Supporting Information

Highly Conductive Carbon-Based Aqueous Inks toward Electroluminescent Devices, Printed Capacitive Sensors and Flexible Wearable Electronics

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Abstract: Carbon-based conductive inks are one of the most important materials in the field of printing electronics. However, most carbon-based conductive inks have large electrical resistance, or as expensive as graphene, lacking a kind of conductive inks with good conductivity and economical price. Here, we propose a low cost and environmentally friendly formula based on dihydroxyphenyl-functionalized multi-walled carbon nanotubes (MWNT-f-OH)/carbon black/graphite as conductive fillers and waterborne acrylic resins as binders for preparing highly conductive carbon-based aqueous inks (HCCA-inks). The study found that when the mass fraction of carbon black, graphite and MWNT-f-OH was 3.0%, 10.2% and 4.1% respectively, optimal conductivity (sheet resistance up to Rs ≈ 29 Ω sq^{-1}) was achieved, and printed the HCCA-inks on paper withstand extremely high folding cycles (>2 000 cycles) the resistance value of the flexible circuit increased 11%. The carbon-based aqueous inks showed high electrical conductivity and excellent mechanical stability which could be effectively used in advanced technologies such as flexible wearable electronics， electroluminescent (EL) devices and printed capacitive sensors and other printed electronics.

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Video files

Video S1. Movie showing the tester's hand moving away and close to the sensing block which made of conductive ink drawn on paper, controlling the opening and closing of the EL device.

Video S2 and Video S3. Are showing the induction sound devices made with different conductive inks, video 2 is touch sound, and video 3 is induction sound. Their difference is the conductivity of the ink and the area of the induction block.

Table S1. Comparison of conductivity of inks prepared by different conductive fillers

Conductive fillers	Electrical conductivity	Thickness	refs
Carbon black / Cellulose acetate	250 Ω sq^{-1}	9 μ m	21
Graphite / Carbon black	38.7Ω sq^{-1} - 252.2 Ω sq^{-1}	6μm - 9.5 μ m	12
Graphitic nanoparticles	220 $Ωsq^{-1}$	25 μ m	22
MWCNT/Wax	$200Ωsq^{-1}$	90 μ m	10
Gr/Polyamic acid	$26Ωsq^{-1}$	15 μ m	24
Gr / MWNT-*f*-OH	25 Ω sq^{-1}	2.86 μ m	2, 23
Carbon black / Graphite / MWNT-*f*-OH	29 Ω sq^{-1}	40 μ m	this work

Table s2. The mass fraction of each conductive filler in the ink in each experimental group and the working parameters of FS-400 laboratory disperser in lapping experiment.

Experimental groups	a	b	c	d	e	f
Carbon black %	0	1.5%	3.0%	4.5.%	6.0%	7.1%
MWNTs-f-OH %	7.1%	5.6%	4.1%	2.6%	1.1%	0
Graphite %	10.2%	10.2%	10.2%	10.2%	10.2%	10.2%
Running speed（rpm）	3600	3600	3600	3600	3600	3600
Grinding time (h)	2	2	2	2	2	2
	3	3	3	3	3	3
	4	4	4	4	4	4
	5	5	5	5	5	5
	6	6	6	6	6	6

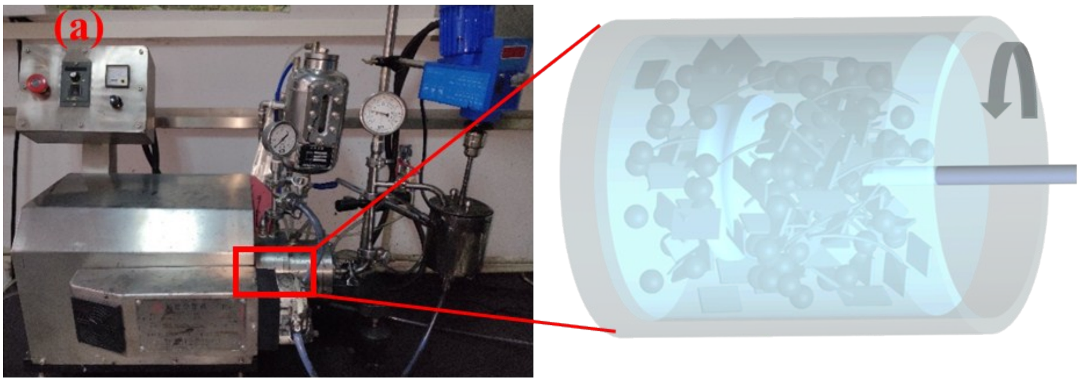


Fig s1. Grinding dispersed conductive fillers.

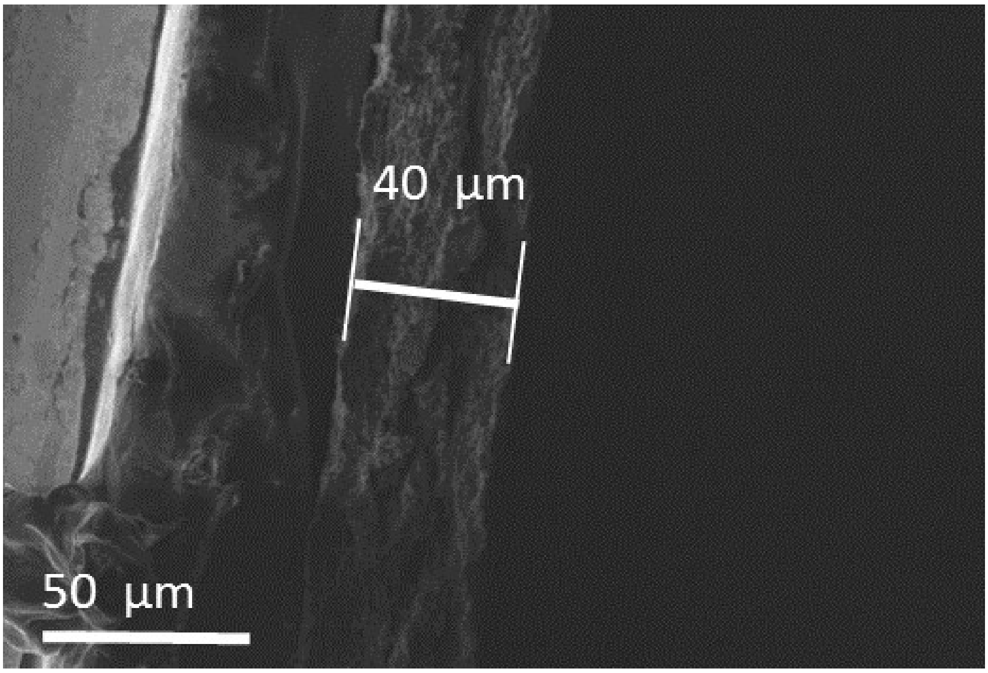
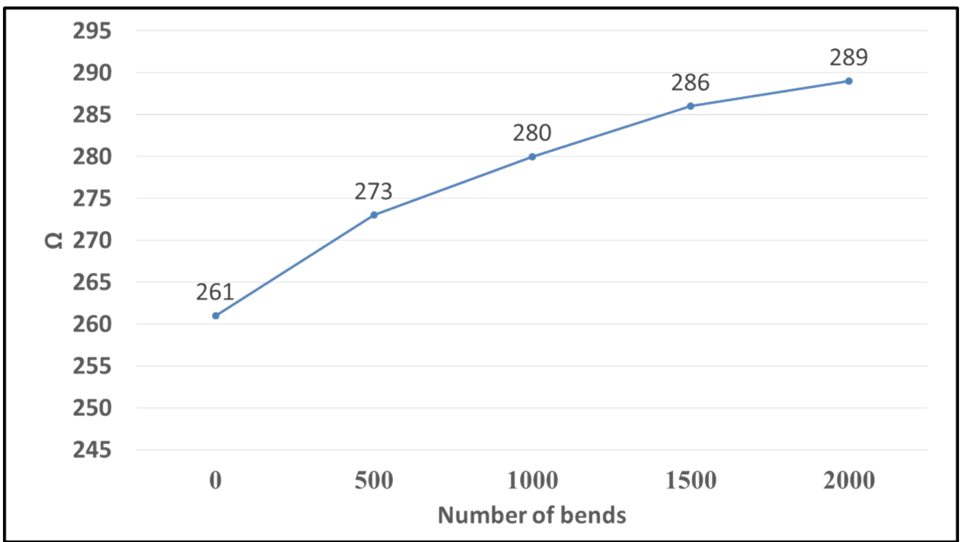
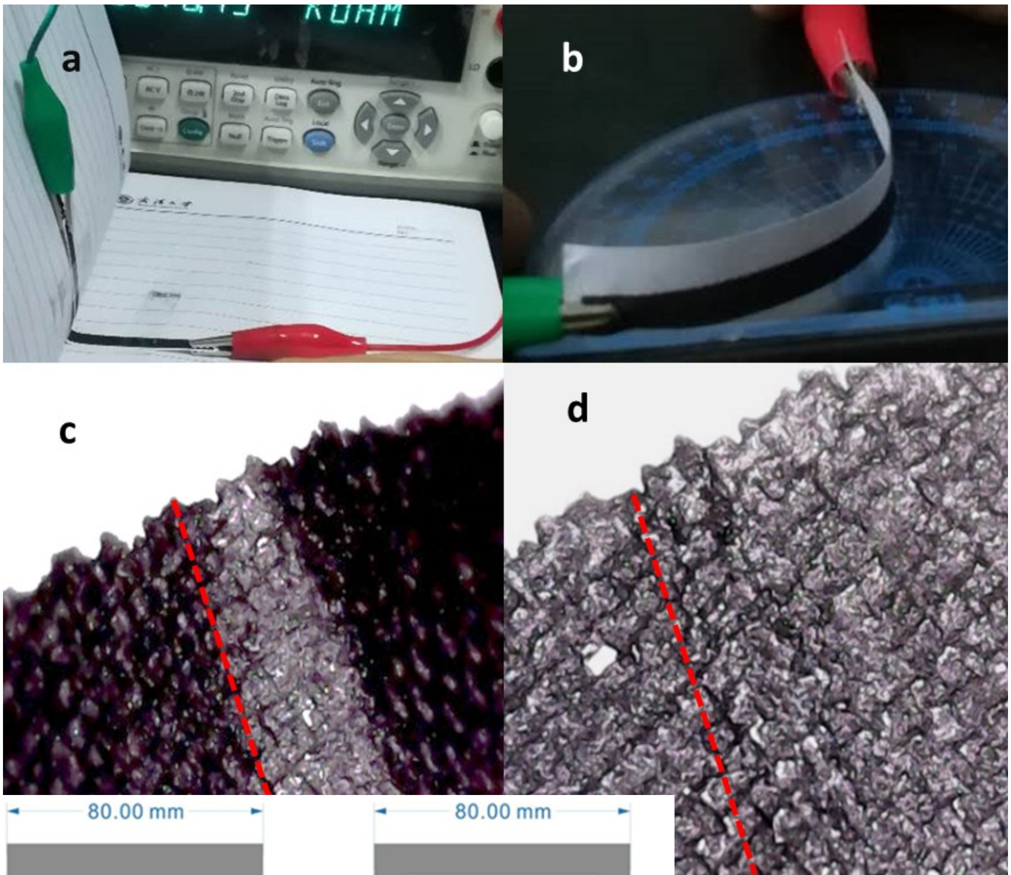
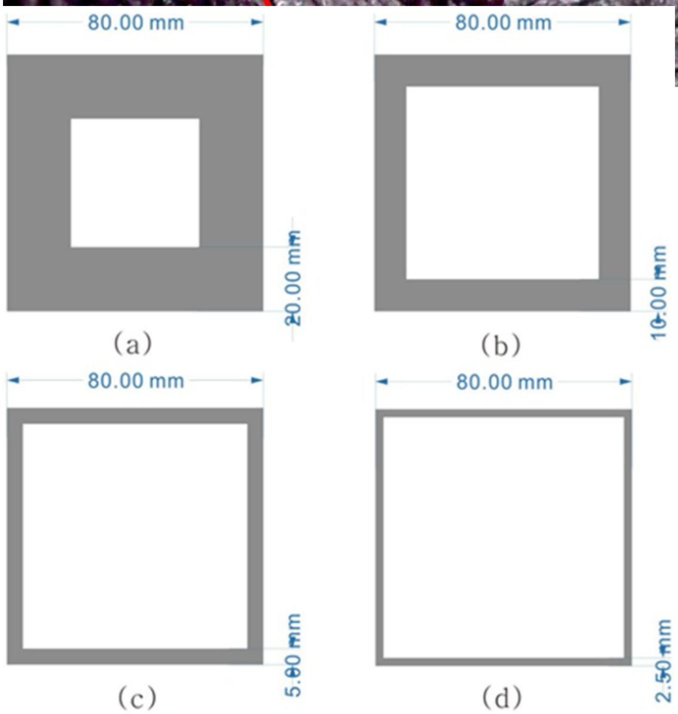


Fig s2. SEM images of the cross-section of the ink layer.



Figs3. The relationship between the resistance and the number of bends of the paper-based flexible circuit





Figs4. (a, b) Bending test of the paper-based flexible circuit. (c, d) The microstructures of the paper-based flexible circuit after bending 500 times and 1000 times respectively.

Fig s5. The sensor with the same side length of 8 cm square. (a) The printing ink track width 20mm. (b) The printing ink track width 10mm. (c) The printing ink track width 5mm. (d) The printing ink track width 2.5mm.

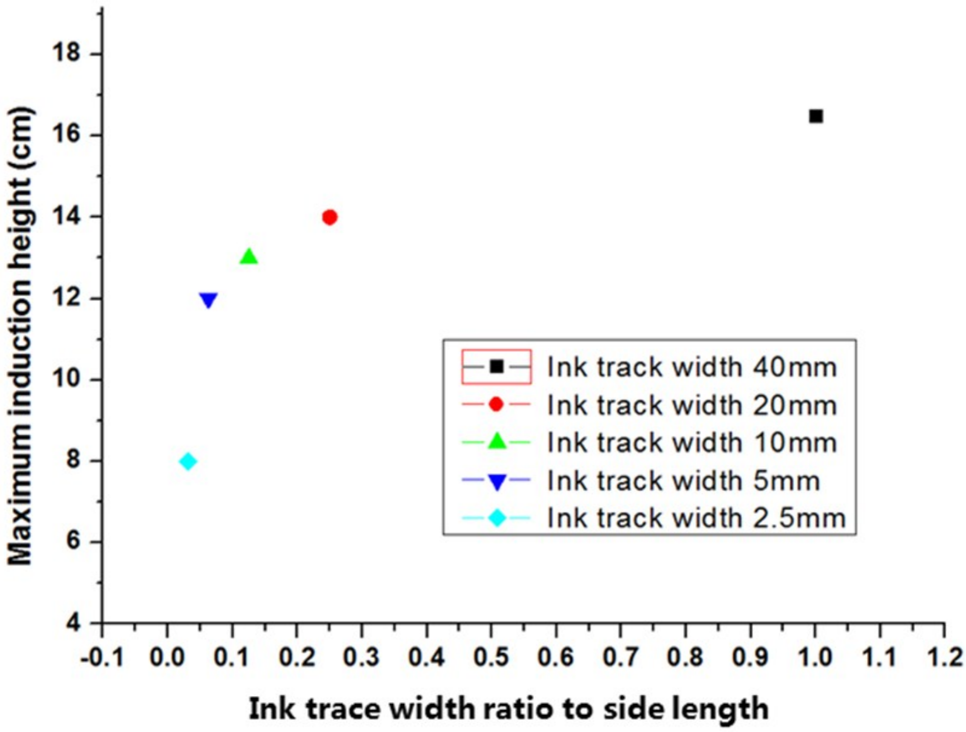


Fig s6. Maximum induction height of different printing ink trace width sensors

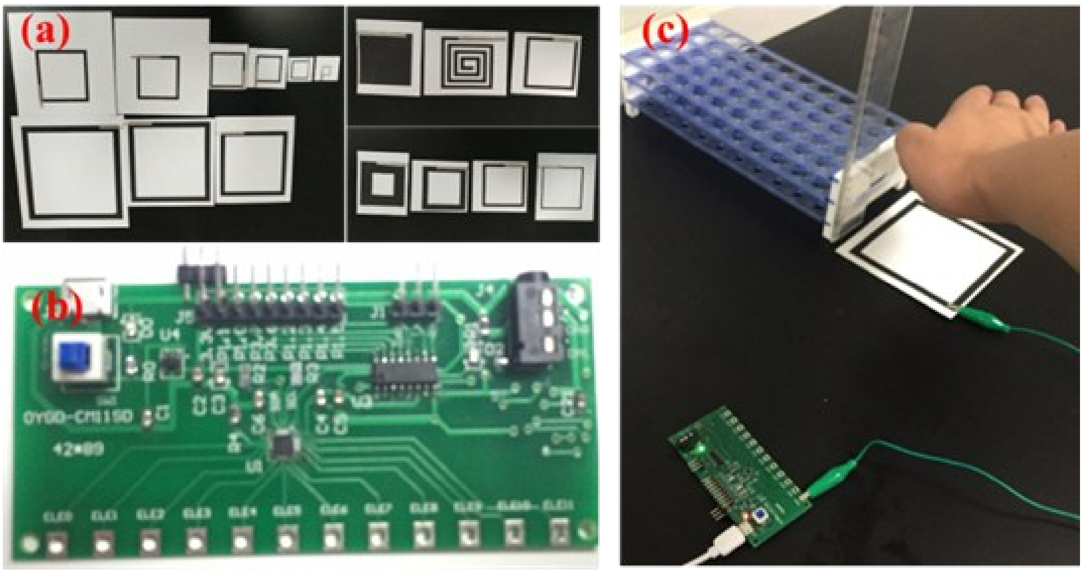
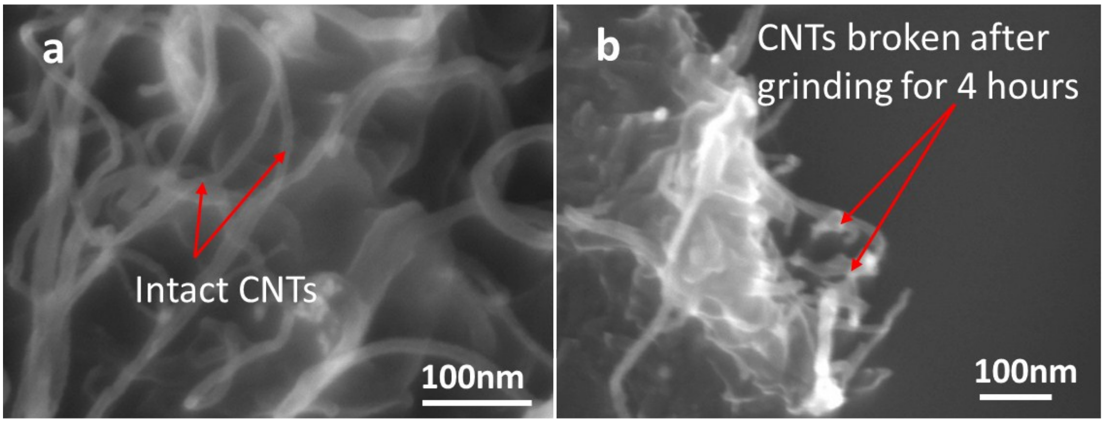


Fig s7: Preparation and test diagram of printed capacitive sensors. (a) Induction modules of different sizes and shapes. (b) Capacitive touch panels. (c) Optimal sensing distance measurement.



Figs8 (a, b) SEM comparison of carbon nanotubes before and after grinding for 4 h.