# Supporting Information

#### Electrical Energy Generated by Silicones Filled with Nanospring-Carbon-Nanotubes

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Figure S1. Photo of the DEG setup used.



Figure S2. SEM images of the cross-section of **PDMS-CN-CNT** (top) and of **P3-Cl-CNT** (bottom) thin film elastomers.



Figure S3. Tensile tests of **PDMS**. Three independent tests were performed.



Figure S4. Tensile tests of Elastosil®Film. Three independent tests were performed.



Figure S5. Tensile tests of **PDMS-CN**. Three independent tests were performed.



Figure S6. Tensile tests of **PDMS-CN-CNT**. Three independent tests were performed.



Figure S7. Tensile tests of P3-Cl. Three independent tests were performed.



Figure S8. Tensile tests of P3-Cl-CNT. Two independent tests were performed.

## DEG Measurements at 33% strain



Figure S9. Output voltage of five different **PDMS** electret DEGs at 33% strain.



Figure S10. Output voltage of five different Elastosil®Film electret DEGs at 33% strain.



Figure S11. Output voltage of four different **PDMS-CN** electret DEGs at 33% strain.



Figure S12. Output voltage of five different **PDMS-CN-CNT** electret DEGs at 33% strain.



Figure S13. Output voltage of five different **P3-Cl** electret DEGs at 33% strain.

## DEG measurements at 66% strain



Figure S14. Output voltage of five different **PDMS** electret DEGs at 66% strain.



Figure S15. Output voltage of five different Elastosil®Film electret DEGs at 66% strain



Figure S16. Output voltage of five different **PDMS-CN** electret-DEGs at 66% strain.



Figure S17. Output voltage of five different **PDMS-CN-CNT** electret DEGs at 66% strain.



Figure 18. Average output voltage obtained from five different DEG *versus* permittivity  $\varepsilon'$  of the DE at 33% and at 66% strain. For the generators strained by 66%, a clear increase in the output voltage with the dielectric permittivity is seen.



Figure S19. Photographs of a dispersion of NS-CNTs in DMF after standing for 1 month at room temperature (left) and high-resolution TEM images of the buckled NS-CNTs (right).

#### **Electret DEG energy considerations:**



 $V_0 = 1000 V$  (Electret surface potential)

 $C_1 = \varepsilon_0 \varepsilon' \frac{A}{d}$  capacity of the DE when stratched  $V'_1 = V'_2$  (Equilibrium of electric potential in the stretched state)  $Q_{total} = C_2 V_0$ (Total amount of charges on the electret surface)

$$Q_{total} = Q'_1 + Q'_2$$

$$V'_{1} = \frac{Q'_{1}}{C_{1}}$$

$$Q'_{1} = \frac{Q_{tot} C_{1}}{C_{1} + C_{2}}$$

$$E_{stretched} = \frac{1}{2}C_{1}V'_{1}^{2} + \frac{1}{2}C_{2}V'_{2}^{2} = \frac{1}{2}V'_{1}^{2}(C_{1} + C_{2})$$

$$E_{open} = \frac{1}{2}C_{2}V_{0}^{2}$$

$$\Delta E = E_{stretched} - E_{open} = \frac{1}{2}\left(\left((C_{1} + C_{2})V'_{1}^{2}\right) - (C_{2}V_{0}^{2})\right)\right)$$

$$= -\frac{1}{2}V_{0}^{2}\frac{C_{1}C_{2}}{C_{1} + C_{2}}$$

Theoretical values of  $\Delta E$  for an electret device where the DE is strained at maximum.

Entry	ε'	S <sub>max</sub>	A <sup>a</sup>	db	<b>C</b> <sub>1</sub>	Q'1	V'1	ΔE	Е
		[%]			[nF]	[µC]	[V]	[J]	[V/µm]
PDMS	2.9	372	5cm × 14.2cm 0.0071 m <sup>2</sup>	42.4	4.3	0.77	179.5	-3.86 10 <sup>-4</sup>	4.2
Elastosil	2.9	485	5cm × 17.6cm 0.0088 m <sup>2</sup>	34.2	6.6	0.99	150.3	-4.95 10 <sup>-4</sup>	4.4
PDMS-CN	3.5	313	5cm × 12.4cm 0.0062 m <sup>2</sup>	48.4	4.0	0.68	171.9	-3.41 10 <sup>-4</sup>	3.6
PDMS-CN-CNT	4.6	270	5cm × 11.1cm 0.0056 m <sup>2</sup>	54.1	4.2	0.62	149.9	-3.13 10 <sup>-4</sup>	2.8
P3-Cl	5.3	114	5cm × 6.4cm 0.0032 m <sup>2</sup>	93.5	1.6	0.34	209.2	-1.69 10 <sup>-4</sup>	2.2
P3-CI-CNT	6.3	95	5cm × 5.85cm 0.0029 m <sup>2</sup>	102.6	1.6	0.31	196.3	-1.56 10 <sup>-4</sup>	1.9

<sup>a</sup>the theoretical surface area of DE films (initial size of 5 cm × 3 cm) that were uniaxially stretched by  $s_{max}$ , <sup>b</sup>d= d<sub>0</sub>/ (1+ s<sub>max</sub>), where d represents the DE thickness after being stretched by  $s_{max}$ , C<sub>1</sub> represents the capacitance of the DE in the stretched state, Q'<sub>1</sub> is the charge on the stretched DE.  $\Delta E$  represents the theoretical amount of energy harvested if the material was stretched at maximum. E represents the theoretical electric field in the DE calculated using V1' and d.

$$C_{2} = \varepsilon_{0} \varepsilon' \frac{A}{d} \ (\epsilon' = 2.1, d_{Teflon} = 140 \ \mu m, A \ depends \ on \ the \ area \ of \ the \ DE)$$

$$V_2 = \frac{\left(Q_{total} - Q'_1\right)}{C_2}$$

Electret	Electret Area <sup>a</sup>	C <sub>2</sub>	<b>Q</b> <sub>total</sub>	V <sub>2</sub>
	[m <sup>2</sup> ]	[nF]	[μC]	[V]
PDMS	0.0071	0.94	0.94	179.8
Elastosil	0.0088	1.17	1.17	150.3
PDMS-CN	0.0062	0.82	0.82	171.9
PDMS-CN-CNT	0.0056	0.74	0.74	149.9
P3-Cl	0.0032	0.43	0.43	209.2
P3-CI-CNT	0.0029	0.39	0.39	196.3

<sup>a</sup>To be able to calculate the maximum energy when the DE materials is stretched at maximum, electrets with different surface area has to be used.

Entry	ε'	A <sup>a</sup>	db	C <sub>1</sub>	Q'1	V'1	ΔE	E
			[µm]	[nF]	[µC]	[V]	[1]	[V/µm]
PDMS	2.9	5cm × 4 cm	150.4	0.34	0.15	437.5	-7.48 10 <sup>-5</sup>	2.9
		0.002 m <sup>2</sup>						
Elastosil	2.9	0.002 m <sup>2</sup>	150.4	0.34	0.14	437.5	-7.48 10 <sup>-5</sup>	2.9
PDMS-CN	3.5	0.002 m <sup>2</sup>	150.4	0.41	0.16	391.9	-8.08 10 <sup>-5</sup>	2.6
PDMS-CN-CNT	4.6	0.002 m <sup>2</sup>	150.4	0.54	0.18	329.0	-8.92 10 <sup>-5</sup>	2.2
P3-Cl	5.3	0.002 m <sup>2</sup>	150.4	0.62	0.19	298.5	-9.32 10 <sup>-5</sup>	2.0
P3-CI-CNT	6.3	$0.002 \text{ m}^2$	150.4	0.74	0.20	263.6	-9.79 10 <sup>-5</sup>	1.8

Theoretical values of  $\Delta E$  when the DE is stretched by 33%.

<sup>a</sup>the surface area of the electret used and of the DE films (5 cm × 3 cm) that were uniaxially stretched by 33%.

$$V_2 = \frac{\left(Q_{total} - Q'_1\right)}{C_2}$$

Electret	C <sub>2</sub>	Q <sub>total</sub>	V <sub>2</sub>
	[nF]	[µC]	[V]
PDMS	0.27	0.27	437.5
Elastosil	0.27	0.27	437.5
PDMS-CN	0.27	0.27	391.9
PDMS-CN-CNT	0.27	0.27	329.0
P3-Cl	0.27	0.27	298.5
P3-CI-CNT	0.27	0.27	263.6

#### Theoretical values at 66% DEG strain

Entry	ε'	A <sup>a</sup>	db	<b>C</b> <sub>1</sub>	Q'1	V′1	ΔE	E
			[µm]	[nF]	[µC]	[V]	[1]	[V/µm]
PDMS	2.9	5cm × 5 cm 0.0025 m <sup>2</sup>	120.5	0.53	0.20	383.9 V	-1.02 10 <sup>-4</sup>	3.2
Elastosil	2.9	0.0025 m <sup>2</sup>	120.5	0.53	0.20	383.9 V	-1.02 10 <sup>-4</sup>	3.2
PDMS-CN	3.5	0.0025 m <sup>2</sup>	120.5	0.64	0.22	340.5 V	-1.10 10 <sup>-4</sup>	2.8
PDMS-CN-CNT	4.6	0.0025 m <sup>2</sup>	120.5	0.84	0.24	282.1 V	-1.19 10 <sup>-4</sup>	2.3
P3-Cl	5.3	0.0025 m <sup>2</sup>	120.5	0.97	0.25	254.3 V	-1.24 10 <sup>-4</sup>	2.1
P3-CI-CNT	6.3	0.0025 m <sup>2</sup>	120.5	1.15	0.26	222.9 V	-1.29 10 <sup>-4</sup>	1.9

<sup>a</sup>the surface area of the electret used and of the DE films (5 cm × 3 cm) that were uniaxially stretched by 66%.

$$V_2 = \frac{\left(Q_{total} - Q'_1\right)}{C_2}$$

Electret	C <sub>2</sub>	<b>Q</b> <sub>total</sub>	V <sub>2</sub>
	[nF]	[µC]	[V]
PDMS	0.33	0.33	383.9

Elastosil	0.33	0.33	383.9
PDMS-CN	0.33	0.33	340.5
PDMS-CN-CNT	0.33	0.33	282.1
P3-Cl	0.33	0.33	254.3
P3-CI-CNT	0.33	0.33	222.9