

Electronic Supplementary Material (ESI) for Energy & Environmental Science.  
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### **Electronic Supplementary Information (ESI)**

Large Piezoelectricity and high Power Density of 3D Printed-Multilayer  
Copolymer in Rugby Ball Structured Mechanical Energy Harvester

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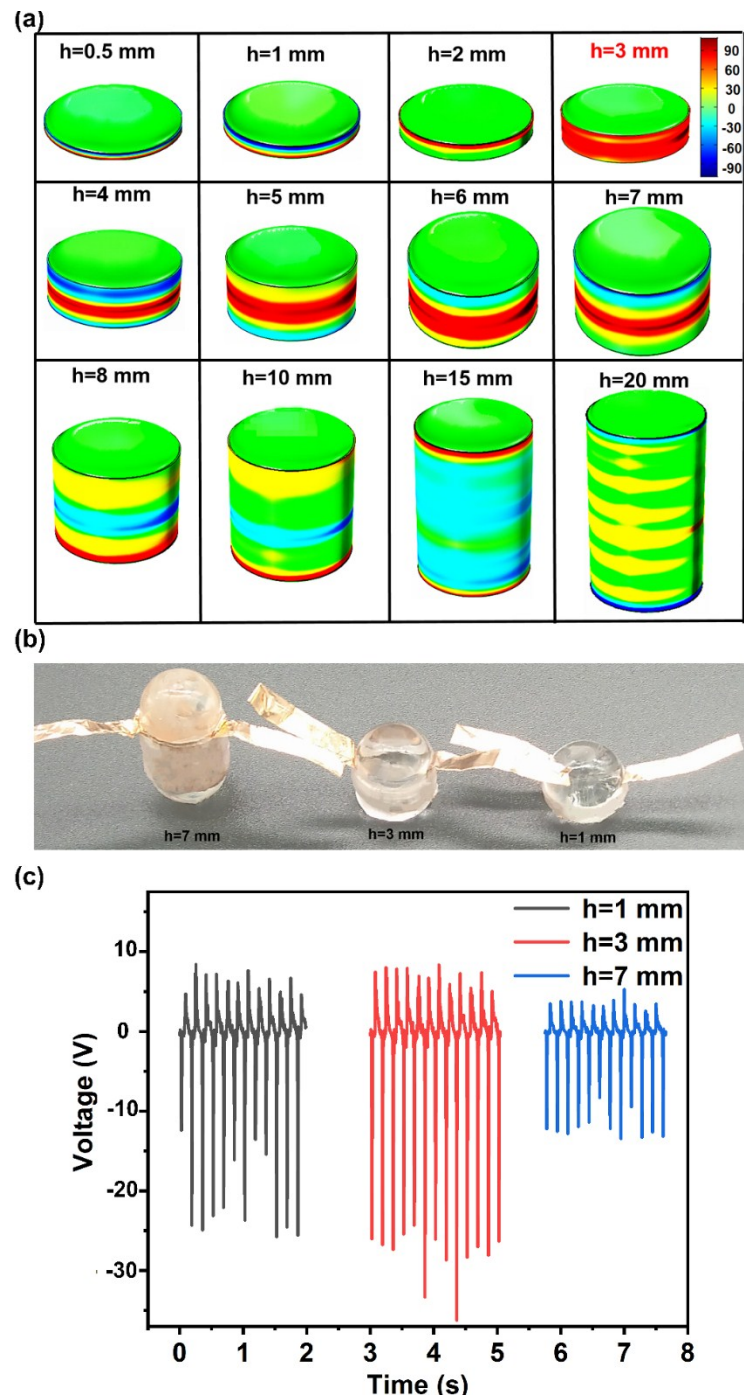
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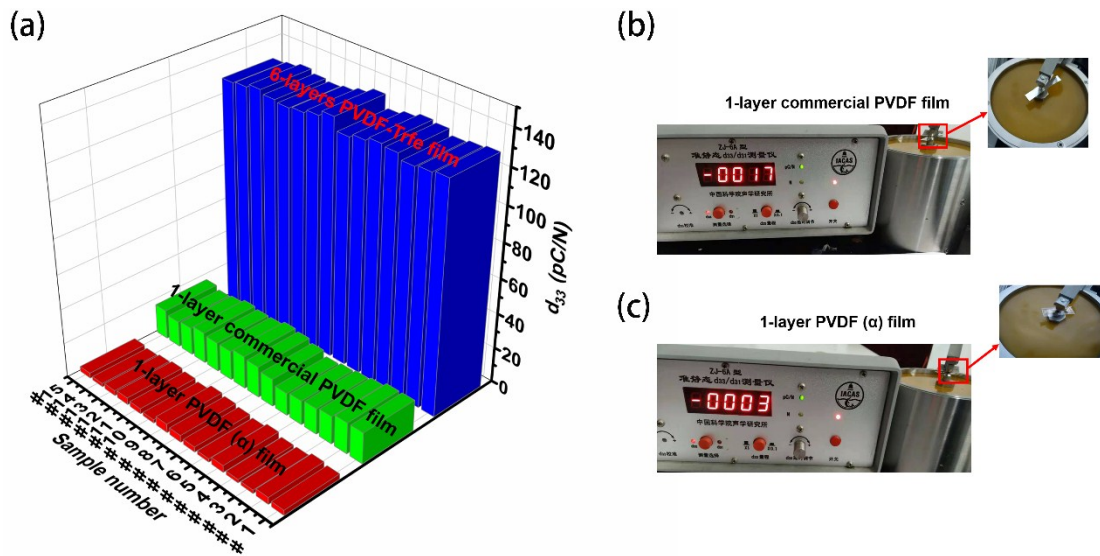
1. The effect of structure parameter height on output voltage in the rugby ball structure



**Figure S1.** (a) The FEM simulated output voltage from the rugby ball structured multilayer PVDF-TrFE energy harvester with different height  $h$  under a given load. (b) The photograph of three rugby ball structured multilayer PVDF-TrFE energy harvester with  $h = 1, 3,$  and  $7$  mm, respectively. (c) The output voltages of the three rugby ball structured energy harvesters under a dynamic pressing of  $0.046$  MPa at  $6$  Hz.

It is found that structure parameter height  $h$  of the rugby ball also influences on stress distribution on PVDF-TrFE belt, therefore, affects its output voltage. An optimum belt width  $h$  ( $h = 3$  mm for a rugby ball with  $D = 10$  mm, see Fig. 1a in text) can be found according to FEM simulation, as shown in Fig. S1 (a). A rugby ball with the optimum  $h$  will produce an uniform potential distribution with high voltage from the PVDF-TrFE belt. From Fig. S1, it can be seen that as  $h$  decreases, the high potential will mainly occur at one end of the belt, while as  $h$  increases, the electric potential distribution tends to disperse and also decrease dramatically. Fig. S1 (b) shows the photograph of three rugby ball structured multilayer PVDF-TrFE energy harvester with  $h = 1, 3,$  and  $7$  mm, respectively. The output voltages of the rugby ball energy harvesters with height  $h = 7$  mm,  $3$  mm,  $1$  mm under a dynamic pressing of  $0.046$  MPa at  $6$  Hz are  $32$  V,  $35$  V and  $17$  V, respectively, which are well consistent with the simulation results.

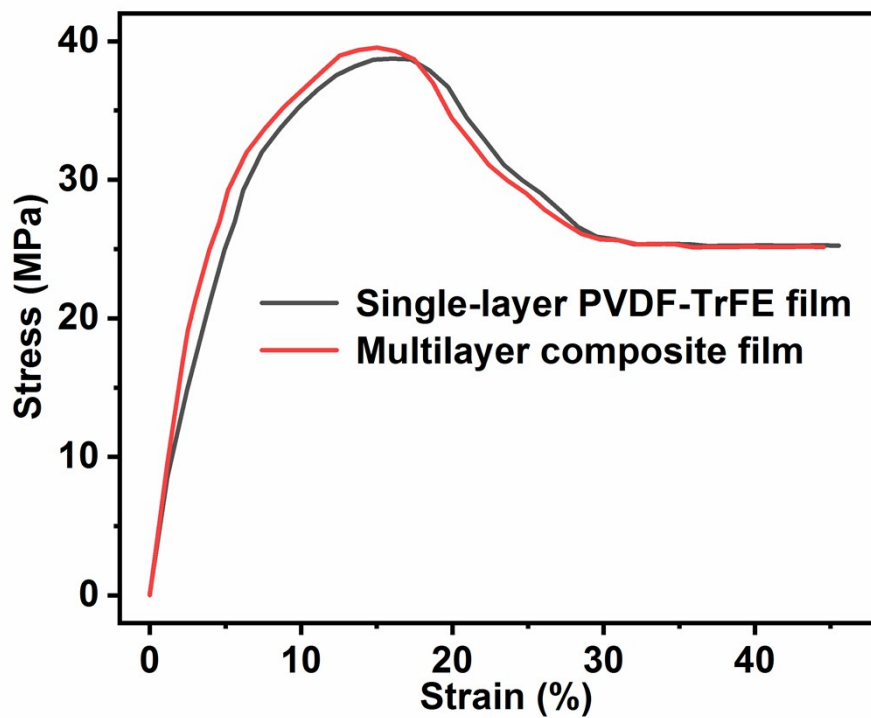
## 2. Piezoelectric coefficient $d_{33}$ of $\alpha$ -PVDF and commercial PVDF film



**Figure S2.** The measured  $d_{33}$  values from 15 samples of 1-layer- $\alpha$ -PVDF film, 1-layer commercial PVDF film and 6-layers PVDF-TrFE film

Fifteen samples of 1-layer  $\alpha$ -PVDF film, 1-layer commercial PVDF film and 6-layers PVDF-TrFE film were tested separately. The average piezoelectric coefficient  $d_{33}$  are 3 pC N<sup>-1</sup>, 17 pC N<sup>-1</sup> and 130 pC N<sup>-1</sup>, respectively.

### 3. Tensile strength of a single PVDF-TrFE film and multilayer composite film



**Figure S3.** Function between stress and strain of single-layer PVDF-TrFE film and multilayer composite film

Fig. S3 shows the tensile stress and strain (tensile strength) relationships for a single layer and the multilayer composite film, and their tensile strength properties are very similar. Although the hardness of the multilayer composite film slightly increased, it does not affect the flexibility of the composite film.

4. The photograph of device test setup

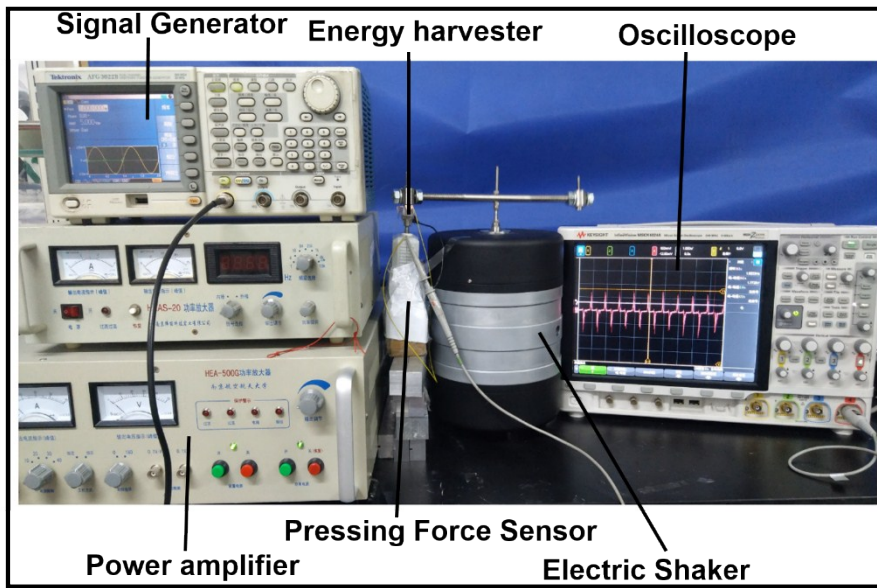
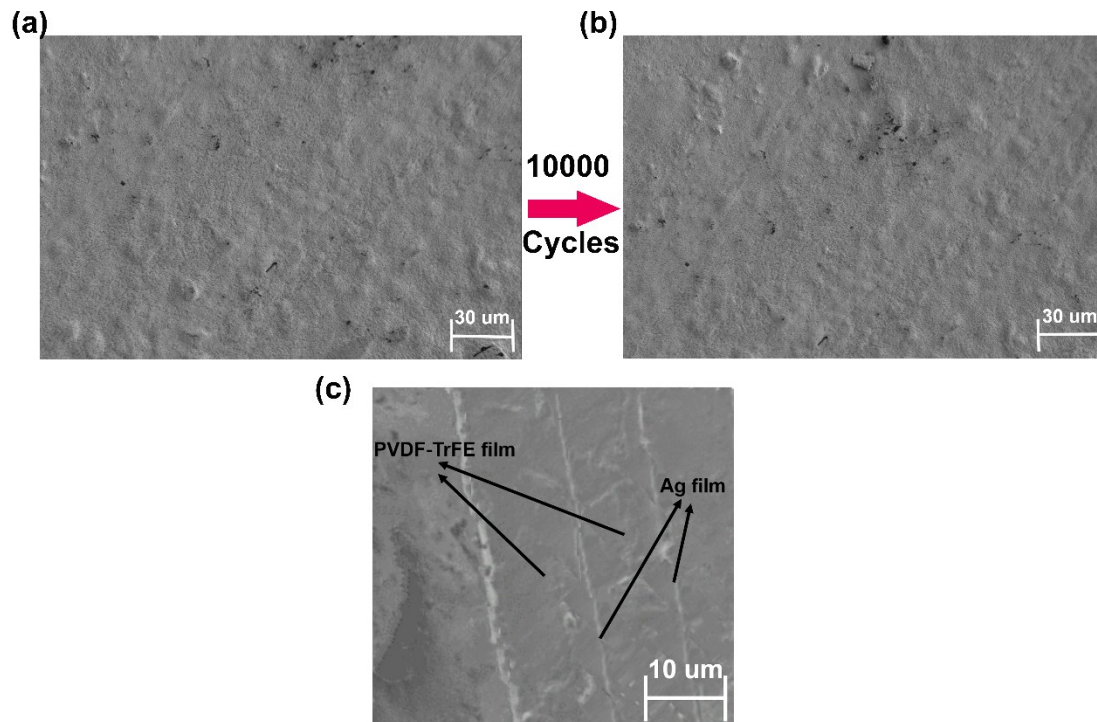


Figure S4. The photograph of test

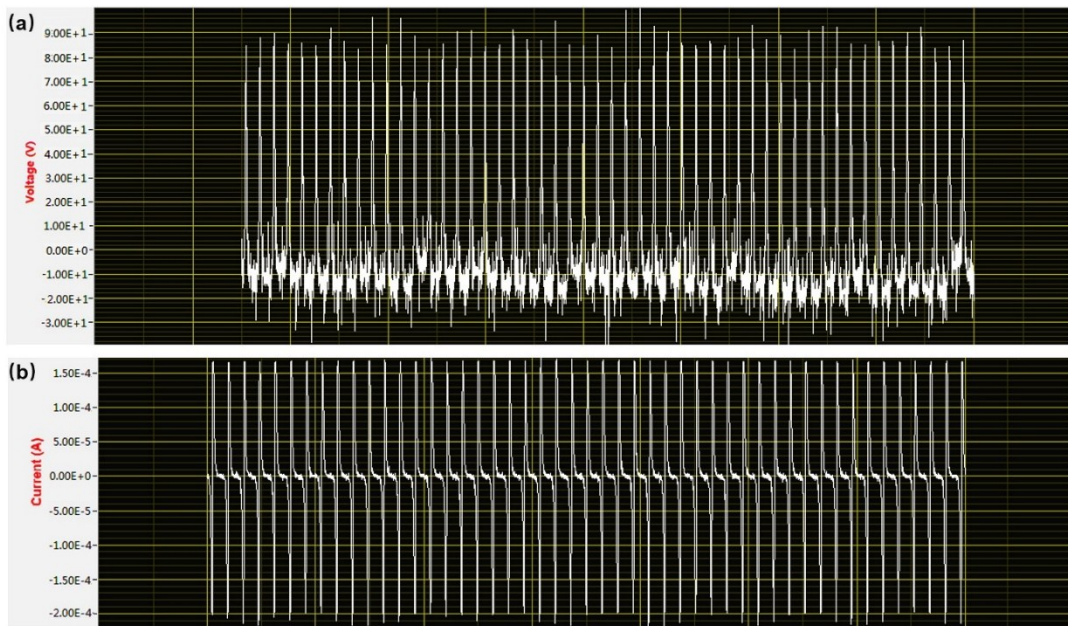
## 5. Mechanical integrity of Ag film and multilayer composite film



**Figure S5.** (a)-(b) The surface of original Ag film and Ag film after 10,000 cycle. (c) The cross section of composite film after 10000 cycles.

Fig. S5 (a) and (b) respectively show SEM photos of the original Ag electrode film and Ag film after 10,000 vibration cycles. It can be seen that the Ag electrode film remains mechanical integrity after 10000 vibration cycles. Fig. S5 (c) presents the cross section of the multilayer composite film after 10,000 vibration cycles. It can be seen that there is no damage or cracking found between Ag films and layers of PVDF-TrFE after 10000 cycles, indicating the rugby ball structure design of piezoelectric energy harvester is rational and reliable.

## 6. Measure $V_{peak}$ and $I_{peak}$



**Figure S6.** The open voltage and short current of the rugby ball structured energy harvesters under a dynamic pressing force of 0.046 MPa at 3.5 Hz

The  $V_{peak}$  and  $I_{peak}$  were measured simultaneously from one rugby ball piezoelectric energy harvester under mechanical vibration exciting. The output signal from the sample was measured by an electrometer (Keithley 6514).