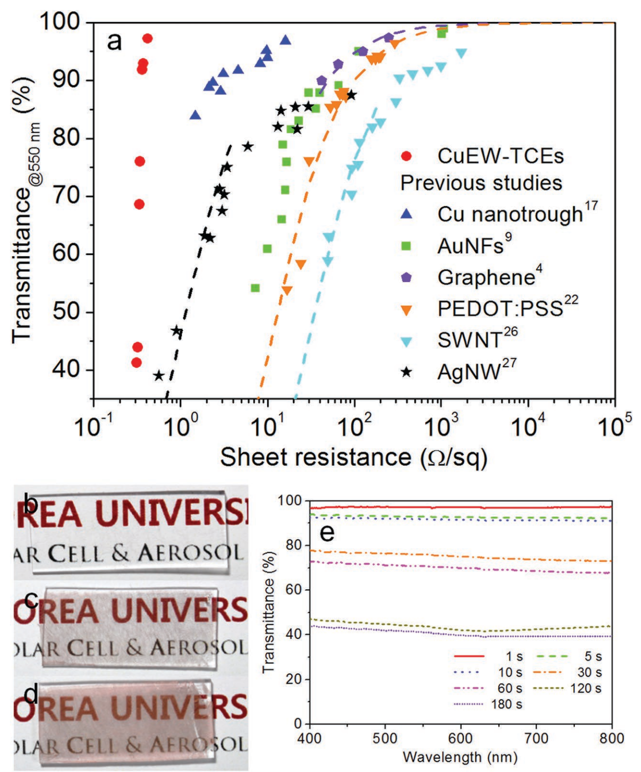


**Figure 3.** Optical and electrical performance. a) Transmittance versus sheet resistance for the CuEW-TCEs in comparison to prior studies. Images of CuEW-TCEs on PDMS with first electrospinning time: b) $t_{1st} = 1$ s, c) $t_{1st} = 60$ s, and d) $t_{1st} = 180$ s. e) Transmittance spectra of CuEW-TCEs after removal of unplated support fibers.

We conducted bending and stretching tests on the CuEW-TCEs, as shown in **Figure 4** and Movies S2, S3, and S4 (Supporting Information). The CuEW-TCEs on PET with $t_{1st} = 5$ and 30 s were used, and sputtered ITO films on PET were prepared for comparison in bending tests. CuEW-TCEs maintained almost constant $R_s$ as the bending radius ($R_b$) decreased from 100 to 1 mm. In contrast, for the ITO film, $R_s$ increased dramatically at $R_b = 10$ mm and could not be measured for $R_b < 6.5$ mm (Figure 4a). Even after 1000 cycles of bending to $R_b = 5$ mm (Figure 4b) the CuEW-TCE retained its electrical conductivity, while $R_s$ increased after a few cycles for the ITO film. Figure 4c,d show that CuEW-TCEs on Eco-flex also have exceptional stretchability relative to previously reported TCEs.[9,17] Lee et al.[28] achieved impressive results for a stretchable, though not very transparent, nanowire electrode, and showed that use of long nanowires was essential for improving stretchability. The electrospinning approach used here takes the concept of using long wires to the maximum possible extent by employing a continuous fiber; the remarkable stretchability results originate from the highly interconnected structure of continuous electrospun nanofiber mats. The CuEW-TCEs samples were stretched up to 580% without a sharp rise in the value of $\Delta R/R_0$, which increases only to 3.6 for the sample with $t_{1st} = 30$ s. Even though some CuEWs are broken after stretching over 200%, the overall CuEW network maintains high conductivity through the many unbroken CuEWs even when stretched by 580% (Figure S8, Supporting Information). As shown in Movie S2 (Supporting Information), electrical performance of CuEW-TCEs is retained under multi-directional stretching. The bulk resistance in the movie differs from the sheet resistance because the ohmmeter for bulk resistance reflects changes in the width and length of the sample as well as changes in sheet resistance.

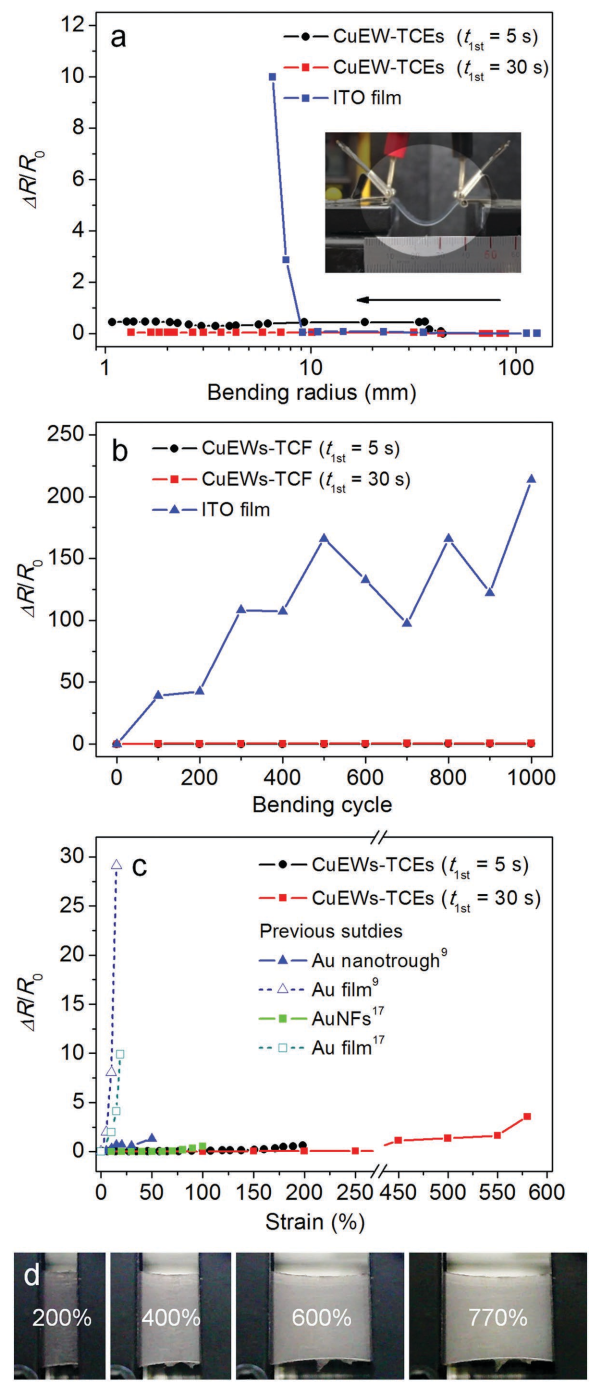


**Figure 4.** Mechanical stability. Changes in the sheet resistance as a function of: a) bending radius, b) bending cycle (CuEW-TCEs on PET), and c) uniaxial strain in tensile test of CuEW-TCEs on Eco-flex. d) Images of CuEW-TCEs on Eco-flex during stretching up to 770% (LED operation is shown in Movie S3, Supporting Information).



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A rigorous percolation model of the network of CuEWs on a surface is detailed in the Supporting Information. We can find the expected normalized conductivity C of the small six-point cell (Figure S10, Supporting Information) or any larger cluster arising from it using the renormalization technique of the percolation theory (Equation S4 and S5, Supporting Information) as

$$C=\frac{\bar{\sigma}}{\sigma_0}=\frac{1}{\sigma_0}\int_0^\infty \sigma P(\sigma)\mathrm{d}\sigma =\frac{11}{3}p^3(1-p)^2+p^2(1-p)^3+\frac{17}{5}p^4(1-p)+p^5 \quad (1)$$

A large percolating cluster, which has $p \to 1$, according to Equation (1) possesses conductivity $C \to p^5$, i.e., its conductivity tends to that of an individual copper nanowire. On the other hand, any large non-percolating cluster, which has inevitably $p \to 0$, possesses conductivity $C \to p^2$, which obviously tends to zero.

The predicted transmittance $T$ is found accordingly as

$$T=1-\frac{\bar{B}}{S}=1-5b\left[p(1-p)^4+4p^2(1-p)^3+6p^3(1-p)^2+4p^4(1-p)+p^5\right]=1-5bp \quad (2)$$

Key parameters in this model are $p$, the probability that a link bond in the percolative network is occupied ($0 < p < 1$), and the shading factor $b$, the fraction of a cell ($0 < b < 1/5$) in the percolative network that is shaded by a single wire (a link in the network). If the dependence of the transmittance on resistance in the form of $Y = \log(T^{-0.5}-1)$ versus $X = \log(1/C)$, as predicted by percolation theory, is superimposed at any constant shading factor $b$ on the experimental results of the present work, the predictions of the percolations theory disagree with the data. The agreement is, however, possible when one finds an appropriate dependence of $b(p)$, with $p$ being the probability of link occupancy, as shown in **Figure 5**. The $p$ values were obtained by substituting the dimensionless $R$ ($=1/C$) by $R_s/R_{single}$ in Equation (1). The experimentally measured sheet resistance, $R_s$ is in the 0.31 to 0.42 Ω $sq^{-1}$ range and the numerically calculated resistance of a single CuEW is roughly $R_{single} = \rho L/A \approx 0.300\ \Omega$, where $\rho$, $L$, and $A$ were $1.68 \times 10^{-8}\ \Omega$ m (resistivity of copper), 12.8 µm (the average length between 200 intersections of CuEWs in SEM images), and $7.15 \times 10^{-13}\ m^2$ (the cross-sectional area of the copper-plated shell of a wire, when the total diameter of CuEW is 1000 nm and the core polymer diameter of CuEW is 300 nm), respectively. Then, $b$ values were finally obtained by using Equation (2), with the $p$ values found using Equation (1), and the experimental values of $T$. All the values used and obtained are listed in **Table 1**. The $p$ values range from 0.9759 to 0.8183 and the $b$ values range from 0.1209 to 0.0073. The $b$ values increase as $t_{1st}$ increases, that is, the dimensionless factor $b$ in the percolation model responsible for the shade of an individual nanofiber appears to be dependent on the deposition time in the experiment due to the nanofiber conglutination revealed by Figure S8 (Supporting Information). This provides a basis for understanding the combined weak dependence of $R_s$ and strong dependence of $T$ upon the first electrospinning deposition time, which determines the total coverage of CuEWs in the TCE. Even at the shortest deposition time, $p$ is already quite high, implying that a dense percolative network of highly conductive CuEWs has already formed. Thus further deposition dramatically increases $b$, the fraction of a cell shaded by a single link in the network, because the network becomes denser. However, it does not dramatically increase $p$, simply because $p$ is already quite high.

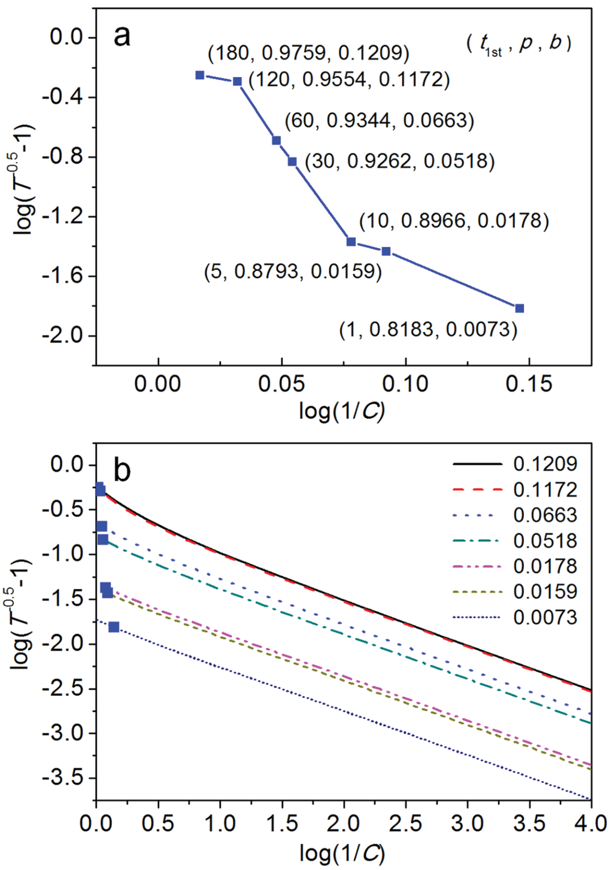


**Figure 5.** Percolation model of conductance and transmittance. Dependence of transmission on resistance: a) Experimental data and the corresponding values of $p$ and $b$ found from the percolation theory. b) Theoretical results based on Equation (1) and (2), with each line corresponding to a constant value of $b$; blue symbols correspond to the theoretical results from panel a).

In summary, we have presented a new approach to low-cost production of TCEs that can be used to generate films with unprecedented combinations of low $R_s$ and high $T$ while also providing exceptional mechanical flexibility and robustness. The key to the high performance of these CuEW-TCEs relative to other wire-based TCEs is the elimination of junction resistance at the intersection of wires, provided by the electroplating process used to deposit the copper wires. This drastic advance in performance, robustness, and cost of TCEs opens up a vast array of new opportunities in flexible displays, electronic skins, and other low-cost, large area optoelectronic devices. Oxidation of CuEW-TCEs can be prevented by encapsulating the transparent conducting film between other device layers, as done in most optoelectronic devices.

**Table 1.** Experimental data versus predictions of the percolation theory.

| Case | Experimental result | | Percolation model | | | |
|---|---|---|---|---|---|---|
| $t_{1st}$ [s] | $R_s$ [$\Omega$ sq$^{-1}$] | $T$ [%] | $R$ | $T$ | $p$ | $b$ |
| 180 | 0.312 | 41 | 1.04 | 41 | 0.9759 | 0.1209 |
| 120 | 0.323 | 44 | 1.08 | 44 | 0.9554 | 0.1172 |
| 60 | 0.335 | 69 | 1.12 | 69 | 0.9344 | 0.0663 |
| 30 | 0.340 | 76 | 1.13 | 76 | 0.9262 | 0.0518 |
| 10 | 0.359 | 92 | 1.20 | 92 | 0.8966 | 0.0178 |
| 5 | 0.371 | 93 | 1.24 | 93 | 0.8793 | 0.0159 |
| 1 | 0.420 | 97 | 1.40 | 97 | 0.8183 | 0.0073 |

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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