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Highly Stretchable or Transparent Conductor Fabrication by a Hierarchical Multiscale Hybrid Nanocomposite

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As is frequently seen in sci-fi movies, future electronics are expected to ultimately be in the form of wearable electronics. To realize wearable electronics, the electric components should be soft, flexible, and even stretchable to be human-friendly. An important step is presented toward realization of wearable electronics by developing a hierarchical multiscale hybrid nanocomposite for highly flexible, stretchable, or transparent conductors. The hybrid nanocomposite combines the enhanced mechanical compliance, electrical conductivity, and optical transparency of small CNTs (d ≈ 1.2 nm) and the enhanced electrical conductivity of relatively bigger Ag nanowire (d ≈ 150 nm) backbone to provide efficient multiscale electron transport path with Ag nanowire current backbone collector and local CNT percolation network. The highly elastic hybrid nanocomposite conductors and highly transparent flexible conductors can be mounted on any non-planar or soft surfaces to realize human-friendly electronics interface for future wearable electronics.

1. Introduction

Recently, wearable smart electronic devices such as information-providing smart glasses (Google Smart Glasses), wristwatch type bendable smart phones, flexible displays or electronic maps are gaining tremendous scientific and social interests. However, they are yet hard to call as true wearable electronics because they still contain a lot of rigid electronic parts. To realize the meaningful wearable electronics, the electronic components should be stretchable or at least flexible because human body is not rigid but soft and elastic. The first generation demonstration of the flexible electronics includes flexible opto-electronics such as flexible organic light-emitting diodes (OLEDs) displays,[1,2] flexible solar cells[3,4] and flexible touch panels.[5,6] These flexible or wearable opto-electronic devices need electrodes not only with high transparency and electrical conductivity but also with good mechanical compliance such as stretchability and flexibility because they are subject to various mechanical deformations including stretching, bending, twisting, and folding during operation, and also they should be compatible with human body which is soft, elastic and curved.[7] As alternatives to fragile and expensive ITO (indium tin oxide) transparent conductor, CNT,[8–12] graphene[5,13,14] and metallic nanowires[6,15–20] are getting huge attention. Carbon based nanomaterials commonly show good flexibility and stretchability but also suffer from relatively poor conductivity thus low transparency to achieve moderate conductivity and difficulty in scalable production with high material cost. Metallic NWs have received the attention recently as transparent electrodes substitutes especially in solar cell application. AgNWs are very conductive because silver has high electron density and AgNWs percolation network shows good mechanical flexibility.[15,21,22]

As Rogers *et al.* suggested, there are two major strategies to address the challenge in stretchable and flexible electronics, namely, "materials that stretch" and "structures that stretch" with good electrical conductivity. These two major strategies have well studied with thin metal ribbon[23–26] structures and soft substrates. However, these approaches have been required advanced complex fabrication process using multi-steps of high temperature vacuum deposition and corrosive chemical etching. One of promising approach in stretchable and flexible conductors is using carbon nanomaterials such as carbon nanotubes (CNTs) and graphene which can be solution processed.

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However, although they show excellent transparency, CNT still suffers from relatively large sheet resistance (>100 ohm/sq)[1] and graphene has large material cost and complex fabrication issue. For examples, inability to obtain bulk quantities of material with suitable purity is one of the major impediments towards implementing CNTs.[27] In case of graphene, multiple boundaries and incorporate defects dramatically decreased the conductivity in practice.[14,27,28] Moreover, it is still difficult to produce single-layer samples of graphene and large scale-pattern growth. In addition, even though conducting polymer has flexibility for various substrates, it still has low conductivity compared to other materials such as metal or metal oxide.[29,30]

Each material has its own strength and weakness, and the resultant conductor based on a single material inevitably brings the demerit of the material in use. Our aim is to combine two different materials in order to compensate the shortcomings of each material with the strength of the other. We try to overcome the current limitations in stretchable electronics by introducing hybrid nanocomposite material using "hierarchical structure that stretches" to compensate the disadvantage of single material while keeping the advantages of each material. The proposed hybrid material is a hierarchical multiscale percolation network of AgNWs/CNTs nanocomposite of very long silver nanowire with relatively larger dimension (d ~ 150 nm, L ~ 50–100 µm) and single wall CNT (SWCNT) with relatively smaller dimension (d ~ 1.2 nm, L ~ 2–10 µm). For the hybrid conductor of hierarchical multiscale AgNW/CNT electrode, highly conductive AgNW percolation network provide backbone current collector electrodes for fast electron transport, while elastic but relatively resistive SWCNT percolation network endows high stretchability and flexibility and further enhances the conductivity through providing local electron transport paths by filling the inter-nanowire space of the backbone electrode of AgNW mesh. For efficient electron transport, this multiscale hierarchical approach could demonstrate the novel highly stretchable conductor by combining the advantages of small diameter, highly stretchable but resistive CNT local percolation network and highly conductive but bigger AgNWs backbone mesh. Besides the stretchable conductor, the characteristics of multiscale hierarchical AgNW/CNT electrode have useful aspects for highly transparent conductor by providing multiscale electron path with AgNW current collector and local CNT percolation network.

In this research, we suggest hierarchical multiscale AgNW/CNT hybrid nanocomposite, which takes advantage of enhanced mechanical compliance and optical transparency of CNTs as well as enhanced electrical conductivity of AgNWs. As a proof of concept for AgNW/CNT hybrid multiscale hierarchical nanocomposite, we demonstrated two types of AgNW/CNT nanocomposite with different AgNW density and requirement (Figure S1): (a) highly stretchable (>460%) conductors with remarkable mechanical reliability over long repeated bending (>10,000 times), large twisting (>540°), and folding (~0°, complete folding) and (b) highly transparent and flexible conductor (T:80 ~ 93%). The fabricated highly stretchable, flexible and transparent conductors were applied to real devices such as stretchable light emitting diode (LED) circuits and touch panel displays.

2. Results and Discussions

Hierarchical multiscale nanocomposite of AgNW/CNT percolation network was prepared by a successive solution filtration of two different nanomaterials and subsequent transfer to the target substrate. Solution filtration of CNTs solution on 0.2 µm pore sized membrane filter was followed by AgNWs solution filtration on CNT deposited membrane filter. The AgNWs/CNTs layered nanocomposite formed on the membrane filter was transferred to target substrates by applying uniform suction on the other side of the membrane filter. In this process, mild suction was applied for both forming AgNWs/CNTs nanocomposite by solution filtration and transferring the nanocomposite to the target substrate, which minimizes the in-plane force which can cause damage on nanostructures. The membrane filter can be detached easily from the substrate, leaving AgNWs/CNTs percolation network nanocomposite on the substrate. Compared with other transfer method, this process is very simple and does not contain etching process for transfer. And the membrane filter can be used multiple times because the process only uses mechanical pressure and mechanical peel-off for transfer without heating or chemical process. The detailed information on the long Ag NW synthesis, CNT solution preparation and solution filtration process can be found in the Experimental Section and Supporting Information (Figure S2).

Figure 1 shows the schematic diagram and SEM picture of the proposed multiscale hierarchical AgNW/CNT hybrid nanocomposite electrode. The hybrid nanocomposite material is hierarchical multiscale percolation network with two different scales of very long AgNW backbone with relatively larger dimension (d ~ 150 nm, red colored) and single wall CNT branch with relatively smaller dimension (d ~ 1.2 nm, grey colored). The CNT mesh forms spider web like meshes in the inter-nanowire space of the AgNW backbone mesh. The multiscale hierarchical AgNW/CNT electrode provides highly effective percolation networks because small and elastic CNT mesh provides local paths for the electrons which will be further collected by

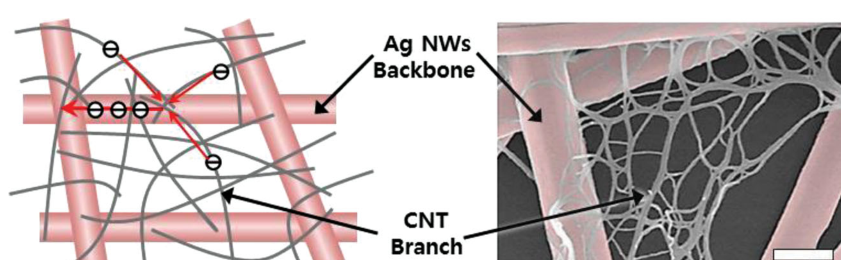

Figure 1. Schematic diagram (left) and SEM image (right) of hierarchical multiscale AgNW/CNT hybrid nanocomposite for highly stretchable conductors or highly transparent/flexible conductors. Note that for efficient electron transport, this multiscale hierarchical approach could demonstrate the novel highly stretchable conductor by combining the advantages of small diameter, highly stretchable but resistive CNT local percolation network (grey colored) and highly conductive but bigger AgNWs backbone mesh (pink colored). Inset scale bar is 300 nm.

the electron freeway of larger diameter AgNW backbone mesh. This hierarchical multiscale nanocomposite structures have various advantages which cannot be achieved by single component percolation network (only AgNW or only CNT cases). Firstly, multiscale hierarchical AgNW/CNT percolation network can make highly stretchable and highly conductive electrode because the hybrid electrode possesses the advantages of both highly stretchable CNTs and highly conductive AgNWs simultaneously. Secondly, it can make highly transparent conductor because CNT mesh between AgNW backbone mesh can further increase conductivity of the percolation network without deteriorating transparency. Therefore, hierarchical multiscale AgNW/CNT hybrid electrode could achieve high electrical conductance, high mechanical compliance, and high transparency simultaneously, which is almost impossible to achieve only with single component material.

When two or more materials make composite, it can form either layered composite structure or uniformly mixed structure. Uniformly mixed structure can be made by mixing materials together in the initial solution. However, AgNW/CNT uniform solution or film is difficult to form because CNTs interrupts the uniform distribution of AgNWs (supplementary Figure S3). Furthermore, CNT-CNT contact has relatively high contact resistance whereas AgNWs show good contact resistance by local welding after thermal annealing process. CNTs covering the AgNWs hindered the ohmic contact between AgNWs, which increases electrical resistance of the hybrid composite film. To overcome those problems, in this research, we fabricated layered structure of AgNW/CNT percolation network nanocomposite in which CNTs cover the AgNW percolation networks and assist AgNW electrode independently. This approach can make full use of high conductivity of AgNWs by filling the space in AgNWs backbone percolation network with CNTs without degrading ohmic contact between AgNWs. The deposition order of the solution filtration material is important for successful AgNW/CNT percolation network nanocomposite for stretchable and transparent electrodes. CNTs thin film was firstly formed on a membrane by CNT solution filtration and then AgNWs solution in ethanol is poured and vacuum-filtered to form AgNW percolation network on top of CNT percolation network film. CNT percolation network was not damaged by carefully poured AgNWs solution because of strong coupling among CNTs while the opposite deposit order was not successful. Depending on the AgNW/CNT nanocomposite electrode application, different concentration level of nanomaterials is applied.

But relatively short percolation network and relatively large diameter (~100 nm) of AgNWs limit the applicable scope for transparent electrodes. By combining the advantages of AgNW and CNT, highly flexible, transparent and conductive electrode could be made. Transparency can be controlled minutely by controlling the amount of CNTs. **Figure 2**A shows that CNT only ('NO AgNW' case in green box) or AgNW only ('NO CNT' case in orange box) cases show good transparency, however, still show very large or infinite resistance. Adding small amount CNT to AgNW networks dramatically reduce the sheet resistance from ∞ to 24 ~ 27 ohm/sq with little transparency degradation. CNTs only percolation network shows high transparency up to 98% easily but with poor conductivity while AgNWs only percolation network was difficult to reach 88% transmittance due to larger diameter. However, AgNW/CNT hybrid conductors in which AgNWs are covered with CNTs shows good conductivity with over 90% transmittance. This signifies that CNTs effectively reduce sheet resistance by assisting connection among isolated AgNWs to decrease discontinuities with small transmission loss, by providing various extra electrical paths over AgNWs window and by firmly joining the Ag NW mesh network especially at the Ag NW junctions to reduce the contact resistance between Ag NWs (further discussion in Supporting Information). CNTs connect the separated AgNWs which were not used as current path but only blocked penetration of light, as in Figure S4. As observed in Figure 2B, increasing CNT content decrease the resistance at the cost of transparency drop and the proper AgNW/CNT concentration should be chosen depending on the requirements. Optical transmittance and electrical conductivity are strongly correlated through the AgNW density. The optical transparency increases for the lower AgNW density, while the mechanical compliance and electrical conductivity increase for the higher AgNW density. There is minimum density of AgNW to achieve successful percolation networks as highly transparent and stretchable metal conductors. Adding CNT to AgNWs allows lowering the required AgNW density thus enhancing optical transmittance and mechanical compliance while maintaining high electrical conductivity. On the contrary, as the CNT concentration gets lower, the required minimum density of AgNW to form percolation network increases, and CNT only ('NO AgNW' case in green box) or AgNW only ('NO CNT' case in orange box) percolation network show insufficient performance to replace ITO, which usually requires sheet resistance lower than 100 ohm/sq at T = 85%. Figure 2C shows the fabricated multiscale hierarchical AgNW/CNT percolation network transparent conductors. Due to flexible nature of AgNW/CNT hybrid conductors, they can be easily applied for flexible or stretchable transparent conductor fabrication while fragile ceramic nature makes the application of ITO for flexible electronics difficult.[27]

Besides the highly transparent conductor, hierarchical multiscale AgNW/CNT hybrid nanocomposite can be applied to the highly stretchable and flexible conductor applications due to CNT's high strength under large strain.[31] AgNW percolation network is also good material for flexible application because the structure can effectively accommodate the elongation without deterioration. Compared with Ag thin film, AgNW has superior mechanical characteristic such as yield strength and elastic modulus,[32,33] which prevents the electrode from permanent deformation and subsequent mechanical failure. Flexibility can be enhanced by making composite with flexible conductor materials such as CNT mesh network. The AgNW electrode was covered with fine percolation network of CNTs and strongly connected CNTs makes the electrode tolerable to high mechanical strain. When strain is applied on the AgNWs electrode, maintaining connected junctions between AgNWs is the most important to hold down the rise of resistance because the total electrical properties are mainly dominated by the point contact resistance of AgNWs junction parts.[21] CNTs network encapsulates the AgNWs junctions beneath and protects the AgNWs junctions from separation.

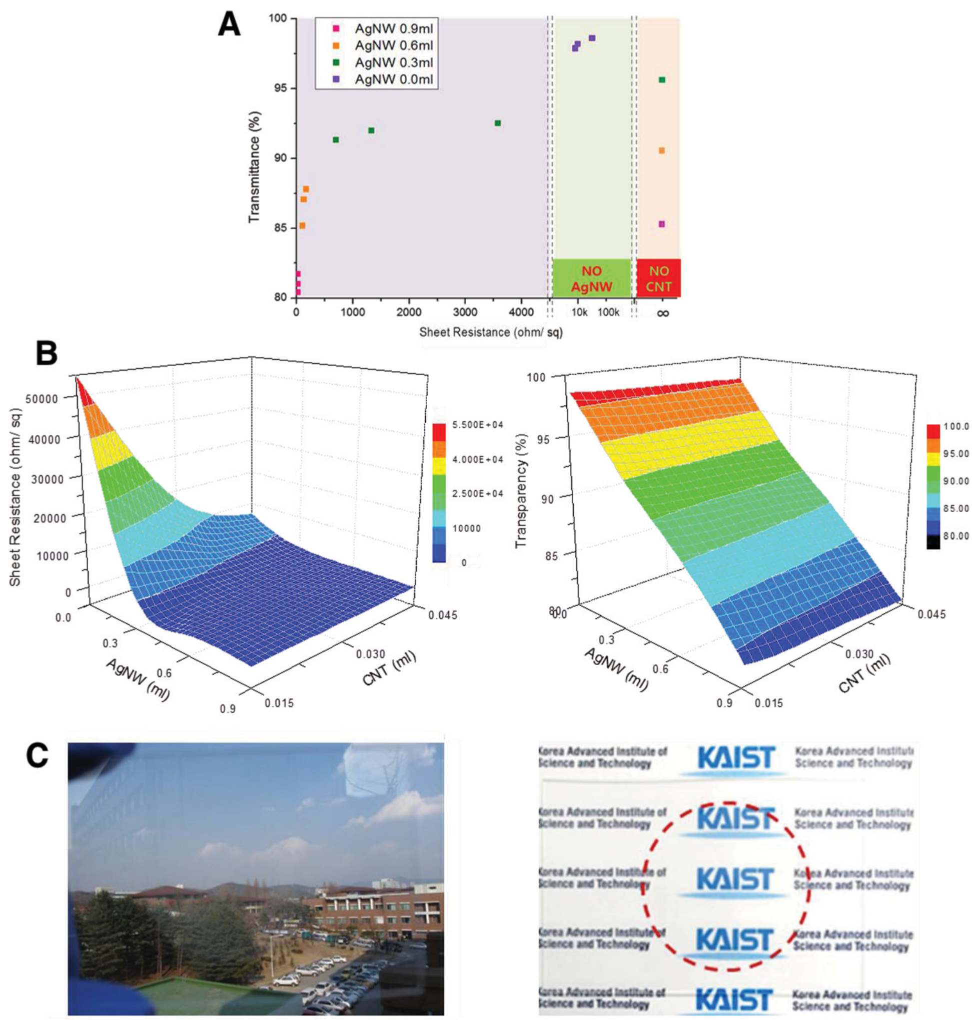

Figure 2. (A) Electrical sheet resistance vs. optical transmittance for various AgNW and CNT concentration. Note that CNT only ('NO AgNW' case in green box) and AgNW only ('NO CNT' case in orange box) cases show very large or infinite sheet resistance while addition CNT to the AgNW percolation network dramatically reduces the sheet resistance of the transparent conductor. (B) Surface plot of electrical sheet resistance (left) and transparency (right) for various AgNW and CNT concentration. (C) Transparent conductor fabricated by hierarchical multiscale AgNW/CNT hybrid nanocomposite. The concentration of AgNW and CNT solution was 0.3 mg/ml (0.03 wt%) and 1 mg/ml (0.3 wt%) respectively.

Highly stretchable conductor electrode is fabricated by transferring hierarchical multiscale AgNW/CNT hybrid nanocomposite on pre-strained (150%) Ecoflex substrate. **Figure 3**A shows the stretchability comparison of AgNW only electrode (black colored symbols) and hierarchical multiscale AgNW/CNT hybrid nanocomposite (red colored symbols). Note the superior stretchability of AgNW/CNT hybrid nanocomposite over AgNW only electrode because addition of CNT can provide enhanced mechanical strength (yield strength, elastic property and fatigue) to the AgNW percolation network. The AgNW only electrode shows much larger resistance increase under the same stress. Figure 3B shows demonstration of highly stretchable LED circuit (150% prestrain) with hierarchical multiscale AgNW/CNT hybrid nanocomposite. The LED was functional over 400% strain without any irreversible degradation. The microscopic behavior of the hierarchical multiscale AgNW/CNT hybrid nanocomposite under various stretching states can be observed in SEM images (Figure 3C). The AgNW/CNT nanocomposite under pre-strain initially shows corrugated surface. As the strain increases and stretching continues, the AgNW/CNT nanocomposite becomes unwrinkled and unraveled while maintaining the AgNW/CNT percolation network, which is advantageous for stretching application. This is because the pentagonal Ag NW is known to have superior yield strength and size dependent elastic modulus (higher Young's modulus than bulk Ag)[34] due to the enhanced stiffness due to internal twin boundary of the NWs,[35] therefore AgNWs easily accommodate tensile stress and deform. Secondly, CNT can hold the AgNW network and more effectively accommodate the deformation without any significant conductivity change by changing the network shapes than the AgNW only film which shows electrical failure under large strain.

Figure 4(A-C) show the superior mechanical compliance and electrical resistance reliability of AgNW/CNT hybrid nanocomposite ('AgNW + CNT' cases in Figure 4) over AgNW only ('AgNW only' cases in Figure 4) percolation network under

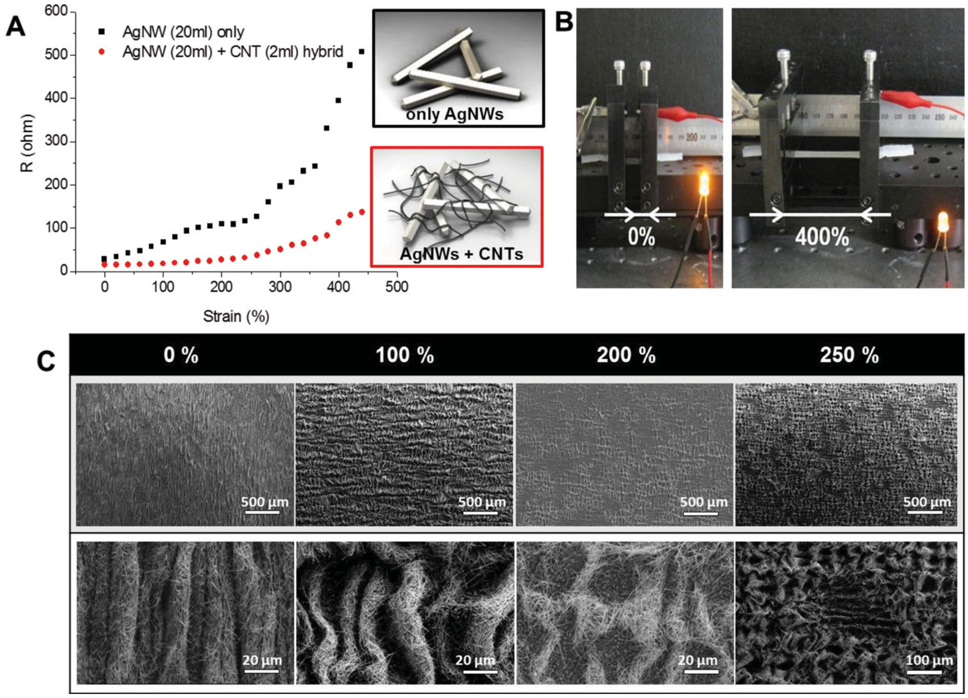

Figure 3. (A) Stretchability comparison of AgNW only percolation network (black colored symbols) and hierarchical multiscale AgNW/CNT hybrid nanocomposite (red colored symbols). Note the superior stretchability of AgNW/CNT hybrid nanocomposite over AgNW only percolation network because addition of CNT can provide enhanced mechanical strength to the AgNW percolation network. The AgNW only percolation network shows much larger resistance increase under the same stress. (B) Highly stretchable LED circuit demonstration with hierarchical multiscale AgNW/CNT hybrid nanocomposite. Note that the LED was functional over 400% strain. (C) Microscopic behavior of hierarchical multiscale AgNW/CNT hybrid nanocomposite by SEM imaging at various strain. Note that corrugated AgNWs/CNTs hybrid electrode becomes unwrinkled and unraveled region appears as stretching continues, which is advantageous for stretching application.

various mechanical stresses (bending, folding, and twisting). The hybrid nanocomposite electrode is highly stretchable not to lose their conductivity over 460%, which shows one of the best performance levels reported up to this day in stretchability aspect. This is similar level of stretchability demonstrated in our previous AgNW stretchable electrode[22] which was demonstrated through complex process and materials/sample preparation such as laser nano-welding on much higher prestrained (~300%) substrate with very long AgNWs (200 µm ~500 µm). However, by simply adding small amount of CNT to the AgNW mesh through the facile all solution process, current process achieved similar or even higher degree stretchability (more than 460% stretchability) without using complex laser nano-welding and under much smaller prestrain (150%). Therefore, current AgNW/CNT hierarchical nanocomposite dramatically simplified the complex fabrication process and stretchability of the AgNW based stretchable conductor. Figure 4A,B show that AgNW/CNT hybrid nanocomposite shows much smaller resistance increase under various bending radius (Figure 4A) and cyclic bending test (Figure 4B) over 10,000 cycles. According to the size of bending radius, the resistance of the AgNW electrode shows slightly changing values. But in case of AgNW/CNTs structure, it shows better conductivity and tolerance for bending. Over repeated bending cycles, AgNW electrode shows growing resistance from ~8,000 bending cycles while AgNW/CNTs structure maintains its electrical properties. At the beginning of cyclic bending test, the resistance of AgNW/CNT hybrid nanocomposite electrode was decreased to some degree in spite of bending. This phenomenon was observed when AgNW electrode was encapsulated on neutral axis using two PDMS layer of same thickness.[6] This may be because the initial substrate bending leaded to normal compression of AgNW as well as in-plane tension of AgNW. CNTs network on AgNW had a good adhesion on the substrate and bending could apply normal compression on unwelded AgNW junction parts besides in-plane tension. Due to the low temperature process nature, the AgNW/CNT hybrid nanocomposite can be fabricated on a paper. Figure 4C shows the repeated folding test. Electrodes are folded 6 times from 180° (flat state) to 0° (totally folded state). The resistance of AgNW only electrode (grey lines) rises abruptly by more than 3 orders of magnitudes as folding test progresses, while AgNW/CNT hybrid nanocomposite shows the obviously superior electrical performance stability over AgNW only sample. Figure 4D shows the superior mechanical compliance and electrical resistance reliability of AgNW/CNT hybrid nanocomposite under various twisting angles. The resistance value well recovered the initial value after the twisting angle is returned to 0°. It showed 8 times increase in electrical conductivity over 540°. The increase in resistance during twisting test may be caused by the simultaneous large stretching (~300% stretching for 540° twisting) due to the large stretchability of Ecoflex substrate and the axial prestrain.

AgNW/CNT hybrid nanocomposites show remarkable stretchable and transparent metal conductors with superior

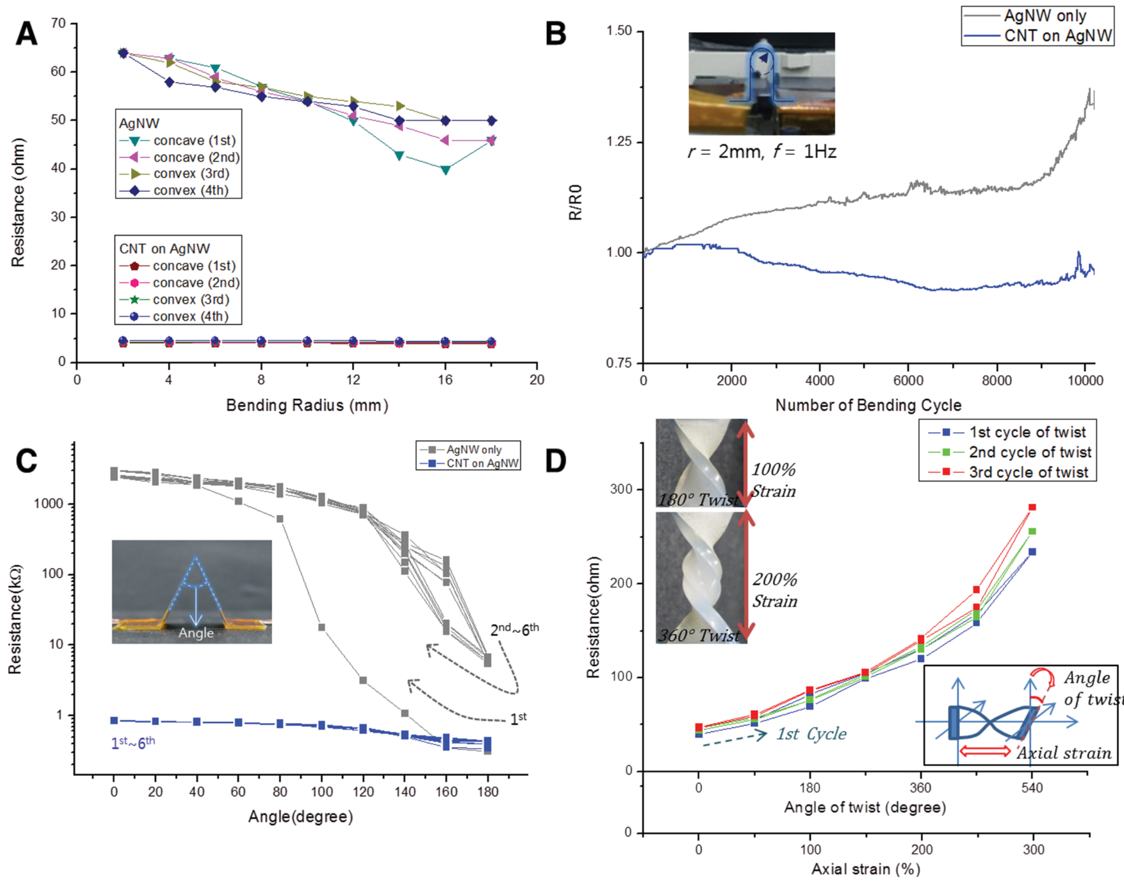

Figure 4. Various mechanical performance comparison of AgNW only percolation network and hierarchical multiscale AgNW/CNT hybrid nanocomposite. (A) Flexibility comparison of AgNW only percolation network (upper line group) and hierarchical multiscale AgNW/CNT hybrid nanocomposite (lower line group) for various bending radius. (B) Cyclic bending test comparison of AgNW only percolation network (pink colored) and hierarchical multiscale AgNW/CNT hybrid nanocomposite (blue colored). (C) Repeated folding test comparison of AgNW only percolation network (grey colored) and hierarchical multiscale AgNW/CNT hybrid nanocomposite (blue colored) for various folding angles. (D) Cyclic twist test of AgNW/CNT hierarchical multiscale nanocomposite for various twisting angles along with the corresponding axial strains information. Note the superior flexibility of AgNW/CNT hybrid nanocomposite because addition of CNT can provide enhanced mechanical strength to the AgNW percolation network.

maximum strain, durability, transparency, and electrical conductivity and they can be directly applied to high performance opto-electronics and stretchable, flexible electronics. **Figure 5** shows various applications of AgNWs/CNTs hybrid nanocomposite material for highly transparent conductor in a touch panel (Figure 5A), highly stretchable conductor in a stretchable LED circuit and its current-voltage measurement (Figure 5B), and highly twistable conductor in a twistable LED circuit (Figure 5C). Transparent touch panel (Figure 5A) is composed of ITO electrode on PET (polyethylene terephthalate) with elastomer spacer array and AgNW/CNT hybrid nanocomposite electrode on glass. AgNW/CNT nanocomposite on stretchable substrate is connected to LED and *I–V* relationship was measured during stretching as shown in Figure 5B. And the resistance change of hybrid electrode is also measured for 360° twisting angles (Figure 5C). Those demonstration signifies that the hierarchical multiscale AgNW/CNT hybrid nanocomposite electrode possess superior and robustness in optical transparency, electrical conductivity and mechanical compliance and can be readily applied to the stretchable and flexible electronics.

3. Conclusions

We developed hierarchical multiscale AgNW/CNT hybrid nanocomposite for highly stretchable conductors and highly transparent and flexible conductors by combining the enhanced mechanical compliance and optical transparency of small CNTs (d ~ 1.2 nm) and the enhanced electrical conductivity of relatively bigger AgNW (d ~ 150 nm) backbone. The AgNW/CNT hybrid nanocomposite can provide efficient multiscale electron transport path with AgNW current backbone collector and local CNT percolation network. The hierarchical multiscale AgNW/CNT hybrid nanocomposite electrode showed much superior and robust mechanical compliance, electrical conductivity and optical transparency over single component materials such as CNT only or AgNW only conductors. The AgNW/CNT hybrid multiscale hierarchical nanocomposite demonstrated highly stretchable (>460%) conductors with remarkable mechanical reliability over long repeated bending (>10 000 times), large twisting (>540°), and folding (~0°, complete folding) and highly transparent and flexible conductor (T:80–93%). The fabricated highly stretchable, flexible and transparent conductors were

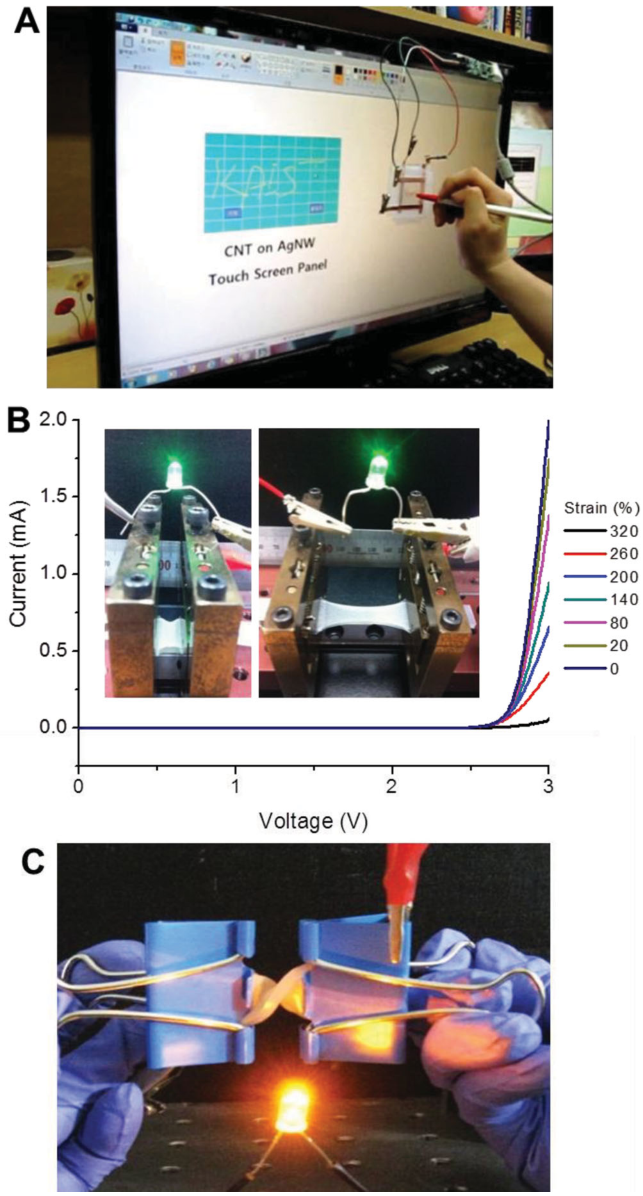

Figure 5. Applications of hierarchical multi-scale percolation networks of AgNW/CNT hybrid nanocomposite for (A) highly transparent conductor in a touch panel, (B) highly stretchable conductor in a stretchable LED circuit and its current-voltage measurement, (C) highly twistable (360°) conductor in a twistable LED circuit.

applied to real devices such as stretchable LED circuits and touch panel displays. We believe that this hierarchical nanocomposite technique can facilitate diverse approach to practical use of flexible and wearable electronics.

4. Experimental Section

Ag NW Synthesis: AgNWs are synthesized by a newly developed successive multistep growth.[6,36,37] 50 ml of ethylene glycol (EG, J. T. Baker) is preheated in an oil bath and maintains 151.5 °C temperature. 0.4 ml of 4 mM $CuCl_2$ (Sigma Aldrich) solution is added on this EG solution. After 10 min, 15 ml of 147 mM poly-(vinylpyrrolidone) (PVP, Sigma Aldrich) solution and 15 ml of 94 mM $AgNO_3$ are injected in turn. And AgNWs are synthesized for 1 hr at 151.5 °C. Grown AgNWs are rinsed with 1 time of acetone cleaning and 2 times of ethanol cleaning at a 10:1 v/v ratio to remove EG and PVP.

CNT Solution Preparation: High purity single walled carbon nanotubes (SWCNT with diameter of 1.0 ~ 1.2 nm, purity of 99%, Iljin Nanotech) produced by an arc-discharge method were dispersed in 1 wt% sodium dodecyl benzene sulfonate (SDBS) solution, sonicated, and then centrifuged.

AgNWs/CNTs Transparent Conductor Fabrication: CNTs dispersed in SDBS solution are vacuum-filtered on 0.2 μm porous membrane filter (D = 49 mm). And then AgNWs dispersed in ethanol solution is poured and vacuum-filtered on CNT percolation network on a membrane filter. After drying the remaining ethanol on the membrane filter at room temperature, a target substrate is gently put on the membrane filter covered with AgNWs/CNTs percolation network nanocomposite. By applying vacuum on the membrane filter side, uniform normal force can be applied on the substrate to induce transfer the target layer to the substrate. The uniform force applied by vacuum makes the substrate immovable on AgNWs/CNTs nanocomposite and transferred to the target substrate from the supporting membrane filter. The membrane filter can be removed by peeling off from the substrate to leave AgNWs/CNTs percolation network nanocomposite layer on the substrate. Thermal annealing (220 °C for 140 mins) is applied on the electrode to enhance the electrical conductivity.

AgNWs/CNTs Stretchable Electrode Fabrication: AgNWs/CNTs composite structure is fabricated with the same process used in the above in AgNWs/CNTs percolation network transparent conductor fabrication process. This nanocomposite material is transferred to the pre-strained (150%) highly elastic polymer (Ecoflex Supersoft 0030, Smooth-On) substrate.

Supporting Information

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

Acknowledgements

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[1] D. Zhang, K. Ryu, X. Liu, E. Polikarpov, J. Ly, M. E. Tompson, C. Zhou, *Nano Lett.* **2006**, *6*, 1880.
[2] X.-Y. Zeng, Q.-K. Zhang, R.-M. Yu, C.-Z. Lu, *Adv. Mater.* **2010**, *22*, 4484.
[3] J. van de Lagemaat, T. M. Barnes, G. Rumbles, S. E. Shaheen, T. J. Coutts, C. Weeks, I. Levitsky, J. Peltola, P. Glatkowski, *Appl. Phys. Lett.* **2006**, 88.
[4] F. Padinger, R. S. Rittberger, N. S. Sariciftci, *Adv. Funct. Mater.* **2003**, *13*, 85.
[5] S. Bae, H. Kim, Y. Lee, X. Xu, J.-S. Park, Y. Zheng, J. Balakrishnan, T. Lei, H. Ri Kim, Y. I. Song, Y.-J. Kim, K. S. Kim, B. Ozyilmaz, J.-H. Ahn, B. H. Hong, S. Iijima, *Nat. Nanotechnol.* **2010**, *5*, 574.
[6] J. Lee, P. Lee, H. Lee, D. Lee, S. S. Lee, S. H. Ko, *Nanoscale* **2012**, *4*, 6408.

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[7] J. A. Rogers, T. Someya, Y. Huang, *Science* **2010**, *327*, 1603.
[8] E. Artukovic, M. Kaempgen, D. S. Hecht, S. Roth, G. Grüner, *Nano Lett.* **2005**, *5*, 757.
[9] G. Gruner, *J. Mater. Chem.* **2006**, *16*, 3533.
[10] T. Sekitani, H. Nakajima, H. Maeda, T. Fukushima, T. Aida, K. Hata, T. Someya, *Nat. Mater.* **2009**, *8*, 494.
[11] Z. Wu, Z. Chen, X. Du, J. M. Logan, J. Sippel, M. Nikolou, K. Kamaras, J. R. Reynolds, D. B. Tanner, A. F. Hebard, A. G. Rinzler, *Science* **2004**, *305*, 1273.
[12] M. Zhang, S. Fang, A. A. Zakhidov, S. B. Lee, A. E. Aliev, C. D. Williams, K. R. Atkinson, R. H. Baughman, *Science* **2005**, *309*, 1215.
[13] G. Eda, G. Fanchini, M. Chhowalla, *Nat. Nanotechnol.* **2008**, *3*, 270.
[14] K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J.-H. Ahn, P. Kim, J.-Y. Choi, B. H. Hong, *Nature* **2009**, *457*, 706.
[15] S. De, T. M. Higgins, P. E. Lyons, E. M. Doherty, P. N. Nirmalraj, W. J. Blau, J. J. Boland, J. N. Coleman, *ACS Nano* **2009**, *3*, 1767.
[16] D. S. Ghosh, L. Martinez, S. Giurgola, P. Vergani, V. Pruneri, *Opt. Lett.* **2009**, *34*, 325.
[17] M. G. Kang, L. J. Guo, *Adv. Mater.* **2007**, *19*, 1391.
[18] M.-G. Kang, M.-S. Kim, J. Kim, L. J. Guo, *Adv. Mater.* **2008**, *20*, 4408.
[19] J. H. Lee, P. Lee, D. Lee, S. S. Lee, S. H. Ko, *Cryst. Growth Design* **2012**, *12*, 5598.
[20] J.-Y. Lee, S. T. Connor, Y. Cui, P. Peumans, *Nano Lett.* **2008**, *8*, 689.
[21] L. Hu, H. S. Kim, J.-Y. Lee, P. Peumans, Y. Cui, *ACS Nano* **2010**, *4*, 2955.
[22] P. Lee, J. Lee, H. Lee, J. Yeo, S. Hong, K. H. Nam, D. Lee, S. S. Lee, S. H. Ko, *Adv. Mater.* **2012**, *24*, 3326.
[23] D. Y. Khang, J. Jiang, Y. Huang, J. A. Rogers, *Science* **2006**, *311*, 208.
[24] D.-Y. Khang, J. A. Rogers, H. H. Lee, *Adv. Funct. Mater.* **2009**, *19*, 1526.
[25] J. Lee, J. Wu, M. Shi, J. Yoon, S. I. Park, M. Li, Z. Liu, Y. Huang, J. A. Rogers, *Adv. Mater.* **2011**, *23*, 986.
[26] Y. Sun, W. M. Choi, H. Jiang, Y. Y. Huang, J. A. Rogers, *Nat. Nanotechnol.* **2006**, *1*, 201.
[27] D. S. Hecht, L. Hu, G. Irvin, *Adv. Mater.* **2011**, *23*, 1482.
[28] V. C. Tung, M. J. Allen, Y. Yang, R. B. Kaner, *Nat. Nanotechnol.* **2009**, *4*, 25.
[29] T. Stöcker, A. Köhler, R. Moos, *J. Polym. Sci. B: Polym. Phys.* **2012**, *50*, 976.
[30] M. Reyes-Reyes, I. Cruz-Cruz, R. n. López-Sandoval, *J. Phys. Chem. C* **2010**, *114*, 20220.
[31] L. Hu, W. Yuan, P. Brochu, G. Gruner, Q. Pei, *Appl. Phys. Lett.* **2009**, 94.
[32] R. Gunawidjaja, H. Ko, C. Jiang, V. V. Tsukruk, *Chem. Mater.* **2007**, *19*, 2007.
[33] J. H. Yoo, S. I. Oh, M. S. Jeong, *J. Appl. Phys.* **2010**, 107.
[34] M. T. McDowell, A. M. Leach, K. Gall, *Nano Lett.* **2008**, *8*, 3613.
[35] T. Filleter, S. Ryu, K. Kang, J. Yin, R. A. Bernal, K. Sohn, S. Li, J. Huang, W. Cai, H. D. Espinosa, *Small* **2012**, *8*, 2986.
[36] K. E. Korte, S. E. Skrabalak, Y. Xia, *J. Mater. Chem.* **2008**, *18*, 437.
[37] B. Wiley, Y. Sun, Y. Xia, *Langmuir* **2005**, *21*, 8077.