

## **Supplementary Materials**

### **Toward Commercialization of Mechanical Energy Harvester: Reusable Triboelectric Nanogenerator based on Closed-loop Mass production of Recyclable Thermoplastic Fluoropolymer with Microstructures**

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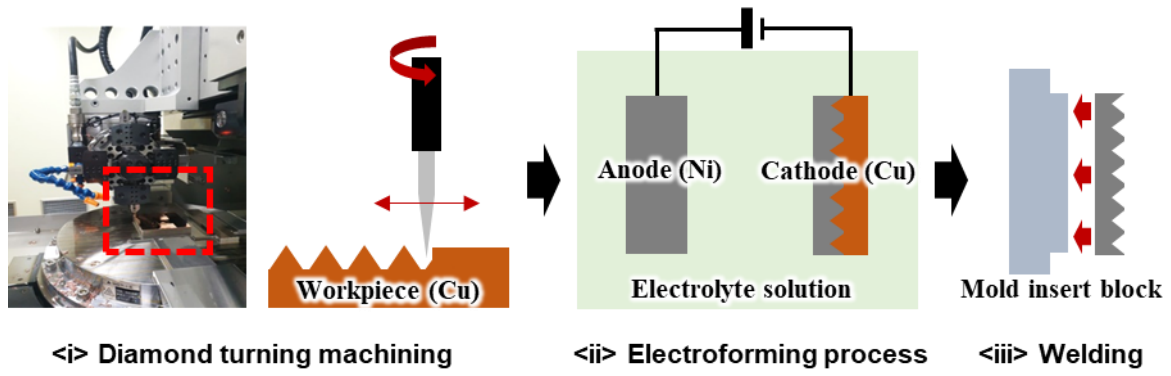
### Optimization of the condition of the injection molding process

To optimize the injection molding conditions for the replication quality of micro pyramid structures on the PFA contact layers, four parameters of mold temperature ( $T_M$ ), injection speed ( $S_I$ ), packing pressure ( $P_P$ ), and packing time ( $t_P$ ) were selected. The processing parameters and their levels are listed in Table S1. The experiments were conducted and analyzed by the Taguchi experimental method for finding out the significant order and the optimal conditions of processing parameters. And, based on the levels of the parameters, the  $L_9(3^4)$  orthogonal array was applied in the design of experiments listed in Table S2. The replication quality of the micro pyramid structures according to the injection molding conditions was evaluated based on the height of them (Figure S2(a)). The 3D laser confocal microscope was used to obtain 3D surface topographical details of the pyramid structures. The peak heights of the pyramid structures were automatically measured by the software. After the measuring of heights, the data obtained was analyzed by calculating the signal-to-noise ratio (S/N), representing the power of a response signal over the variation in the signal due to unwanted noise. This optimization of the injection molding parameter was considered as a static problem with larger-the-better S/N ratio, which were expressed as Equation (S1).

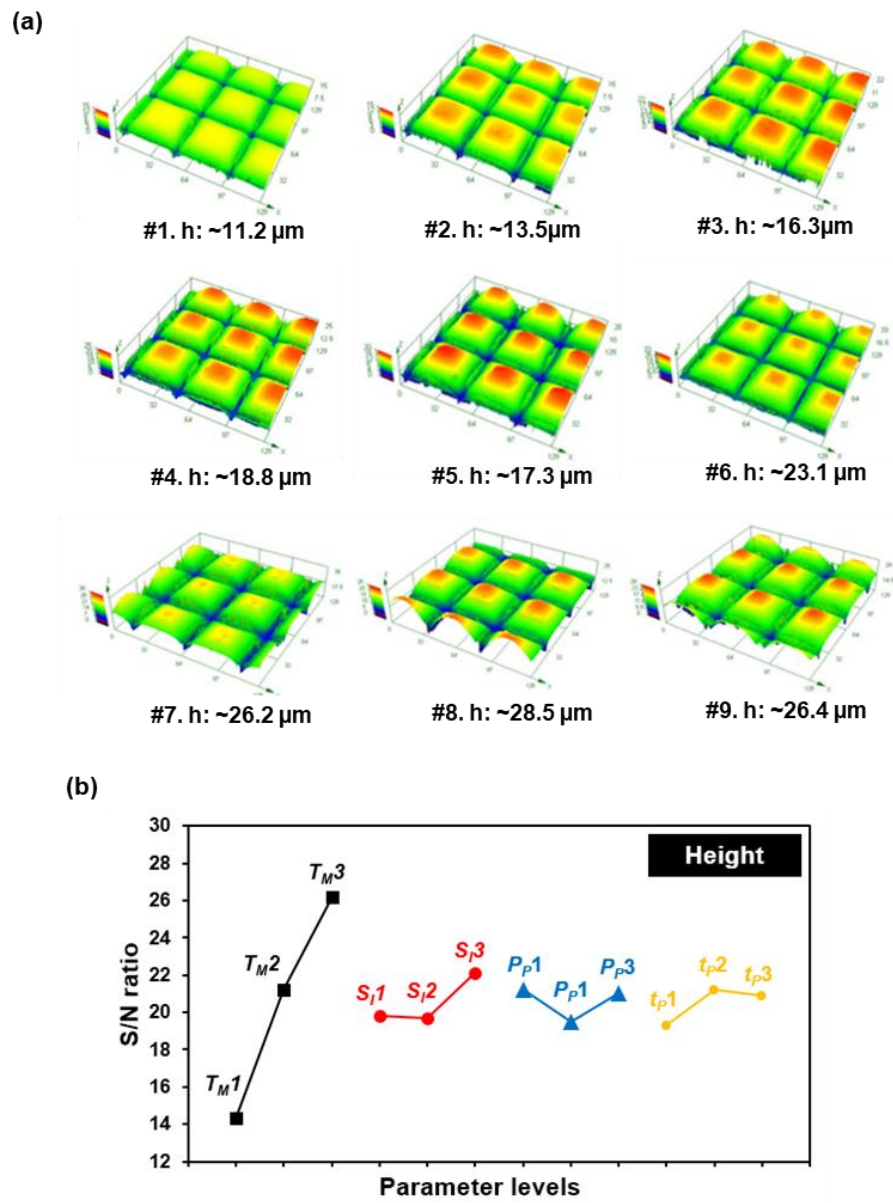
$$S/N_{Larger} = -10 \log \left[ \frac{1}{n} \sum \frac{1}{y_i^2} \right] \quad (S1)$$

where  $y_i$  is the measured value and  $n$  is the number of repetitions being tested in each test run. The optimum factor and levels can then be known by plotting the S/N against factor levels. Figure S2(b) shows signal to noise (S/N) ratios according to the applied processing parameters of injection molding. From the S/N ratios analysis, the significant order of processing parameters became mold temperature, injection speed. The results indicated that there were positive effects of higher mold temperature and injection speed. Based on the proper processing window derived from the previous experiment, the next experiments were performed to find

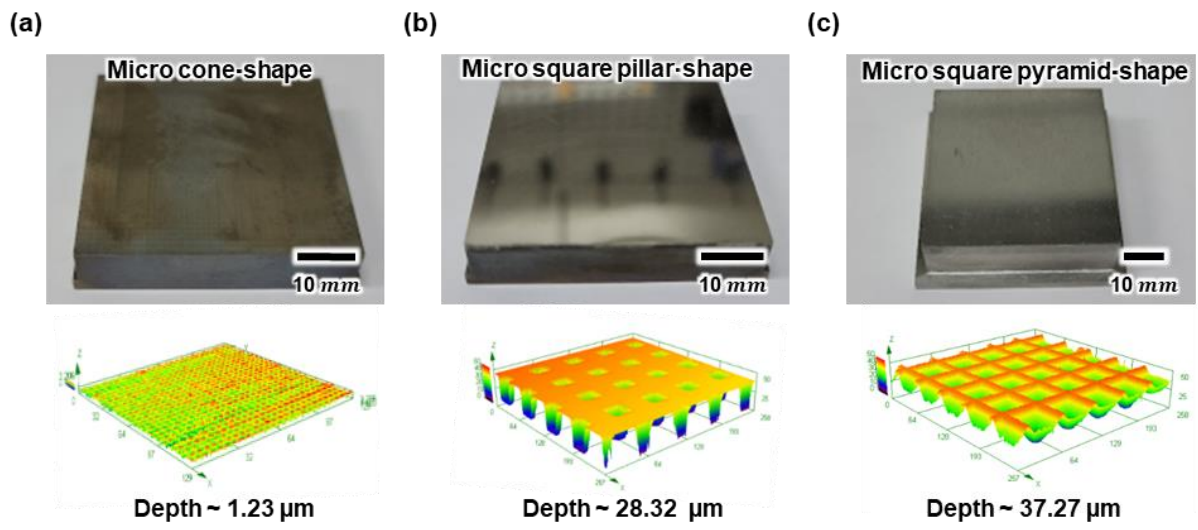
optimal conditions by changing the injection speed in the range 6 – 50 mm/s. Therefore, the optimal condition was determined as mold temperature of 190 °C, injection speed of 50 mm/s, holding pressure of 130 MPa, and holding time of 1 s.



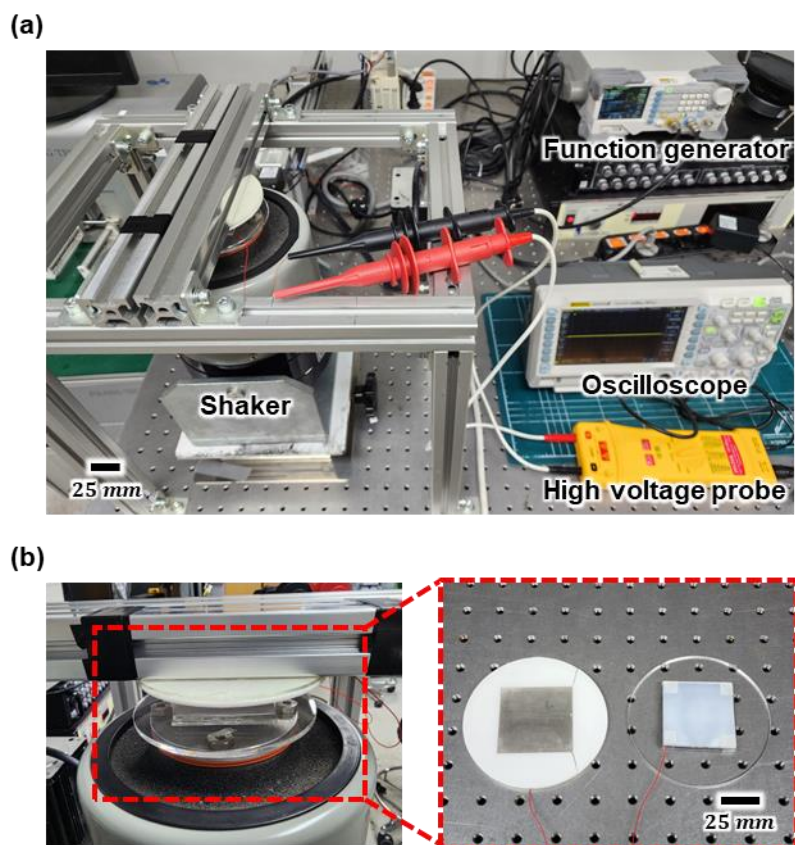
**Figure S1.** Diamond turning machining (DTM) and electroforming process for fabricating the mold insert block.



**Figure S2.** (a) Change of the surface topography of the injection molded PFA contact layers and (b) S/N ratio in the optimization of the injection molding process.

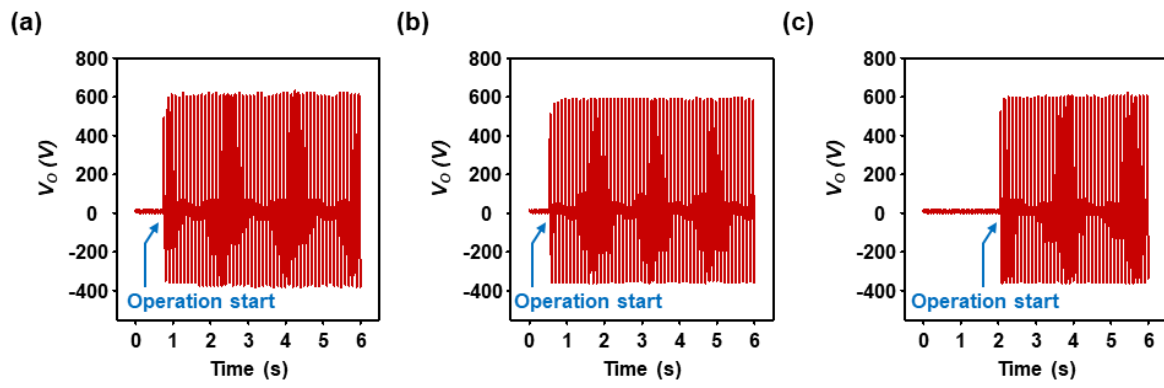


**Figure S3.** Photographs and surface topographies of the mold inserts with three types of negative surface structures of (a) micro cone-shape, (b) micro square pillar-shape, and (c) micro square pyramid-shape.

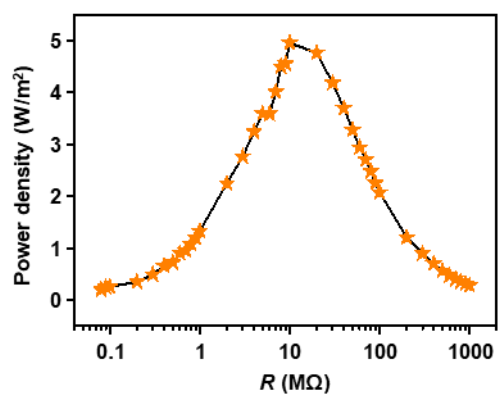


**Figure S4.** (a) Photograph of experimental setup for operation of S-S TENG. (b) S-S TENG mounted on the shaker. The inset is photograph of the PFA contact layer and APTES counter layer with aluminum electrodes.

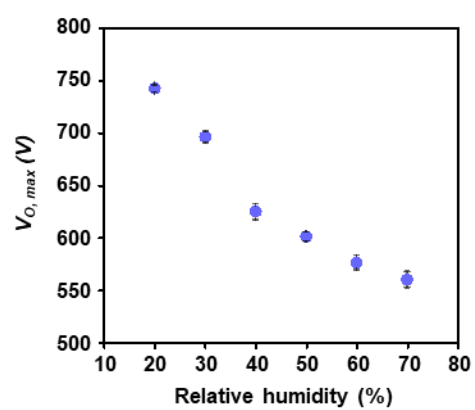




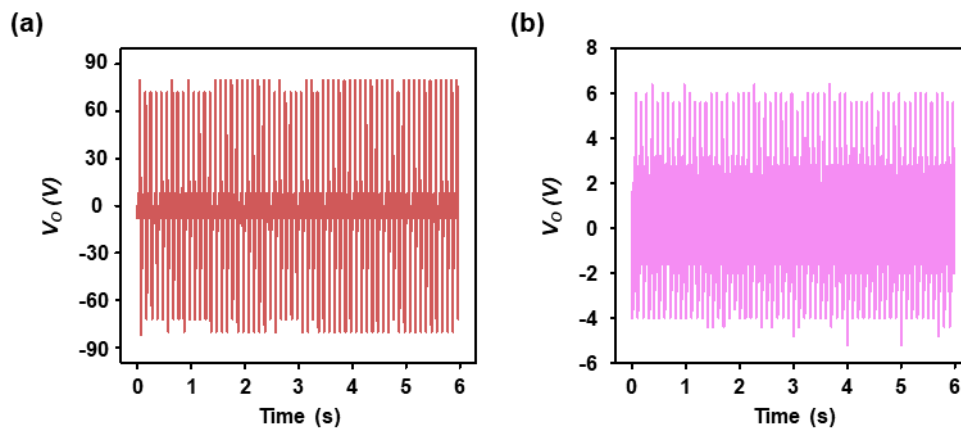
**Figure S5.** Gradual increase and saturation of electrical output of S-S TENG with its continuous operation.



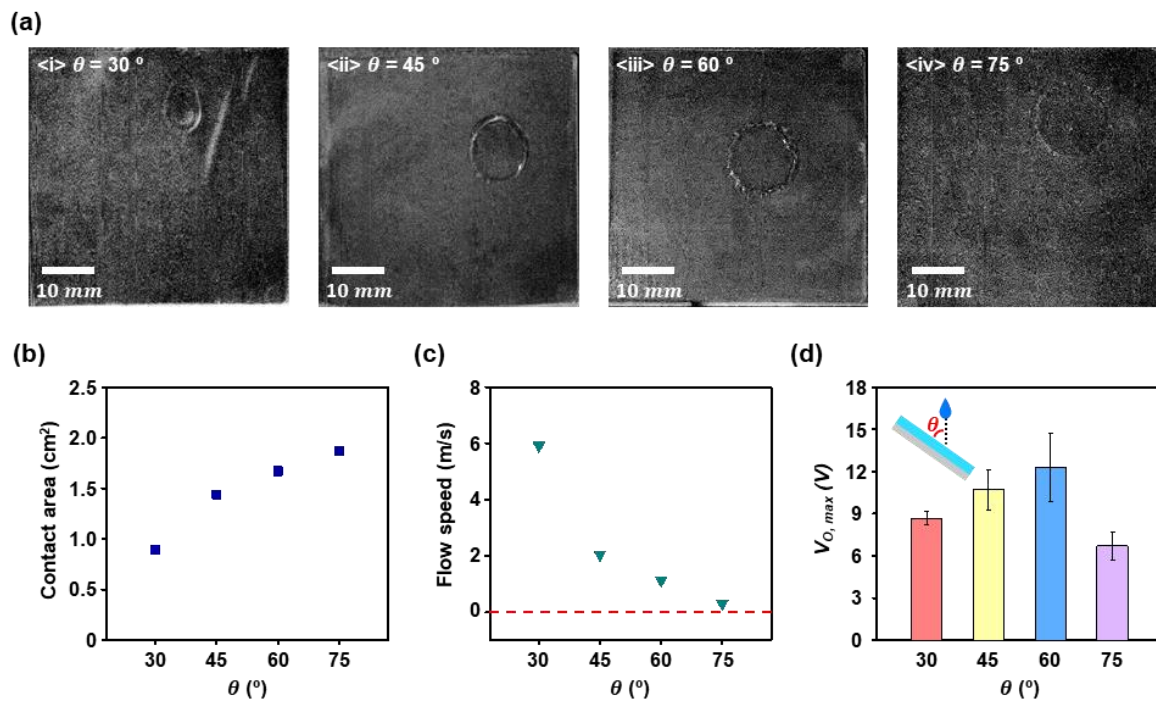
**Figure S6.** Power density generated by S-S TENG by varying the external load resistance.



**Figure S7.** Electrical output performance of S-S TENG with the injection-molded PFA contact layer according to the relative humidity.



**Figure S8.** Output voltage of S-S TENG when the (a) PLA and (b) PVA sheets are used as the contact layer.



**Figure S9.** (a) Photographs captured by the high-speed camera of the moments that a droplet and tilted PFA contact layer come into contact when  $\theta$  is (i) 30, (ii) 45, (iii) 60, and (iv) 75°. (b) Contact area, (c) flow speed, and (d) electrical output of L-S TENG depending on  $\theta$ .

**Table S1.** Parameter design for injection molding.

<b>Parameters</b>	<b>Level</b>			<b>Unit</b>
	1	2	3	
Mold temperature ( $T_M$ )	130	160	190	°C
Injection speed ( $S_I$ )	4	5	6	mm/s
Packing pressure ( $P_P$ )	70	100	130	MPa
Packing time ( $t_P$ )	0.5	1	2	sec

**Table S2.** Design of experiments (Orthogonal arrays of  $L_9 3^4$ ).

<b>Exp. #</b>	<b><math>T_M</math></b>	<b><math>S_I</math></b>	<b><math>P_P</math></b>	<b><math>t_P</math></b>
<b>1</b>	1	1	1	1
<b>2</b>	1	2	2	2
<b>3</b>	1	3	3	3
<b>4</b>	2	1	2	3
<b>5</b>	2	2	3	1
<b>6</b>	2	3	1	2
<b>7</b>	3	1	3	2
<b>8</b>	3	2	1	3
<b>9</b>	3	3	2	1

**Table S3.** Processing conditions applied to the injection molding process for the replication of PFA contact layers.

<b>Processing parameter</b>	<b>PFA contact layers</b>
Melt temperature (°C)	360
Mold temperature (°C)	130
Injection speed (mm s <sup>-1</sup> )	30
Injection pressure (MPa)	120
Packing pressure (MPa)	130
Packing time (s)	1
Cooling time (s)	30



**Table S4.** Cost of the injection molding process for manufacturing the of PFA contact layers.

<b>Category</b>	<b>\$/10,000ea</b>	<b>Category</b>	<b>\$/Month</b>
Injection molding	5,000	PFA contact layer (17,300 ea/Month)	16,600
Mold insert fabrication	4,000	Labor cost	3500
Material (PFA)	600	Incidental costs (Electricity usage, etc.)	900
Total	9,600	Monthly total	21,000