

Supporting Information

Self-compensation stretchable conductor based on viscous fluid for wide-range flexible sensors

*Minxuan Xu **, *Dafei Wei*, *Tengyue Song*, *Chun Dou*, *Yueqin Shi*, *Xin Li*, *Zhe Kong*, *Qi Zhang **

¹ Center for Advanced Optoelectronic Materials, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, P. R. China

² Key Laboratory of Novel Materials for Sensor of Zhejiang Province, Hangzhou Dianzi University, Xiasha Higher Education Zone, Hangzhou, 310018, P. R. China

*Corresponding authors, E-mail: gizhang@hust.edu.cn

Table of contents

1. Materials and Methods
2. Supplementary Table
3. Supplementary Figures
4. Supplementary References

1. Materials and Methods

Material: The CNF powders (99.6%, diameter: 4~10nm, length: 1~3 μ m) was purchased from Guilin Qihong Technology Co., Ltd. NaCl crystal ($\geq 99.5\%$) was provided by Sinopharm Chemical Reagent Co., Ltd. The Ecoflex silicone rubber (00-30) was provided by Smooth-On and the acrylic tape (3mm thick, 4965PV0) was purchased from TESA. All these reagents were used without further purification.

Preparation of viscous GCB/CNF/NaCl fluid: At room temperature, 1g of CNF powders was added into deionized water (DI, 100ml) and stirred using an Ultrasonic Cell Pulverizer (15000 rpm) for 10 minutes. Then, GCB (0.5g) and NaCl (1g) were dispersed in 5 g of the above fluid to form a uniform and stable liquid. The obtained black viscous GCB/CNF/NaCl fluid was in a mass ratio of 1:10:2.

Fabrication of the SCD: The cleaned PET substrate (100 μ m thick) was cut into size of 40mm \times 30mm and was patterned in a rectangular shape (20mm \times 5mm \times 3mm) with acrylic tape covered around the PET. Then, the rectangular shape was filled up with 2g mixed Ecoflex silicone rubber (mass ratio A: B=1:1) after a wire (d=1mm) was left in the middle. The Ecoflex was cured after 8h and the acrylic tape was detached from the PET slide. The wire was pulled out and left a cylindrical cavity (length: 20mm, diameter: 1mm). A pair of platinum strip electrodes was used to block the cylindrical cavity after the as-obtained GCB/CNF/NaCl fluid injected into. Finally, the highly stretchable conductor (20mm \times 5mm \times 3mm) was manufactured after sealing and peeling off the PET.

Characterizations and testing: A scanning electron microscope (SEM, *Apereo S HiVac*) was used to measure the morphology of the solid GCB/CNF/NaCl composite. The conductance of the viscous ionic liquid was explored by the LCR (*VC4091A*). A mechanical viscometer (*NDJ-1*) was used to investigate the viscosity of different fluids, and the stress-strain curves were recorded by a tensile testing machine (*ZQ-990A*). Finally, the tensile deformation of the SCD was applied by a self-developed smart tensile table. The electromechanical properties of the SCD were measured with a digital source meter (*Keithley DMM-7510*).

2. Supplementary Table

Table S1. Comparison of our SCD with other stretchable conductors in terms of their performance. The “-” indicates not given.

Materials	Conductivity	Sensitivity (GF)	Working range	Cycles	Stability	Ref.
GCB/CNF/NaCl	30.6 S/m	0.066	100%	5000	72h	This work
AgNWs/PDMS	14 Ω /sq	3.3	20%	-	20d	(1)
Au nanomesh/PDMS	21 Ω /sq	0.21~1.94	160%	1000	-	(2)
AgNW/CNT	-	1.1	460%	10000	-	(3)
Metal/graphene	1 Ω /sq	1.2	50%	-	-	(4)
Graphene/PDMS	-	1.5	30%	1000	-	(5)
3D graphene/PDMS	1.16×10^3 S/m	5.34	200%	5000	-	(6)
Metal-mesh	2 Ω /sq	1.8	55%	500	-	(7)
Ag/PTFE	5.71×10^3 S/m	20~33	50%	10000	-	(8)
TPU fiber /CNT/PDMS	20~27 S/m	0.0714~0.39	100%	700	-	(9)
AuNS	5 Ω /sq	8	50%	1000	-	(10)
PEDOT:PSS/WPU	4×10^4 S/m	0.42	30%	430	-	(11)
BSS bundle mesh	151 Ω /sq	25.75	40%	-	-	(12)
PTFE/APC-PTFE/PTFE	33.6 Ω /sq	25	40%	10000	-	(13)
SEBS/CNT@PANi	240 S/m	1.4	100%	1000	-	(14)

3. Supplementary Figures

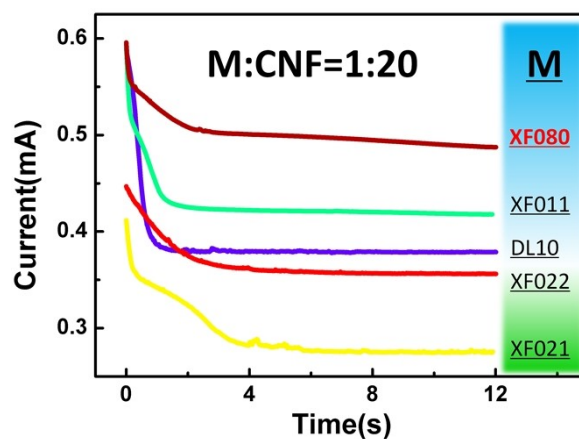


Figure S1. Conductivity of different carbon materials.

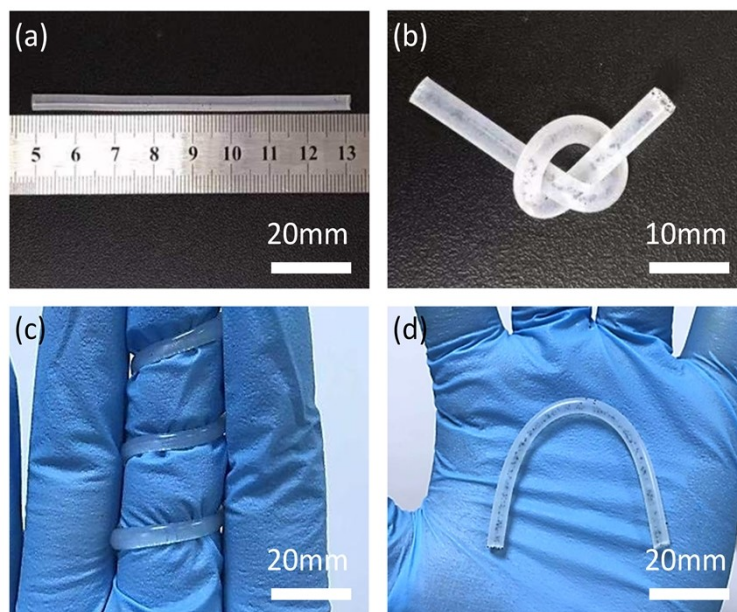


Figure S2. Optical photographs of cylindrical SCD in various states. (a) Straight. (b) Knotted. (c) Tangled. (d) Bent.

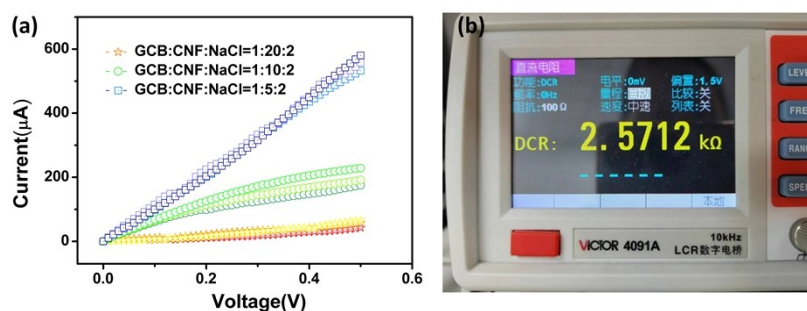


Figure S3. Electrical performance of the viscous fluids. (a) Current voltage (I-V) curves of viscous fluids with different mass ratios. (b) The DC resistance of the viscous fluid (GCB:CNF:NaCl=1:10:2).

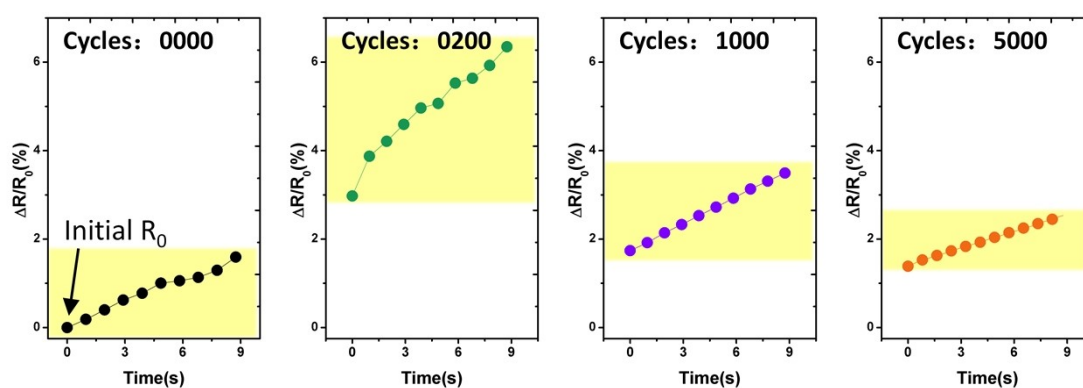


Figure S4. Real-time resistance within 9 seconds after 0, 200, 1000, and 5000 cycles of stretching ($\epsilon=50\%$).



Figure S5. The image records of AC resistance within 4 days from the conductor without (upper) or with (bottom) deformation, respectively.

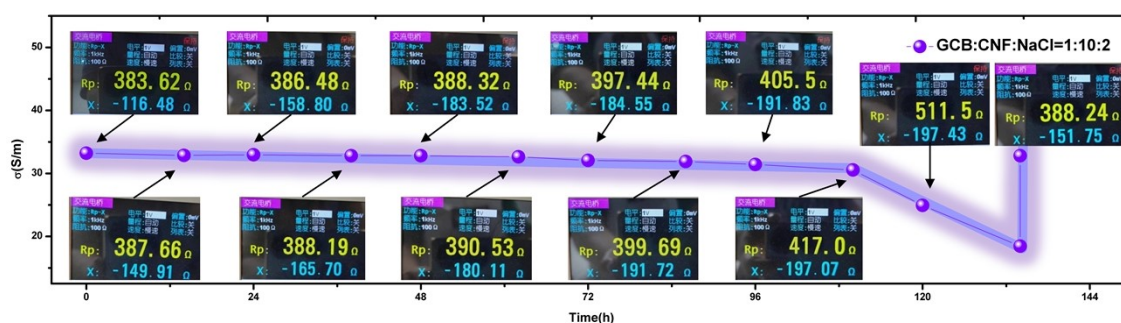


Figure S6. The conductivity of a SCD based on a poor vapor permeability epoxy resin varies upon time.

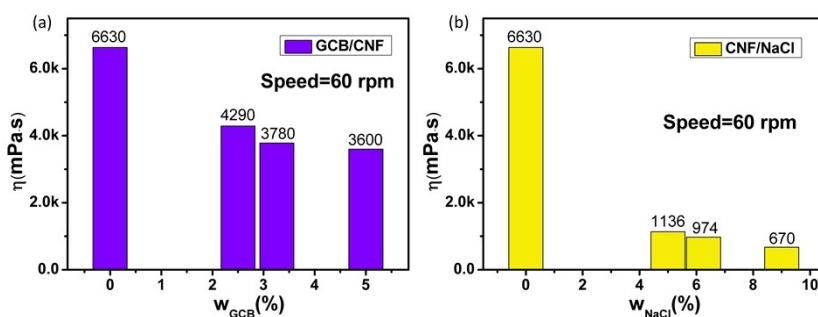


Figure S7. Effect of additive mass ratio on fluid viscosity. (a) The additive is GCB. (b) The additive is NaCl.

4. Supplementary References

- [1] T. Cheng, Y-Z. Zhang, W-Y. Lai, Y. Chen, W-J. Zeng, W. Huang, *J Mater. Chem. C*, **2014**, 2, 10369.
- [2] C. F. Guo, T. Y. Sun, Q. H. Liu, Z. G. Suo, Z. F. Ren, *Nat. Commun.*, **2014**, 5,

3121.

- [3] P. Lee, J. Ham, J. Lee, S. Hong, S. Han, Y. D. Suh, S. E. Lee, J. Yeo, S. S. Lee, D. Lee, S. H. Ko, *Adv. Funct. Mater.*, **2014**, *24*, 5671-5678.
- [4] B. W. An, B. G. Hyun, S-Y. Kim, M. Kim, M-S. Lee, K. Lee, J. B. Koo, H. Y. Chu, B-S. Bae, J-U. Park, *Nano Lett.*, **2014**, *14*, 6322-6328.
- [5] J-Y. Hong, W. Kim, D. Choi, J. Kong, H. S. Park, *ACS Nano*, **2016**, *10*, 9446-9455.
- [6] F. Pan, S-M. Chen, Y. H. Li, Z. C. Tao, J. L. Ye, K. Ni, H. Yu, B. Xiang, Y. B. Ren, F. X. Qin, S-H. Yu, Y. W. Zhu, *Adv. Funct. Mater.*, **2018**, *28*, 1803221.
- [7] C. P. Zhang, A. Khan, J. X. Cai, C. W. Liang, Y. J. Liu, J. H. Deng, S. Y. Huang, G. X. Li, W-D. Li, *ACS Appl. Mater. Interfaces*, **2018**, *10*, 21009-21017.
- [8] J-E. Lim, S. Yoon, B-U. Hwang, N-E. Lee, H-K. Kim, *Adv. Mater. Interfaces*, **2019**, *6*, 1801936.
- [9] L. Wang, Y. Chen, L. W. Lin, H. Wang, X. W. Huang, H. G. Xue, J. F. Gao, *Chem. Eng. J*, **2019**, *362*, 89-98.
- [10] S. Jeong, S. Heo, M. Kang, H-J. Kim, *Mater. Design*, **2020**, *196*, 109178.
- [11] L. Zhang, K. S. Kumar, H. He, C. J. Cai, X. He, H. X. Gao, S. Z. Yue, C. S. Li, R. C-S. Seet, H. L. Ren, J. Y. Ouyang, *Nat. Commun.*, **2020**, *11*, 4683.
- [12] E. Y. An, S. Y. Lee, S. G. Lee, E. Lee, J. J. Baek, G. Shin, K. H. Choi, J. H. Cho, G. Y. Bae, *Nanomaterials*, **2021**, *11*, 2865.
- [13] S. Yoon, H-M. Sim, S. Cho, H. Ko, Y. Park, Y. Kim, H-K. Kim, *Adv. Mater. Interfaces*, **2021**, *8*, 2001500.
- [14] M. Zhu, T. Yang, L. T. Wang, M. Y. Xiong, W. J. He, Y. W. Chen, W. L. Deng, X. Y. Li, *Chem. Eng. J*, **2022**, *430*, 132911.