

Supporting information

Highly Sensitive Piezo-Resistive Graphite Nanoplatelet-Carbon Nanotube
Hybrids/ Polydimethylsilicone Composites with Improved Conductive
Network Construction

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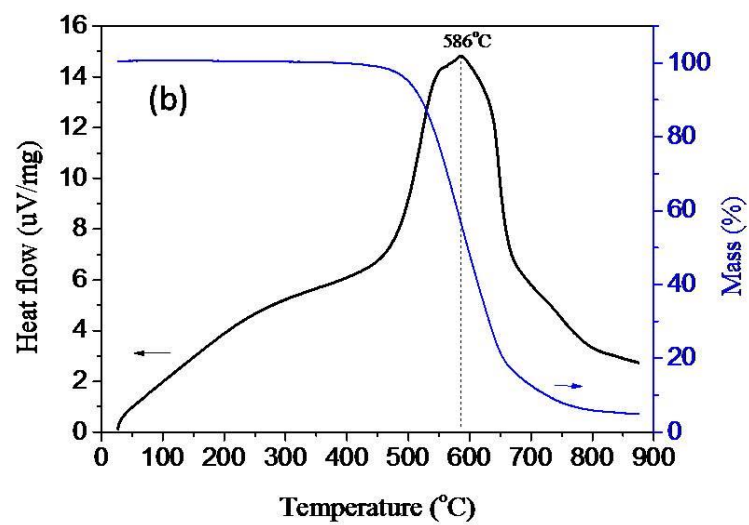


Figure S1. The TGA and DSC curves of the GCH particles.

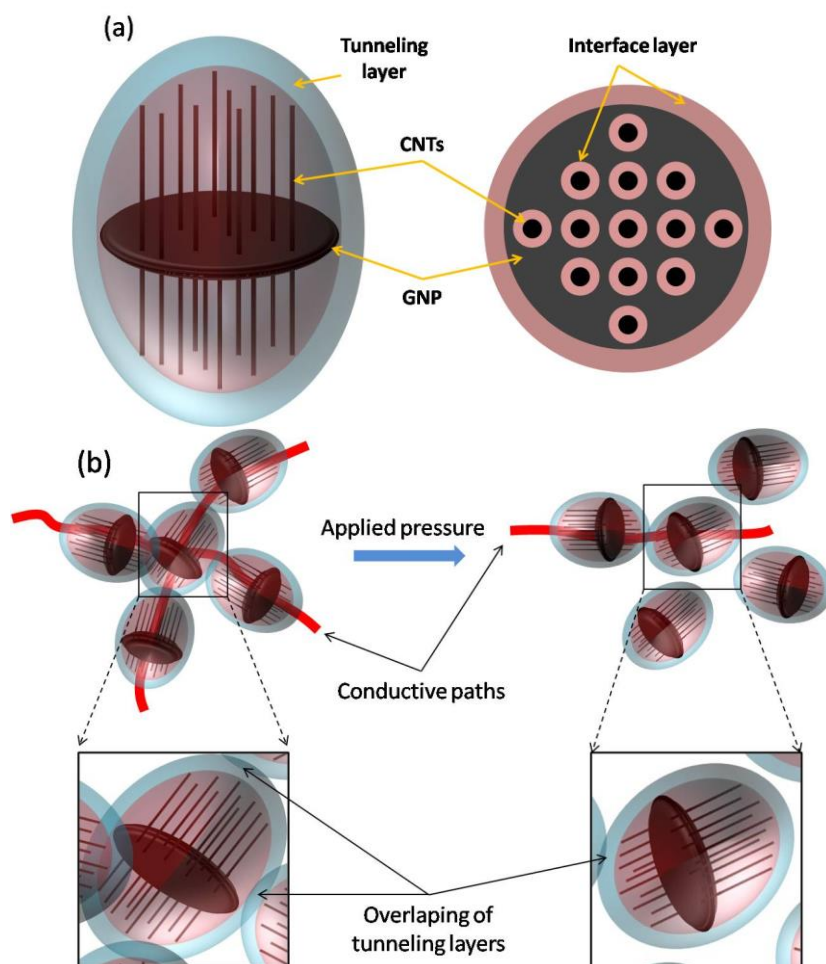


Figure S2. Schematic of (a) the construction of GCHs with improved interfacial area and effectively conductive space. (b) the “hub” structure of GCHs is capable of multi-point communicating with neighboring particles via direct contact or tunneling effect. While, the multi-point communicating is easily influenced by applied pressure, transforming the conductive “traffic hub” to the “isolated island”

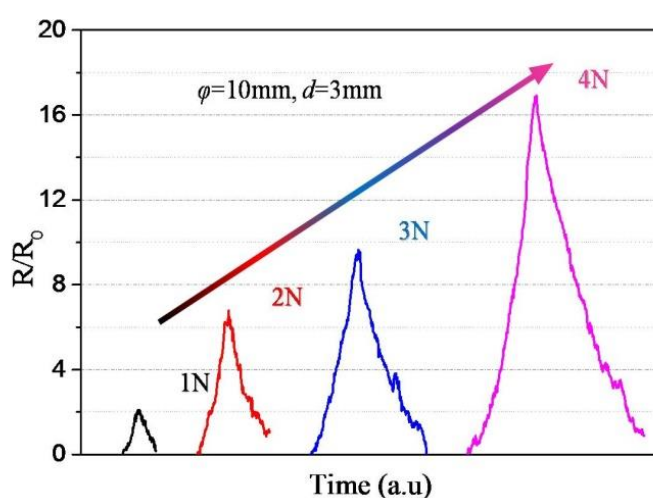


Figure S3. The relative resistance change of composite with GCHs 0.8 vol% under a very low charge, showing an excellent piezo-resistive sensitivity.

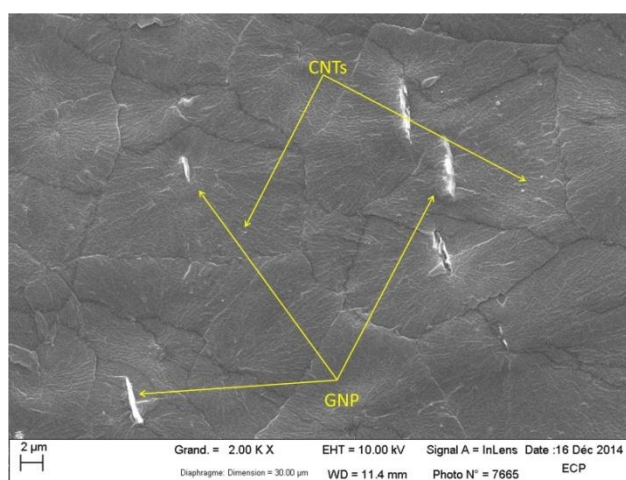


Figure S4. Typical SEM images of fractured surface of GNPs/CNTs/PDMS three-phase composite.

Table S1 The different composition of 4 PDMS-based composite samples.

Sample ¹	GCHs content (wt%)	CNTs content ² (wt%)	GNPs content ² (wt%)
1	1.52	-	-
2	2.28	-	-
3	-	0.76	0.76
4	-	1.14	1.14

¹Sample 3 and 4 are samples that GNPs/CNTs/PDMS three-phase composite with equivalent contents of GNPs and CNTs to GCHs/PDMS composites, sample 1 and 2, respectively.

²The mass ratio between CNTs and GNPs substrate of GCHs particles is approximately equal to 1.

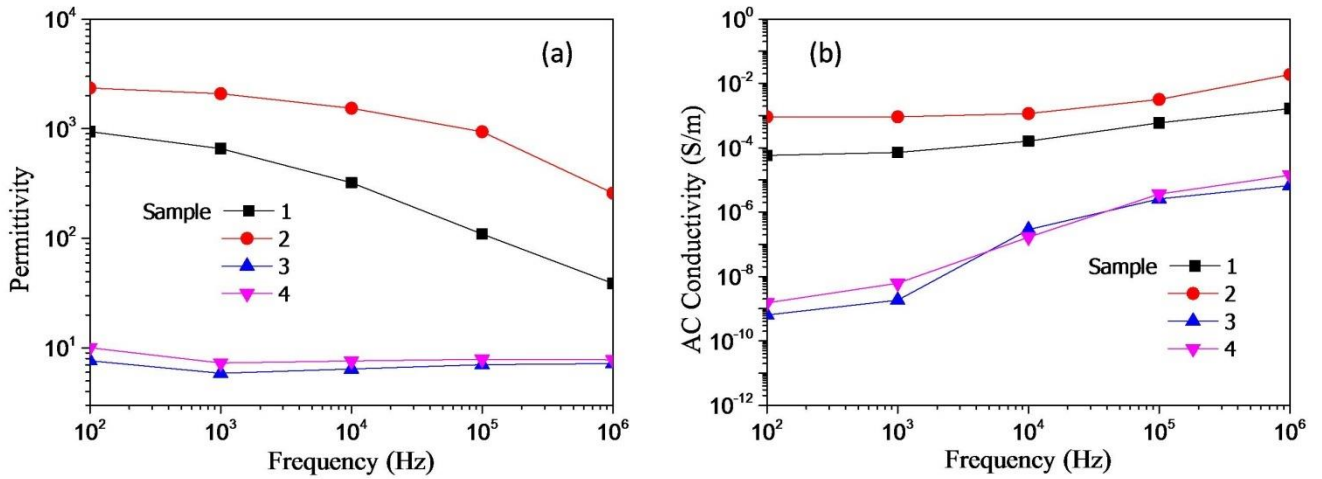


Figure S5. Frequency-dependent (a) dielectric permittivity and (b) AC conductivity of GCHs/PDMS composite and GNPs/CNTs/PDMS three-phase composite, respectively.

The fitting models were derived from the previous electrical resistance model,^{S2} which are as follows:

$$\frac{R}{R_0} = \left(\frac{Ns}{N_0s_0} \right) \exp \left[\frac{4\pi}{h} \sqrt{2m\phi} (s - s_0) \right] \quad (S1)$$

$$s = s_0 \left[1 + \left(\frac{|l - l_0|}{l_0} \right) \right] \quad (S2)$$

$$p = E \left(\frac{|l - l_0|}{l_0} \right) \quad (S3)$$

Where R , R_0 , N , N_0 , s , s_0 are the total resistance of composite, the number of conductive paths

and the minimum distance between conductive fillers of loading state and initial unloading state, respectively. l is the thickness of disk-shaped composite under applied compression, the l_0 is the initial thickness. The h the Plank constant and the m the electron mass. ϕ is the height of the potential barrier between neighboring fillers. p and E are the applied compression pressure and elastic modulus of composite, respectively.

For our GCHs/PDMS composite system, combining of the Equation 1, 2 and 3, the relationship between resistance change of the “tunneling state” and applied pressure is as follow:

$$N = \frac{N_0}{\exp[A_1(|l - l_0|/l_0)]} \quad (S4)$$

$$R/R_0 = (1 + p)\exp(A \times p) \quad (S5)$$

Where A_1 is a constant, $A = [\frac{4\pi}{h}\sqrt{2m\phi} + A_1]/E$.

For the “transitional state”, the conductive paths start to be influenced by the applied pressure:

$$N = \frac{N_0}{\exp[A_2(|l - l_0|/l_0) + B(|l - l_0|/l_0)^2]} \quad (S6)$$

$$R/R_0 = (1 + p)\exp(A \times p + B \times p^2) \quad (S7)$$

Where A_2 and B are constants, $A = [\frac{4\pi}{h}\sqrt{2m\phi} + A_2]/E$.

For the “discontinuous state”, the conductive network is strongly broken by the further increasing applied pressure:

$$N = \frac{N_0}{\exp[A_3(|l - l_0|/l_0) + B(|l - l_0|/l_0)^2 + C(|l - l_0|/l_0)^3]} \quad (S8)$$

$$R/R_0 = (1 + p)\exp(A \times p + B \times p^2 + C \times p^3) \quad (S9)$$

Where A_3 , B and C are constants, $A = [\frac{4\pi}{h}\sqrt{2m\phi} + A_3]/E$.

Table S2. Fitted coefficients and R^2 of three states.

	Initial state	Transitional state	Discontinuous state
A	17.094	8.416	-21.212
B	-	28.997	245.643
C	-	-	-391.309
R^2	0.966	0.99	0.998

Table S3. Comparison of the piezo-resistive sensitivity of carbon-fillers/PDMS composites under the small applied pressure.

Type (specific)	f_c (vol.%)	PS* (kPa^{-1})	GF	Conditions	Ref
GNP-PDMS	0.9	0.067		0.3 MPa	S2
CB-PDMS	-	-0.001		0.3 MPa	S3
CB-PDMS		<0.001		0.3 MPa	S4
CNT-PDMS	-		1.25	Strain~0.4	S5
CNT film-PDMS	-	~0.33		3 kPa	S6
Carboxyl-MWCNT-PDMS	-	0.013		3 MPa	S7
CNT-PDMS	-		~2-3	Strain~100%	S8
P3HT-CNT-PDMS	-	0.0017		0.12 MPa	S9
CNT-CB-PDMS	-				S10
(AR of CNT=117)		0.01		0.3 MPa	
(AR of CNT=437)		0.01		0.3 MPa	
Alkyl-Graphene-PDMS	0.63	<0.1		1 MPa	S11
GCHs-PDMS	0.64	0.6	~1000	0.32 MPa	

*The pressure or strain sensitivities were calculated and estimated from the curves in relevant references.

Supporting References

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