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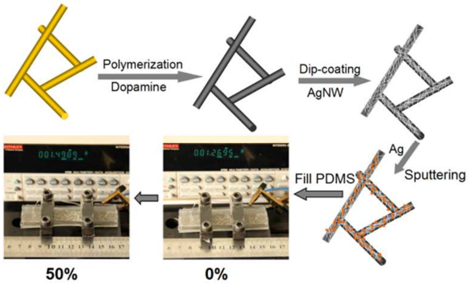
## Graphic abstract:

**A conductive ternary network structure composed of AgNWs/AgNPs based on polydopamine-modified 3D PU sponge** was fabricated, which shows excellent electromechanical ability owing to the combined effects from the ternary network.



# Conductive Ternary Network of Highly Stretchable AgNWs/AgNPs Conductors Based on Polydopamine-Modified Polyurethane Sponge**

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**Abstract:** A highly stretchable silver nanowires/silver nanoparticles (AgNWs/AgNPs) conductor based on polydopamine (PDA) modified three dimensional polyurethane (PU) sponge was fabricated. PU sponge as a skeleton was first modified by PDA to improve adhesion to the conductive nanomaterials. The conductive micro-network was fabricated by dip-coating of the PDA modified PU sponge into the solution of AgNWs, and AgNPs were further sputtered onto its surface to improve conductivity, resulting a conductive ternary network structure. The resistances of PDA-PU-AgNW-Ag-PDMS stretchable conductor decreased obviously with bending or stretching cycles firstly, and keep stable even up to 1000th cycles. The composites exhibit high electrical conductivity (exceeding 27.78 S $cm^{-1}$) and excellent electromechanical stability under high tensile strain (50%) owing to the combined effects from the ternary network. The significant electromechanical properties was also demonstrated by a simple stretchable circuit composed of a light-emitting diode (LED) and stretchable conductor during bending, twisting or stretching.

# 1. Introduction

Flexible and stretchable conductors that have the capacity to accommodate large levels of strain without significant degradation in their electronic performance can enable a wide variety of applications such as wearable displays, deformable antenna and capacitors, stretchable solar cells, electronic skins, camera eyes, and biological actuators.[1-4] As we known, conventional conductive materials such as metals show high electrical performance and stability, while sustain a small strain value.[5] In order to retain its both high conductivity and large deformation stability, different conductive and stretchable structure in elastic polymer have been designed for the fabrication of conductive elastic composites, which include i) the fabrication of buckled conductive interconnects or high loading conductive nanoparticles; ii) the formation of two-dimensional (2D) conductive networks, and iii) the configuration of three-dimensional (3D) conductive architectures.[6-8] The conductivity under substantial stretching and bending can both maintain to a certain extent through shape deformation of designed conductive structures in composites.

As we known, conductive and stretchable structures in polymer were core in the fabrication of conductive elastic composites. Although several conductive and stretchable structures have been explored, each one has its own unique properties arising from its specific network. In order to further enhance the stretchable conductivity, multiple network combining different structures was a promising way. [9] Ko et.al recently reported a highly stretchable and conductive metal conductor which was prepared by formation of wavy and 2D silver nanowires (AgNWs) network on a highly stretchable polymer substrate. [10] The binary structure network here by combining “buckled” and “2D” conductive strategies was responsible for the highly conductive and stretchable ability. In the meantime, a binary structure network was reported by Yu et.al through combining 2D and 3D networks. [11] However, to the best of our knowledge, only a few investigations have reported the design of conductive ternary network structures. It is interesting to enhance the properties of stretchable conductor through the combination of three different networks in elastic polymer.

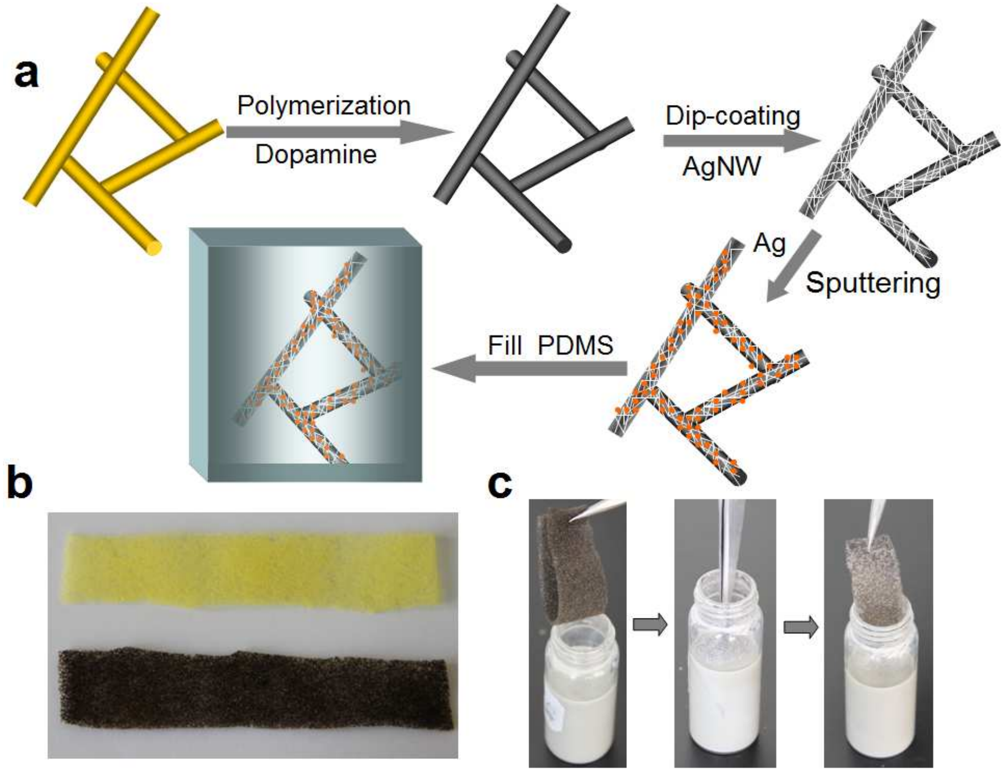
During those different conductive and stretchable structures, using stretchable sponge/foam as skeleton to construct 3D conductive architectures have been widely investigated because the conductivity of those composites during bending or stretching will keep stable by shape deformation before breaking the conductive 3D networks. [12-14] For example, Zheng et. al reported continuous 3D conductive metal networks, which was fabricated through solution deposition of metal thin films (Cu, Ag, Au) onto chemically functionalized porous polyurethane (PU) sponges. [6] Lee et al. have prepared another stretchable conductive composite film by using an e-beam evaporation process to deposit Ti and Au onto the skeleton of porous poly(dimethylsiloxane) (PDMS). [15] Moreover, as one of the most important conductive materials, AgNWs have been widely investigated in much progress for formation of 2D network to realize its electromechanical properties. [16-18] Here, it is promising to design the stretchable conductor structures through combining 3D modified polymer sponge with 2D AgNWs film to improve its performance. Moreover, as reported in literatures, nanoparticles in a polymer matrix has greater freedom for reversible nanoscale restructuring than nanotubes or nanowires hence represent a more dynamic system, which is essential for stretch ability. [19] Thus, the combination of Ag nanoparticles (AgNPs), 2D AgNWs network with 3D polymer skeleton is promising to construct stretchable conductors with unique properties for various applications.

During the design of conductive ternary network, one important part is to fabricate high adhesion between the conductive nanomaterials and polymer sponge to construct the 3D conductive network. [6, 20] But the adhesion of most commercial polymer sponges with conductive nanomaterials are not satisfactory, which needs multiple dip-coating process or P(METAC-*co*-MPTS) copolymer as adhesion layer. [6, 11, 21] As we known, polydopamine (PDA) coating is an amazing method to modify various surfaces arising from its abundant catechol and amine groups, where the lone pair electrons of the functional groups afford a strong binding interaction with the subsequent inorganic or organic layers.[22] Moreover, dopamine can easily self-polymerized at room temperature.[23, 24] Although various PDA modified materials such as graphene, carbon nanotube and PDMS film were reported, [25, 26] 3D polymer architecture which is difficult to modify by most common method such as oxygen plasma irradiation wasn't investigated at present. [20] In comparison with P(METAC-*co*-MPTS) copolymer as adhesion layers produced by multistep for electroless deposition of metal over porous 3D PU sponge, [6] the present procedure here resulted polydopamine as adhesion layer is more straightforward and without the use of any initiator, organic solvent, or high temperature.

In this paper, a conductive ternary network structure was fabricated, where the adhesion of PU microfiber was enhanced by PDA coating. Reduced-resistance was developed by sputtering a thin film of AgNPs onto PDA-PU-AgNWs surface, resulted a ternary structures containing two conducting networks. The PDA-PU-AgNW-AgNP-PDMS stretchable conductor showed a decreased resistance upon stretching or bending firstly, and then kept unchanged, which exhibited markedly high stretchability and small resistance increase. As an example of applications, a simple stretchable circuit composed of a light-emitting diode (LED) and stretchable conductor is fabricated to demonstrate its significant electrical properties during bending, twisting or stretching.

# 2. Results and Discussion

## 2.1 Fabrication of stretchable conductor



**Scheme 1. (a)**Schematic illustration of the fabrication of PDA-PU-AgNW-Ag-PDMS stretchable conductor; (b) Pictures of PU spong before (yellow) and after (black) PDA-modification; (c) Dip coating process of AgNWs absorbing on the surface of PDA-PU sponge.

A schematic representation of the fabrication process of the PDA-PU-AgNW-Ag-PDMS stretchable conductor is shown in Scheme 1a. Yellow, precleaned PU sponge was immersed into a solution of dopamine, where the self-polymerization of dopamine occurs on the surface of PU to form the adhesion layer, resulted the color change from yellow to black (Scheme 1b). [23-26] After drying step, black polydopamine-modified PU sponge with 3D-interconnected microfiber networks was dipped into isopropyl alcohol solution of AgNWs, where the microfibers were uniformly wrapped by the AgNWs after evaporating the isopropyl alcohol arising from increased adhesion by polydopamine-modification (Scheme 1c) . In order to further increase conductivity, a thin layer of Ag nanoparticles was coated the PDA-PU-AgNW substrate by magnetron sputtering. Finally, PDMS was introduced into the 3D PDA-PU-AgNW-Ag conductive sponge to form the desired stretchable conductor.

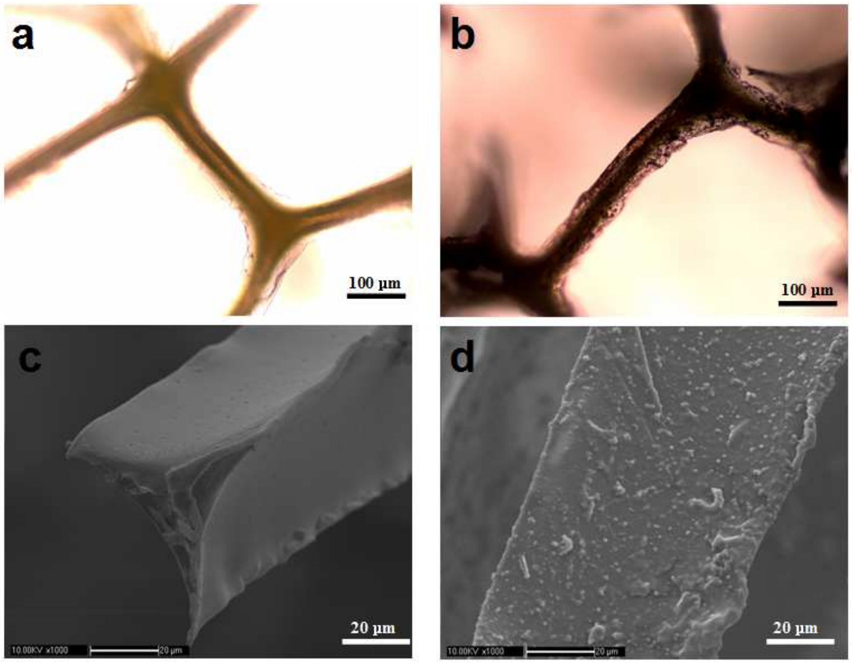


**Figure 1.** Optical microscope and SEM images of PU (a, c) and PDA-PU (b, d).

To construct PU sponge networks as a 3D skeleton to support the 2D AgNW, the key is to enhance the adhesion between PU and nanowire. As we known, polydopamine-coating is an amazing method to modify various surfaces arising from its abundant catechol and amine groups. And here it was expected to provide additional adhesion between the macroporous architecture and the subsequent AgNWs. As shown in Figure 1, the dramatic color change is observed in PU sponge modified with and without PDA. The pure PU sponge was yellow (Figure 1a), whereas the PDA-PU sponge was black (Figure 1b). [24, 25] A thin, adherent PDA layer on the surface of PU sponge surface after polymerization of dopamine are clearly recognized in the corresponding optical microscope images (Figure 1a, b). The SEM images in Figure 1c and d also reveal that PDA aggregations were coated uniformly on smooth PU surfaces. And the resulting PDA modified microfibers were uniformly wrapped by the thick layer of AgNWs, resulting in a conductive sponge with binary AgNW nano/micro network structures (Figure 2c, d). Without using PDA coating, only a thin layer of AgNWs with a large number of defects and obvious uncovered area on PU microfiber could be observed (Figure 2a, b ), indicating that the presence of the functional polymer layer is vital for enhancing the adhesion between AgNWs and PU surfaces. That's because the lone pair electrons of the functional groups (catechol and amine groups) on the surface of polydopamine layer cause a strong binding interaction with the subsequent AgNWs. [25, 26] Importantly, in comparison with similar method proposed by multistep using of the P(METAC-*co* -MPTS) copolymer modified PU sponge, [6] dopamine as a constituent molecule mimicking adhesive proteins was easily self-polymerized in the buffer solution at a weak alkaline pH without the use of any initiator, organic solvent, or high temperature.

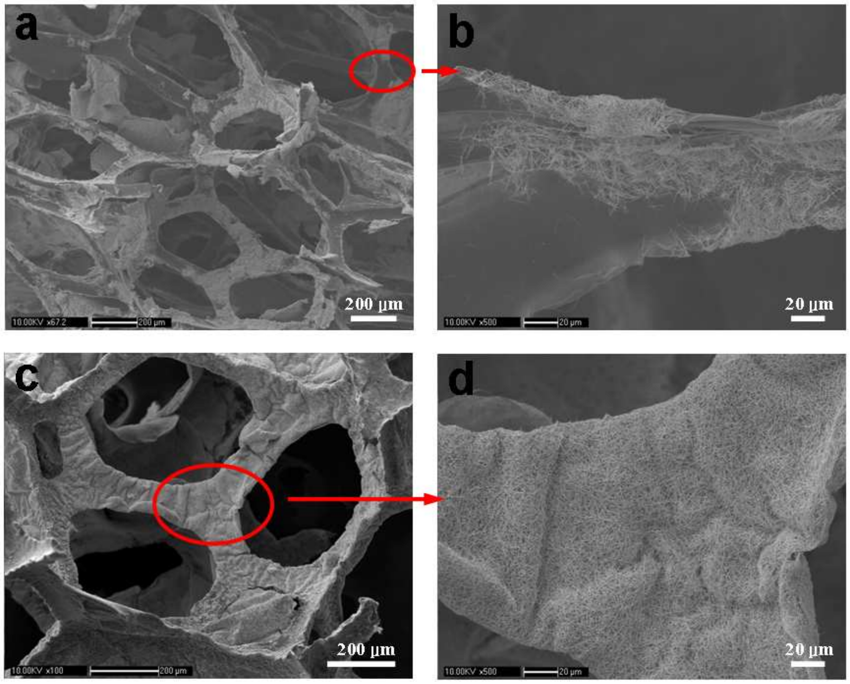


**Figure 2.** SEM images of PU-AgNW (a, b) and PDA-PU-AgNW (c, d) at low and high magnifications, respectively.

The morphology of AgNW coatings on PU microfiber was changed from sparse to dense by modification of PDA, indicating an increased conductivity, which was verified by the decreased resistance after the same fabrication procedure. The PU specimens of the same size (5.0 × 1.5 × 0.1 cm) were all coated with AgNWs in the beginning, and then infiltrated by PDMS. The resistance of PU-AgNW-PDMS was 8.51 Ω, while PDA modified one was 6.56 Ω, suggesting that PDA layer coated on PU surface plays a critical role in the fabrication and final properties of the conductive composite. The result was also confirmed by surrounding optical microscope images of PDMS composites, where AgNW on the PDA modified microfiber was more dense than unmodified one (Figure S2). Compared to the recently reported research of using P(METAC-*co*-MPTS) copolymer as adhesion layer for electroless deposition of metal over porous 3D PU sponges, [6] the reason that makes this route unique is obvious decrease of resistance after several bend or stretch process. For example, the resistance of PU-AgNW-PDMS conductor was reduced from 6.51 Ω to 4.50 Ω after several bend cycles. More details will be discussed in later sections.

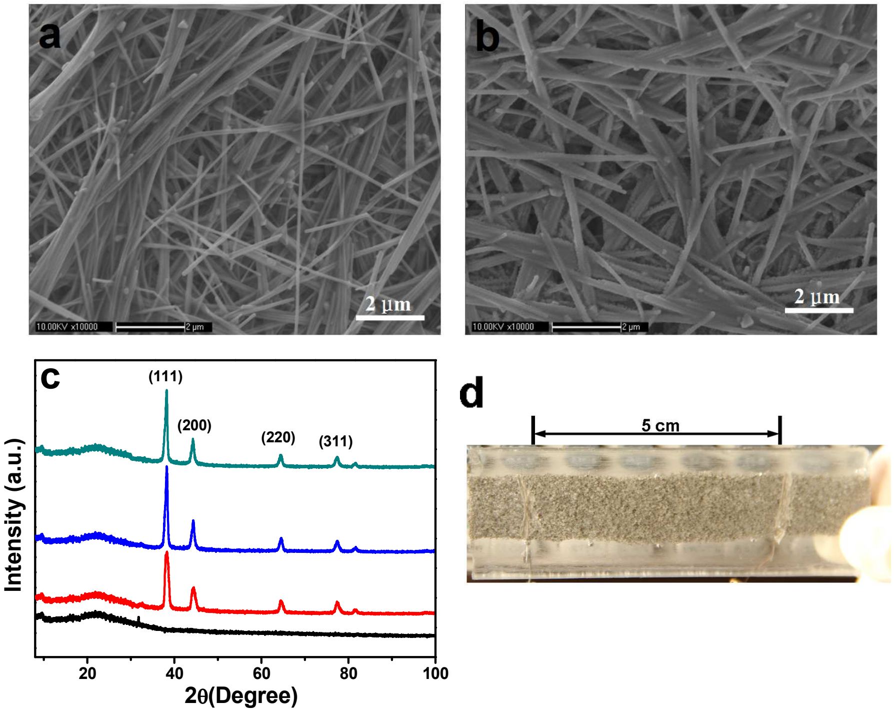


**Figure 3.** SEM images of PDA-PU-AgNW (a) before and (b) after AgNPs film was sputtered. (c) XRD spectra of PU (black), PU-AgNW (red), PDA-PU-AgNW (blue), PDA-PU-AgNW-Ag (green). (d) Optical images of PDA-PU-AgNW-Ag-PDMS stretchable conductor. The size of PU sponge embedded in PDMS between two copper wires is 5.0×1.5×0.1 cm.

In order to further improve conductivity, deposition of AgNPs film on the PDA-PU-AgNW substrate was explored through magnetron sputtering. As shown in Figure 3, AgNWs coated on the surface of the PDA-PU sponge become remarkable thicker upon sputtering, due to deposition of Ag nanoparticles. The decrease of resistance of PDA-PU-AgNW-PDMS was significant, from 6.5 Ω to 1.7 Ω after sputtering. The conductivity was around 19.06 S $cm^{-1}$ and increased to 27.78 S $cm^{-1}$ after 1000 stretching cycles from 0% to 50% strain, which is higher than that of PUS-AgNW-PDMS after eight dip-coating cycles and the procedure here is much easier. [11] **Figure 3** c shows the XRD patterns of PU, PU-AgNW, PDA-PU-AgNW and PDA-PU-AgNW-Ag. A broad peak centered at $2\theta = 21.0°$ were ascribed to the diffraction peaks of pure PU. And the typical diffraction peaks at 38.10, 44.20, 64.34, and 77.39° that represents Bragg reflections from the (111), (200), (220), (311) planes of Ag were observed, [27, 28] which indicates the existence of Ag in the stretchable conductor respectively. In addition, as shown in Figure 3d, PDMS was transparent and the modified PU sponge embedded in PDMS can be seen clearly. It should be noted that the distance of two copper wires embedded into the two side of the conductor with silver paste was 5 cm (Figure 3d), and the size of the PU sponge for all measured samples is uniform, whether modified or not, to ensure the best comparative value.

## 2.2 Bendability and stretchability of PDA-PU-AgNW-Ag-PDMS conductors

The specially designed PDA-PU-AgNW-Ag-PDMS architecture combines the advantages of both flexible 3D interconnected junction-free PU network at the microscale as polymer skeletons and conductive AgNWs/AgNPs network at the nanoscale, which imparts the composite with both excellent electrical and mechanical properties. The resistance variation of PDA-PU-AgNW-Ag-PDMS conductors under bending deformation was investigated. For bending tests, PDA-PU-AgNW-Ag-PDMS composite was bent to different radius of curvatures until the samples were folded. As shown in Figure 4a, during the first bending cycle, the resistance of PDA-PU-AgNW-Ag-PDMS composite had a slight decrease from 1.71 Ω to 1.65 Ω until bending to fold. The stability of resistance here during the bending process is due to the shape deformation of the PU skeleton at the microscale and the resulting AgNWs micronetwork which guarantees its

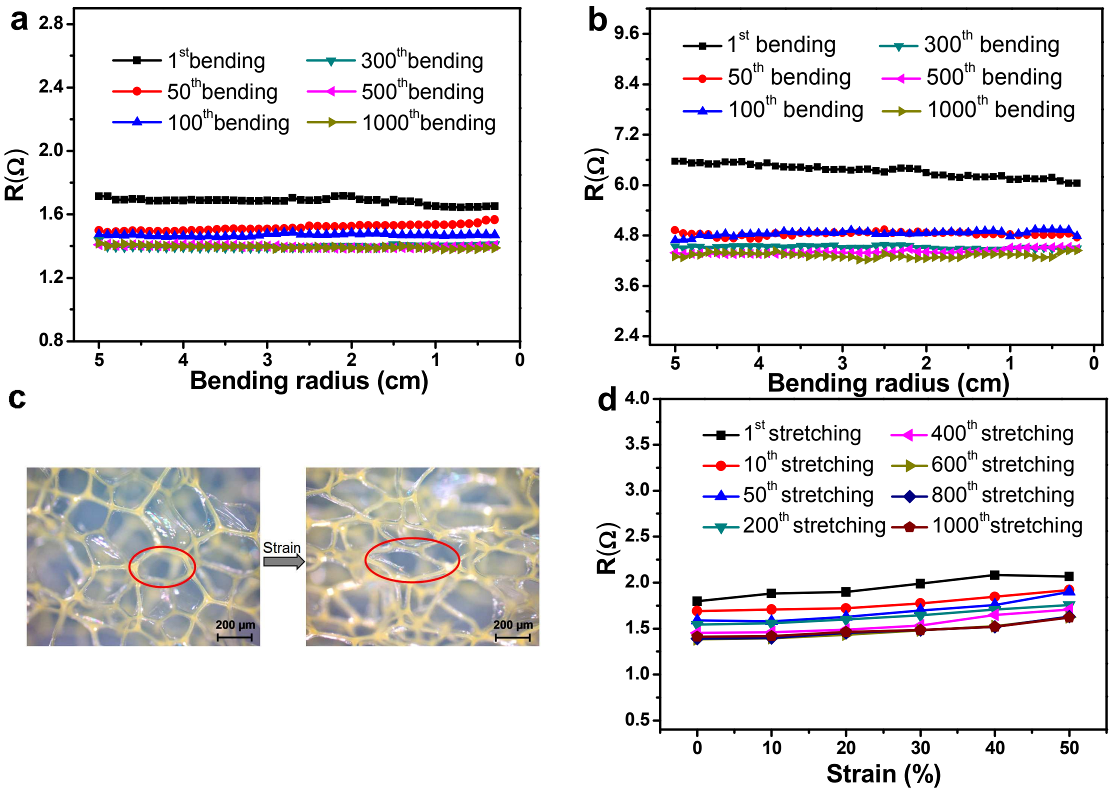


**Figure 4.** Electrical resistance of (a) PDA-PU-AgNW-Ag-PDMS and (b) PDA-PU-AgNW-PDMS stretchable conductor as a function of bend radius at different bend cycles; The size of PU sponges embedded in PDMS between two copper wires for all measured samples is 5.0×1.5×0.1 cm. (c) Optical microscope images of PU sponge micro-network describe the effects of elongation. (d) Resistance response of PDA-PU-AgNW-Ag-PDMS conductor as a function of strain at different stretching cycles. The distance of two copper wires embedded into the two side of conductor with silver paste was 5 cm, where 2 cm of conductor in length were conducted in stretching tests.

electrical conductivity. [11] Compared with the results mentioned above, increasing bending cycles obviously decreased the resistance firstly, and, afterwards, keep stable at 1.39 Ω when beyond 300th bending cycles, even up to 1000th cycles (Figure 4a). Interestingly, the similar decreased trend was also observed for PU-AgNW-Ag-PDMS composite during the bending cycles as shown in Figure 4b. The resistance variation behaviors as a function of bending cycles were clearly shown in Figure S4. Compared to the stable resistance of PDA-PU-AgNW-Ag-PDMS composite after 300th bending cycles, the resistance of PU-AgNW-Ag-PDMS composite still decreased slowly after high bending cycles which may be attributed to the absence of AgNPs. The stable resistance after higher bending cycles revealed the excellent electromechanical stability of PDA-PU-AgNW-Ag-PDMS composites and their great potential for high-performance stretchable and flexible conductors.

The reason of unchanged resistance under deformation is due to the PU miro-network as a polymer skeleton. As shown in Figure 4c, when the tensile strain on PDA-PU-AgNW-Ag-PDMS composites increased, the macropores of PU sponge (marked by the red window) shrank perpendicular to the direction of the tensile force and elongated in the direction of the applied force, resulting share the tensile strain applied on the composite conductive pathway. [11, 16] Therefore, the 3D architectures embed in composite remarkable enhanced its flexible conductivity than just only 2D nanowire networks. In addition, the AgNWs coated on the modified PU microfibers was connected together to form percolation network which can effectively accommodate the deformation without any significant change in conductivity by alter the shape of network. [10, 29] Moreover, even with defect section existing, Ag nanoparticles with greater matrix mobility would overcome this negative effect. In one word, the conductive ternary network structure containing 3D polymer architectures, 2D AgNWs percolation network and AgNPs formed in PU-AgNW-Ag-PDMS composite would greatly enhance their performances as stretchable conductors as confirmed by stretching test.

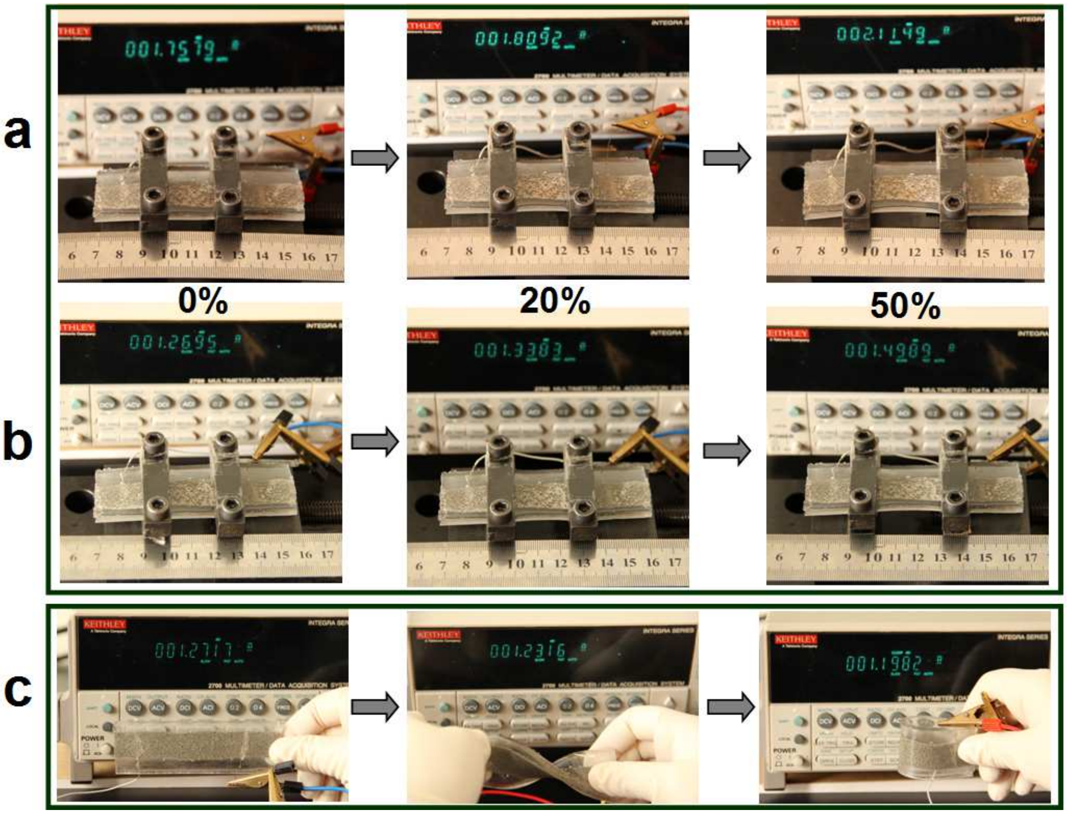


**Figure 5.** Resistance of PDA-PU-AgNW-Ag-PDMS conductor at 0%, 20%, 50% strain during the 1st (a) and 1000th stretching (b). (c) Resistance of PDA-PU-AgNW-Ag- PDMS conductor under twisting and folding after 1000th stretching. The size of PU sponges embeded in PDMS between two copper wires for all measured samples is 5.0×1.5×0.1 cm, where 2 cm of conductor in length were conducted in stretching tests.

The resistance variations of stretchable conductors were studied as a function of tensile strain. The stretching tests were carried out from 0% to 50% strains for 1000 cycles each. As revealed in Figure 4d, during the first stretching, the resistance of PDA-PU-AgNW-Ag-PDMS composite increase from 1.75 Ω to 2.10 Ω at the 50% strain, which is much better than for the AgNW/PDMS stretchable conductors (ca. from 0.82 Ω to 2.6 Ω at 50% strain during first stretching. [29] Digital pictures in Figure 5a shows resistance of stretchable conductor at specific strains of 0%, 20%, and 50% measured with a four-probe method, which clearly exhibits the stretching process as well.

From the viewpoint of repeatability, as the stretching cycle number increased, the resistance of stretchable conductor by different stretching cycles was decreased obviously as shown in Figure 4d. After 400 times stretching, the resistance variation of the PDA-PUS-AgNW-Ag-PDMS composites with increasing cycles gradually became stable under stretching deformation. And the resistance variation curve at the 600th, 800th, and 1000th cycle was completely overlapped, demonstrating the resistance stability of composites which can be used as ideal interconnects in flexible electronic applications. The excellent stability of resistance was also confirmed by comparing with the previous reported PU-CuAg-PDMS conductors where the resistance increased by 20% at the first cycle and gradually increased to 2.2-fold after 1000 cycles under 40% strain. [6] Under the 1000th stretching cycle, the resistant of stretchable conductor at 0%, 20%, and 50% strain was reduced to 1.26 Ω, 1.33 Ω, and 1.49 Ω respectively. As revealed in Figure 5a, b, compared with the resistance during the 1st and 1000th stretching cycles, a decrease of 0.50 Ω, 0.47 Ω, and 0.62 Ω of the PDA-PU-AgNW-Ag-PDMS composites were observed at 0%, 20%, and 50% strain respectively. The increase of resistance is about 0.35 Ω under 50% strain for the 1st stretching cycles, while an increase of 0.23 Ω was observed for the 1000th stretching cycles, which is much better than PUS-AgNW-PDMS conductor. [11]

Although in most works the resistance was increased after hundreds of the stretching cycles arising from destruction of conductive network, the contrary trend reported here was also observed in other nanowire composite such as CNT/PU, and CNT/ bismaleimide composites stretchable conductors. [30, 31] The resistance decreasing phenomenon may be induced by alignment enhancement of nanowires along the stretching direction of composites under the stretching/releasing process. During stretching, the AgNWs self-assembled and aligned themselves along the load direction without damaging the connection of the conductive network, which induced the decrease of resistance. [30, 31] In addition, as expected, the stretchable conductor after 1000th stretching cycle mentioned above show remarkable stability upon bending and twisting and the resistance kept unchanged around 1.2 Ω (Figure 5c).

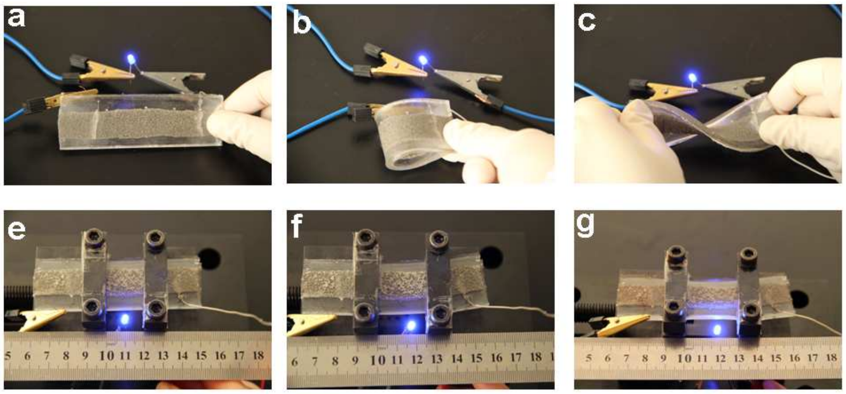


**Figure 6.** Optical images of an LED illuminated by using a PDA-PU-AgNW-Ag- PDMS film as the connecting wire under a) no strain, b) bending , c) twisting, and at e ) 0%, f) 20% and g) 50% strain.

As a proof-of-principle demonstration for stretchable electronics, a circuit containing a commercial light-emitting diode (LED) was fabricated with as-prepared PDA-PU-AgNW-Ag-PDMS stretchable conductor as connecting wire. As shown in Figure 6, the digital pictures show that the brightness of the LED lights almost kept unchanged after bending, twisting or stretching the stretchable conductors to a tensile strain of 50% under constant voltage, which implies that the fabricated conductors could retain constant resistance with shape deformation to a certain extent. These results are well demonstration of the excellent electromechanical stability of PDA-PU-AgNW-Ag-PDMS composites and their great potential for high-performance stretchable and flexible conductors.

## 3. Conclusion

In summary, we have fabricated AgNWs/AgNPs stretchable conductors based on 3D polydopamine-modified PU sponge. The PDA modified sponge as a skeleton shown improved adhesion to AgNWs. The conductivity of the micro-network was further improved by sputtering AgNPs onto the surface, resulted a ternary network structure. The resistances of PDA-PU-AgNW-Ag-PDMS stretchable conductor decreased obviously with bending or stretching cycles firstly, and keep stable even up to 1000th cycles. The composites exhibit high electrical conductivity (exceeding 27.78 S $cm^{-1}$) and excellent electromechanical stability under high tensile strain (50%) owing to the combined effects from the ternary network. Further fabrication of these ternary network stretchable conductors could aim to meet the demands of applications in flexible, stretchable, and foldable electronic devices.

## 4. Experimental Section

*Materials*: Dopamine hydrochloride was received from Sigma-Aldrich, and tris(hydroxymethyl) aminomethane was obtained from TCI chemicals. Silver nitrate ($AgNO_3$), polyvinylpyrrolidone (PVP, K30), sodium chloride and isopropyl alcohol (IPA) was purchased from Sinopharm. High-purity silver paste was obtained from SPI. PDMS (Sylgard 184) was received from Dow Corning. All the reagents were used as received without further purification. Polyurethane sponges are commercial products. The averaged size of pores, thickness of walls, and density are 590 ± 100 µm, 65 ± 6 µm and 0.12± 0.05 $g\cdot cm^{-3}$.

*Synthesis of AgNWs:* AgNWs were synthesized according to the method reported previously. [11] Briefly, PVP (5.86 g) were added into glycerol (190 mL) in a 500 mL round bottle flask and heating at 90 °C to dissolve under stirring. After the solution was cooled down to 50 °C, $AgNO_3$ powder (1.58 g) and a solution containing glycerol (10 mL), NaCl (59 mg) and $H_2O$ (0.5 mL) was added into the above solution respectively. Then the mixture was heated up to 210 °C under gentle stirring (~50 rpm). Subsequently, the as-obtained gray-green mixture was transfer immediately into a beaker, and 200 ml deionized water was added slowly. After stabilized for one week, a layer of sediment at the bottom of the beaker was collected and washed with isopropanol several times to remove the residual PVP.

*Preparation of PDA-PU:* A macroporous PU sponge was cleaned by ethanol for several times, and dried at 40 °C. The pre-cleaned PU sponge was cutted into small pieces with size of 5.0×1.5×0.1 cm. 0.446 g of tris(hydroxymethyl) aminomethane was dissolved in 180 g distilled water, and the pH was maintained at 8 ~ 9. After that, 0.258 g of dopamine was added and dissolved. All sponge pieces were dipped into the above solution at ambient temperature for 24 h, and then picked out and transferred into a dry oven at 40 °C for 2 h.

*Preparation of PDA-PU-AgNW-Ag and PU-AgNW-Ag:* All sponge pieces modified with and without polydopamine were dipped into the isopropyl alcohol solution of AgNWs (~20 mg/mL) prepared by the above method, then picked out and transferred into a petri dish, followed by adding AgNWs solutions (~1.3 g) through dropwise to enhance its uniformity. Afterwards, the obtained nanocomposite was transferred into a dry oven at 80 °C for 30 min to obtain PDA-PU-AgNW and PU-AgNW nanocomposites. A thin AgNP film was sputter coated on PDA-PU-AgNW and PU-AgNW nanocompsite using EMITECHK550X sputter coater at room temperature. Before deposition, the vacuum chamber of pressure was decreased down to $1\times10^{-1}$ mbar. The deposition time was 12 min with a current of 25 mA.

*Preparation of PDA-PU-AgNW-Ag-PDMS and PU-AgNW-Ag-PDMS stretchable conductor:* Copper wires were embedded and connected to the as-prepared conductors with silver paste. The distance between two copper wires was 5 cm. Pre-cured PDMS (mixing the "base" and the "curing agent" with a ratio of 10:1) were poured into the as-prepared *PDA-PU-AgNW-Ag* and PU-AgNW-Ag pieces, followed by degassing in vacuum oven for 30 min and thermally cured in a dry oven at 100 °C for 2 h. Then PDA-PU-AgNW-Ag-PDMS and PU-AgNW-Ag-PDMS composites were obtained through peeling off the films from the tailor-made moulds. The size of PU sponges embed in PDMS between two copper wired is 5.0×1.5×0.1 cm and is uniform for all measured samples.

*Electronic measurement:* Stretching tests were conducted using home-made equipment. The electrical resistance of the film during tensile or bending testing was measured using a Keithley 2700 multimeter in a four-probe method setup under ambient condition.

For the stretchable circuit device experiment, a simple circuit consisting of commercially available light-emitting diode (LED) light was fabricated. PDA-PUS-AgNW-Ag- PDMS film was used as electrical wires in the circuit where the copper wires were adhered to the two sides of the films.

*Characterization*: The surface morphology of sample was characterized using a CAMSCAN APOLLO 300 field-emission scanning electron microscope (SEM). The X-ray diffraction (XRD) patterns of the samples were obtained using a Bruker D8 X-ray diffractometer with Cu Kα irradiation at λ = 0.15418 nm. The optical images of PU and PDA-PU sponge were obtained on a Zess Axio Scope A1 microscope.

## Reference

[1] T. Sekitani, T. Someya, *Adv. Mater.* **2010**, *22*, 2228.

[2] M. L. Hammock, A. Chortos, B. C. Tee, J. B. Tok, Z. Bao, *Adv. Mater.* **2013**, *25*, 5997.

[3] K. Xie, B. Wei, *Adv. Mater.* **2014**, *26*, 3592.

[4] S. Yao, Y. Zhu, *Adv. Mater.* **2015**, *27*, 1480.

[5] X. Wang, H. Hu, Y. Shen, X. Zhou, Z. Zheng, *Adv. Mater.* **2011**, *23*, 3090.

[6] Y. Yu, J. Zeng, C. Chen, Z. Xie, R. Guo, Z. Liu, X. Zhou, Y. Yang, Z. Zheng, *Adv. Mater.* **2014**, *26,* 810.

[7] (a) M. Chen, L. Zhang, S. Duan, S. Jing, H. Jiang, C. Li, *Adv. Funct. Mater.* **2014**, *24*, 7548; (b) T. A. Kim, S. Lee, H. Kim, M. Park, *RSC Adv.*, **2012**, 2, 10717.

[8] Y. Sun, W. M. Choi, H. Jiang, Y. Y. Huang, J. A. Rogers, *Nat. Nanotechnol.* **2006**, *1*, 201.

[9] Z. Niu, H. Dong, B. Zhu, J. Li, H. H. Hng, W. Zhou, X. Chen, S. Xie, *Adv. Mater.* **2013**, *25*, 1058.

[10] 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.

[11] J. Ge, H. B. Yao, X. Wang, Y. D. Ye, J. L. Wang, Z. Y. Wu, J. W. Liu, F. J. Fan, H. L. Gao, C. L. Zhang, S. H. Yu, *Angew. Chem. Int. Ed.* **2013**, *52*, 1654.

[12] Z. Shi, H. Gao, J. Feng, B. Ding, X. Cao, S. Kuga, Y. Wang, L. Zhang, J. Cai, *Angew. Chem. Int. Ed.* **2014**, *53*, 5380.

[13] J. Zhi, W. Zhao, X. Liu, A. Chen, Z. Liu, F. Huang, *Adv. Funct. Mater.* **2014**, *24*, 2013.

[14] Z. P. Chen, W. C. Ren, L. B. Gao, B. L. Liu, S. F. Pei, H. M. Cheng, *Nat. Mater.* **2011**, *10*, 424.

[15]G. S. Jeong, D. H. Baek, H. C. Jung, J. H. Song, J. H. Moon, S. W. Hong, I. Y. Kim, S. H. Lee, *Nat. commun.* **2012**, *3*, 977.

[16] H. L. Gao, L. Xu, F. Long, Z. Pan, Y. X. Du, Y. Lu, J. Ge, S. H. Yu, *Angew. Chem. Int. Ed.* **2014**, *53*, 4561;

[17] (a) W. Zeng, L. Shu, Q. Li, S. Chen, F. Wang, X. M. Tao, *Adv. Mater.* **2014**, *26*, 5310; (b) H. Lee, B. Seong, H. Moon, D. Byun, *RSC Adv.*, **2015**, *5*, 28379.

[18] M. Park, J. Park, U. Jeong, *Nano Today* **2014**, *9*, 244.

[19] Y. Kim, J. Zhu, B. Yeom, M. Di Prima, X. Su, J. G. Kim, S. J. Yoo, C. Uher, N. A. Kotov, *Nature* **2013**, *500*, 59.

[20] L. Cai, J. Li, P. Luan, H. Dong, D. Zhao, Q. Zhang, X. Zhang, M. Tu, Q. Zeng, W. Zhou, S. Xie, *Adv. Funct. Mater.* **2012**, *22*, 5238.

[21] H. W. Engels, H. G. Pirkl, R. Albers, R. W. Albach, J. Krause, A. Hoffmann, H. Casselmann, J. Dormish, *Angew. Chem. Int. Ed.* **2013**, *52*, 9422.

[22] J. Sedo, J. Saiz-Poseu, F. Busque, D. Ruiz-Molina, *Adv. Mater.* **2013**, *25*, 653.

[23] R. Li, K. Parvez, F. Hinkel, X. Feng, K. Mullen, *Angew. Chem. Int. Ed.* **2013**, *52*, 5535.

[24] Q. Wei, F. Zhang, J. Li, B. Li, C. Zhao, *Polym. Chem.* **2010**, *1*, 1430.

[25] M. Ji, N. Jiang, J. Chang, J. Sun, *Adv. Funct. Mater.* **2014**, *24*, 5412.

[26] T. Akter, W. S. Kim, *ACS Appl. Mater. Interfaces* **2012**, *4*, 1855.

[27] C. Zhang, K. Lv, N. Hu, L. Yu, X. Ren, S. Liu, S. Yu, *Small* **2012**, *8,* 2936.

[28] J. Chen, H. Bi, S. Sun, Y. Tang, W. Zhao, T. Lin, D. Wan, F. Huang, X. Zhou, X. Xie, M. Jiang, *ACS Appl. Mater. Interfaces* **2013**, *5*, 1408.

[29] F. Xu, Y. Zhu, *Adv. Mater.* **2012**, *24*, 5117.

[30] S. Shang, W. Zeng, X. Tao, *J. Mater. Chem.* **2011**, *21*, 7274.

[31] Q. Cheng, J. Bao, J. Park, Z. Liang, C. Zhang, B. Wang, *Adv. Funct. Mater.* **2009**, *19*, 3219.