Supplementary Information

Synergetic effects in composite-based flexible hybrid mechanical energy harvesting generator

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Part 1

Table S1 Detailed composition of the generators.

Sample	**Sample contents**	**PDMS (A:B = 10:1) / ml**	**PVDF/g**	**MWCNT/g**	**$BaTiO_3$/g**	**Content[(a)] (wt%)**
S1	PDMS	5ml (6.35g)	0	0	0	-
S2	PVDF	5	1.5	0	0	19.1
S3	MWCNT	5	0	0.1	0	1.55
S4	$BaTiO_3$	5	0	0	1.2	15.9
S5	PVDF & MWCNT	5	1.5	0.1	0	20.12
S6	MWCNT & $BaTiO_3$	5	0	0.1	1.2	17
S7	PVDF & $BaTiO_3$	5	1.5	0	1.2	29.83
S8	PVDF & MWCNT & $BaTiO_3$	5	1.5	0.1	1.2	30.6

Note: (a) Content (wt%) = $\frac{Weight\ of\ added\ component}{Total\ weight\ of\ PDMS\ mixture\ solution}$

Part 2 Fabrication process of PDMS composite films.

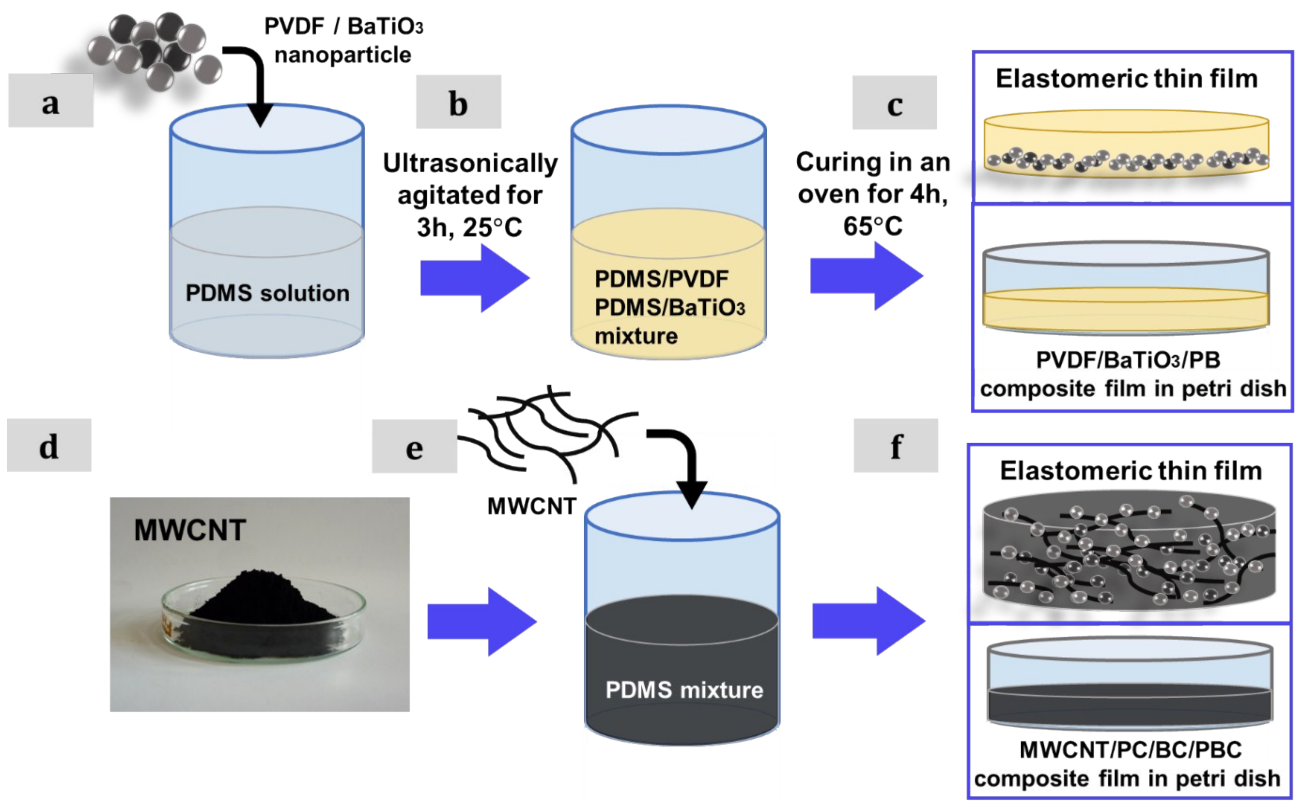

Fig. S1 Schematic diagrams of the process for fabricating PDMS composite films, (a), (d) Add the functional materials to PDMS solution. (b), (e) Agitate the PDMS composite ultrasonically for 3 hours at room temperature. (c), (f) Dry the as-mixed uniformly PDMS composite in an oven and cure for 4 hours at 65°C to form the elastomeric thin film (PCF).

Part 3 Polarization station set-up for poling of PCF containing piezoelectric materials.

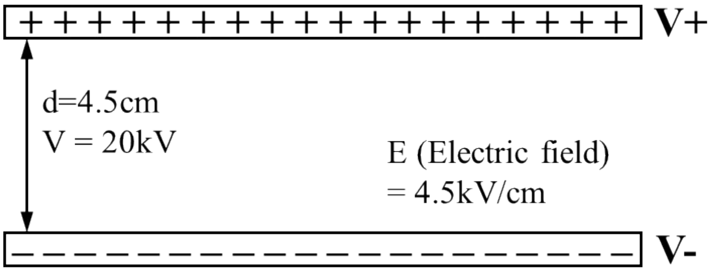

Fig. S2 Set up of the polarization station for poling the PCF containing piezoelectric materials

Part 4 Piezoelectric coefficient measurement

The piezoelectric coefficient (d_{33}) represents the ability of piezoelectric materials to convert mechanical deformation to electrical signal and plays a key role in the device performance. To verify the piezoelectric properties of PCF, we measured the piezoelectric coefficient (d_{33}) for each sample under a constant oscillation frequency of 190Hz using SS01 Piezo-d Meter (Sensor Tech. Ltd.). While many samples showed relatively high d_{33} larger than 100pC/N demonstrating that the piezoelectric composites possess a potential for generating high-power output, S1-PDMS and S3-CNT showed no d_{33} indicating no piezoelectric response.

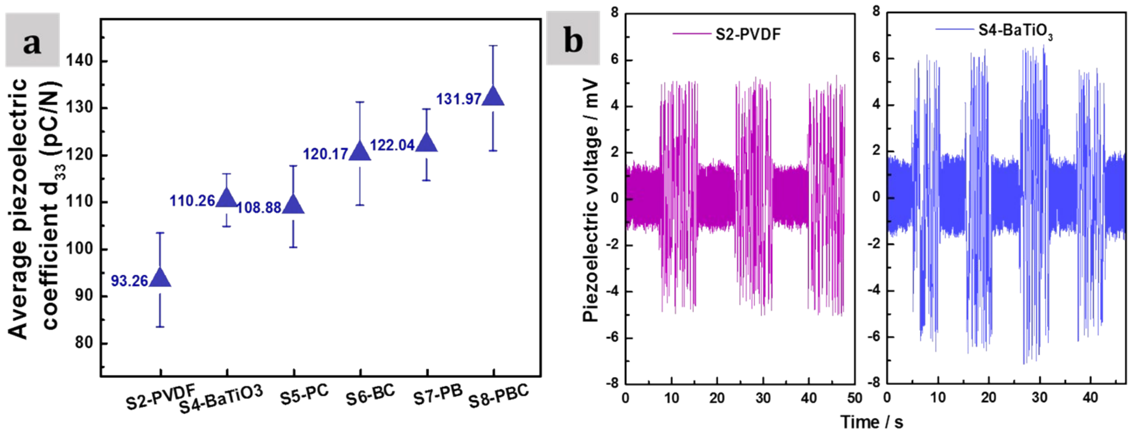

Fig. S3 (a) Average piezoelectric coefficient (d_{33}) measurements. (b) Piezoelectricity response of S2-PVDF (~5mV) and S4-$BaTiO_3$ (~7mV) under the constant oscillation with a frequency of 190Hz and a mechanical compression of 2N.

Part 5

Table S2 Calculation and derivation process of the composition ratio between triboelectric and piezoelectric potential for each generator.

Row		S1	S2	S3	S4	S5	S6	S7	S8
1	**Item** / **Sample contents**	**PDMS**	**PVDF**	**MWCNT**	**$BaTiO_3$**	**PVDF & MWCNT**	**$BaTiO_3$ & MWCNT**	**PVDF & $BaTiO_3$**	**PVDF & $BaTiO_3$& MWCNT**
2	**Total tribo-piezoelectric potential[(a)]/V (with spacer)**	15.23	16.67	17.17	19.26	23.94	25.77	21.33	48.46
3	**Type (with spacer)**	TEG	TPEG	TEG	TPEG	TPEG	TPEG	TPEG	TPEG
4	**Total piezoelectric potential / Non-piezo potential / V (without spacer)**	0.056[(b)]	2.78	1.78[(b)]	4.25	5.76	6.43	6.79	8.57
5	**Type (without spacer)**	/	PEG	/	PEG	PEG	PEG	PEG	PEG
6	**Net triboelectric potential of PCF[(c)]/V (excluding piezoelectric potential)**	15.23[(d)]	13.89	17.17[(d)]	15.01	18.18	19.34	14.54	39.89
7	**Net piezoelectric potential of individual and composite[(e)] (excluding PDMS)**	0[(f)]	2.724	0[(f)]	4.194	5.704	6.374	6.734	8.514
8	**Net triboelectric potential of individual and composite (V) (excluding PDMS)** [(g)]	15.23	-1.34	1.94	-0.22	2.95	4.11	-0.69	24.66
9	**Sum tribo-piezoelectric potential of PCF (S5-S8)** [(h)]	/	/	/	/	21.534	23.324	20.404	24.124
10	**T:P ratio[(i)]**	/	5.1:1	/	3.6:1	3.2:1	3:1	2.2:1	4.7:1

Supplementary Notes

(a) Total tribo-piezoelectric potential is the original output of all the generators based on PCF. Since some of the generators (S2, S4-S8) contain both triboelectric and piezoelectric materials, therefore, the hybrid output has combined tribo-piezoelectric potential.

(b) Since not all the materials possess piezoelectric property, e.g. S1-PDMS and S3-MWCNT, the generators containing these two materials have no piezoelectric output in absence of spacers. Both S1 (0.056V) and S3 (1.78V) registered an output during the piezoelectric potential measurement, though PDMS and MWCNT have no piezoelectric property. This small voltage is attributed to the triboelectric effect as explained in the discussion section due to a micro gap between Al and the PCF.

(c) Net triboelectric potential of PCF (Row 6) = Total tribo-piezoelectric potential (with spacer, Row 2) – Total piezoelectric potential (without spacer, Row 4).

(d) The net triboelectric potential of S1-PDMS and S3-CNT equal to their total tribo-piezoelectric potential, which are 15.23V and 17.17V, respectively. Because S1 and S3 do not have the capability of piezoelectricity generation, the generated potentials in absence of spacers (Row 4) are attributed to the presence of micro gap between the Al and the PCF as discussed in note (b).

(e) Net piezoelectric potential of individual and composite (excluding PDMS, Row 7) = Total piezoelectric potential (Without spacer, Row 4) – Output potential of PDMS without spacer (0.056V)

(f) Net piezoelectric potential of S1-PDMS and S3-CNT should be zero since they do not possess piezoelectric effect.

(g) Net triboelectric potential of individual material and composite (excluding PDMS, Row 8) = Net triboelectric potential of PCF (Row 6) – Triboelectric potential of S1-PDMS (15.23V). In this case, the net triboelectric of PDMS is 15.23V, however, under such assumption, the net triboelectric potential of individual PVDF, $BaTiO_3$ and composite PB achieved negative values of -1.34V, -0.22V and -0.69V, respectively. Here, the triboelectricity from all the PCF based generators cannot be generated independently without the PDMS substrate. Therefore, the negative values can only be used in the derivation process for analyzing the synergetic effects.

(h) Sum tribo-piezoelectric potential of S5-S8 (Row 9) = Sum of the net triboelectric potential of individual material (Row 8) + Triboelectric potential of PDMS (15.23V) + Net piezoelectric potential (Row 7)

For examples:

S5 (PC) = PDMS + PVDF + MWCNT + net piezo-potential of PC = 15.23 + (-1.34) + 1.94 + 5.704 = 21.534V

S6 (BC) = PDMS + $BaTiO_3$ + MWCNT + net piezo-potential of BC = 15.23 + (-0.22) + 1.94 + 6.374 = 23.324V

S7 (PB) = PDMS + PVDF + $BaTiO_3$ + net piezo-potential of PB = 15.23 + (-1.34) + (-0.22) + 6.734 =20.404V

S8 (PBC) = PDMS + PVDF + $BaTiO_3$ + MWCNT + net piezo-potential of PBC = 15.23 + (-1.34) + (-0.22) + 1.94 + 8.514 = 24.124V

(i) The T:P ratio for each generator was calculated including the potential of PDMS since all the generators were PDMS based and triboelectricity generation was also attributed to the presence of PDMS.

Part 6

Table S3 Internal resistance of TPEG.

	S1-PDMS	**S2-PVDF**	**S3-CNT**	**S4-$BaTiO_3$**	**S5-PC**	**S6-BC**	**S7-PB**	**S8-PBC**
Actual $R_{internal}$ = V_{oc}/I_{sc} (MΩ)	11.7	14.5	14.8	17.5	10.8	14.2	14.62	16.7
Approximate $R_{internal}$ (MΩ)	10.5	15	15	18	10.5	15	15	15

Part 7 Energy conversion efficiency and three-phase movement of linear motor.

The energy conversion efficiency of TPEG is defined as the ratio of the output electrical energy transferred to the external load and the input mechanical energy as given in the following equation.

$$\eta\% = \frac{Output\ electrical\ energy}{Input\ mechanical\ energy} = \frac{Energy\ delivered\ to\ external\ load\ resistance}{Work\ done\ on\ PCF\ by\ motor}$$

$$= \frac{E_R}{E_m} = \frac{\int I^2 R\, dt}{{}^1/_2 m v_1^2 - {}^1/_2 m v_0^2}$$

During the output test, the TPEG is subjected to two forces, the motor tapping force and its own gravity. However, the device only does work when the motor applies the reciprocal tapping where there's no work done on its gravity direction. The working mode of the motor is a process of first accelerating, travel at constant speed, and then decelerating, so called as "three-phase" movement of linear motor (Figure S3). When the motor probe approaches the movable PCF, it will be accelerated to a speed identical to the probe at last and thus converting kinetic energy to electrical energy. Therefore, we only consider the kinetic energy on the movable substrate stimulated by the tapping probe on the horizontal direction and ignore the work done on the gravity direction.

According to the equation, the average mass of the movable PCF is ~3.93g and the velocity when the two triboelectric substrates are about to come into contact is 1m/s as set by the motor controller. Therefore, the average input mechanical energy E_m equals to 1.965mJ. The trend of energy transferred to external resistance with different resistance values is demonstrated in Figure S2.

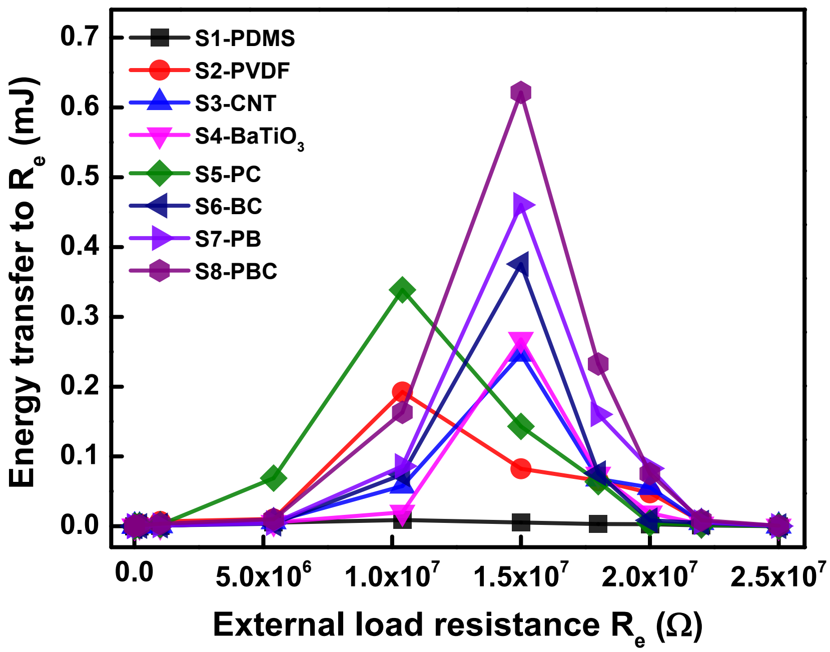

Fig. S4 Energy transferred to external load resistance with different load values.

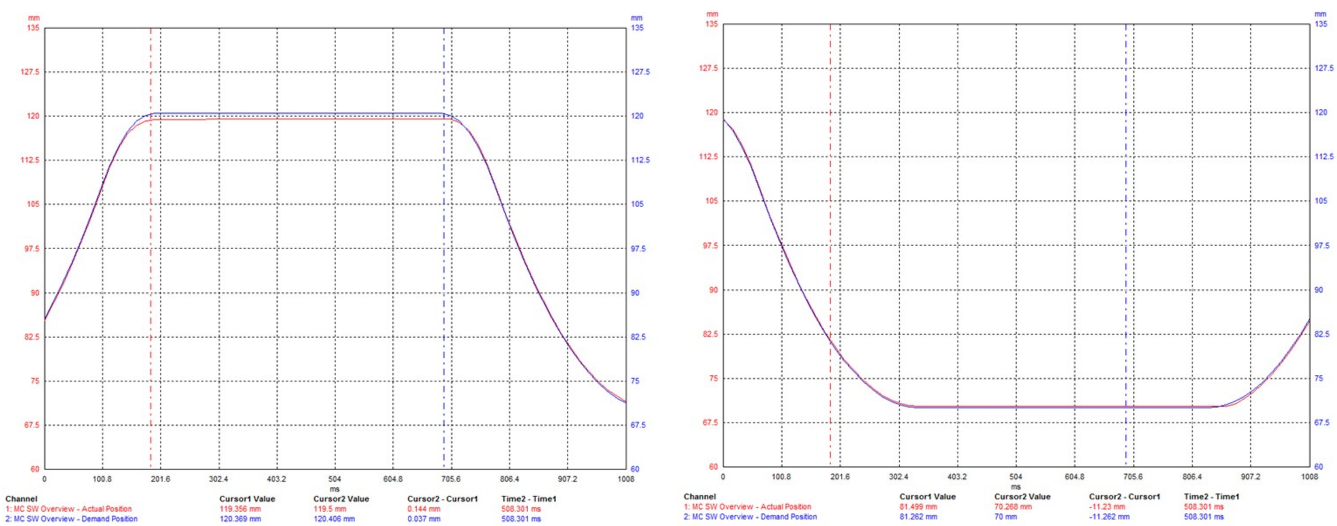

Fig. S5 Real-time relationship between the probe travel distance and time, "three-phase" movement of linear motor.