

Supplementary Information

Super elastic and negative triboelectric polymer matrix for high performance mechanoluminescent platforms

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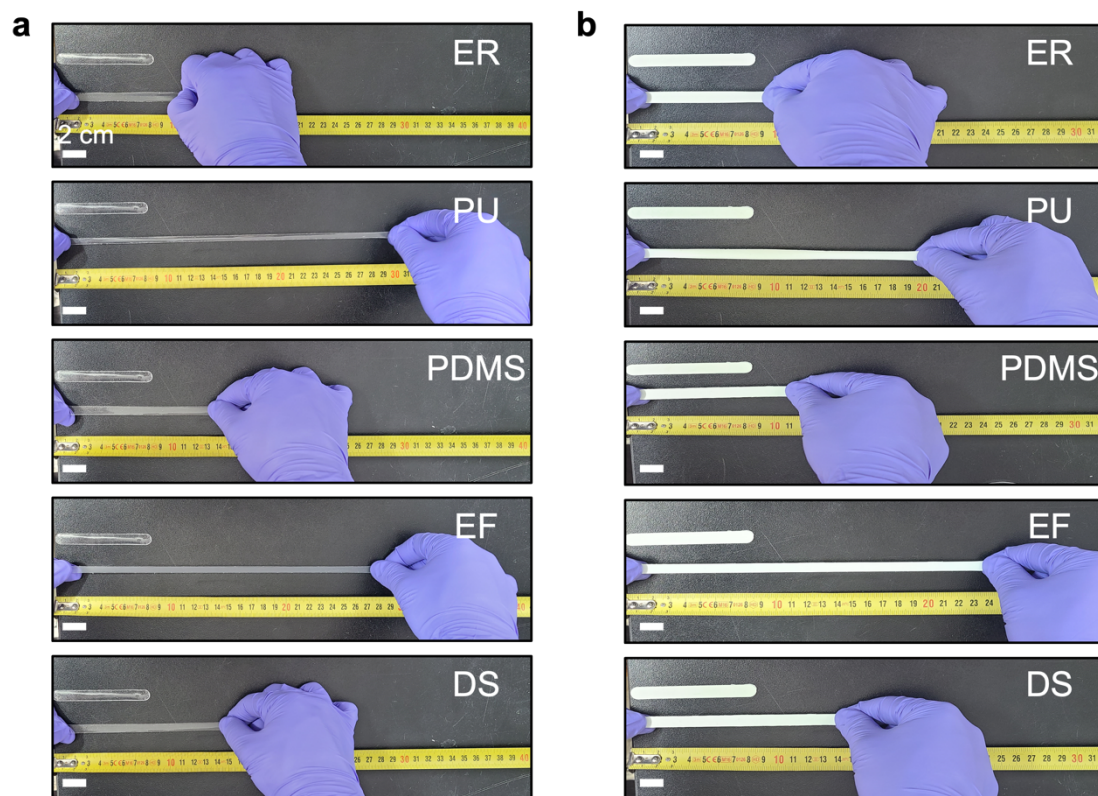
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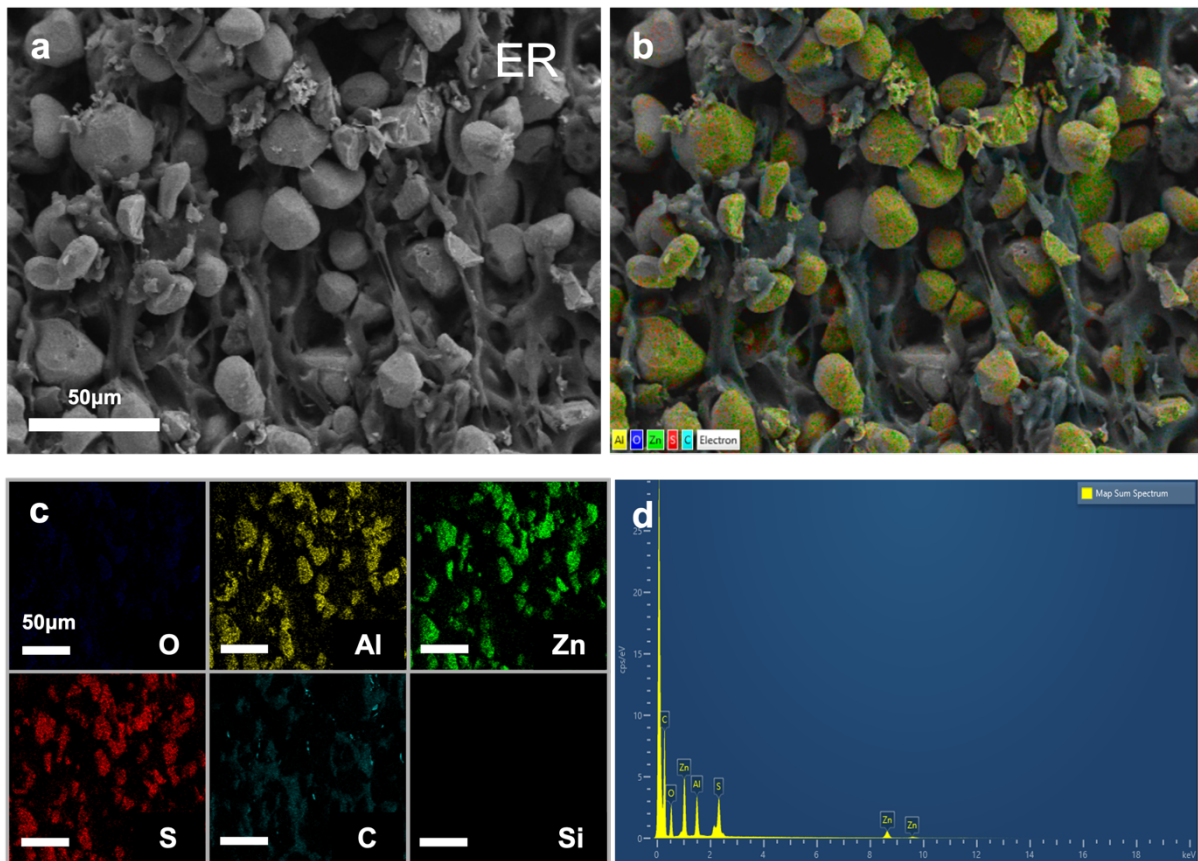
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Supplementary Fig. 1 | Durability test of polymer matrices

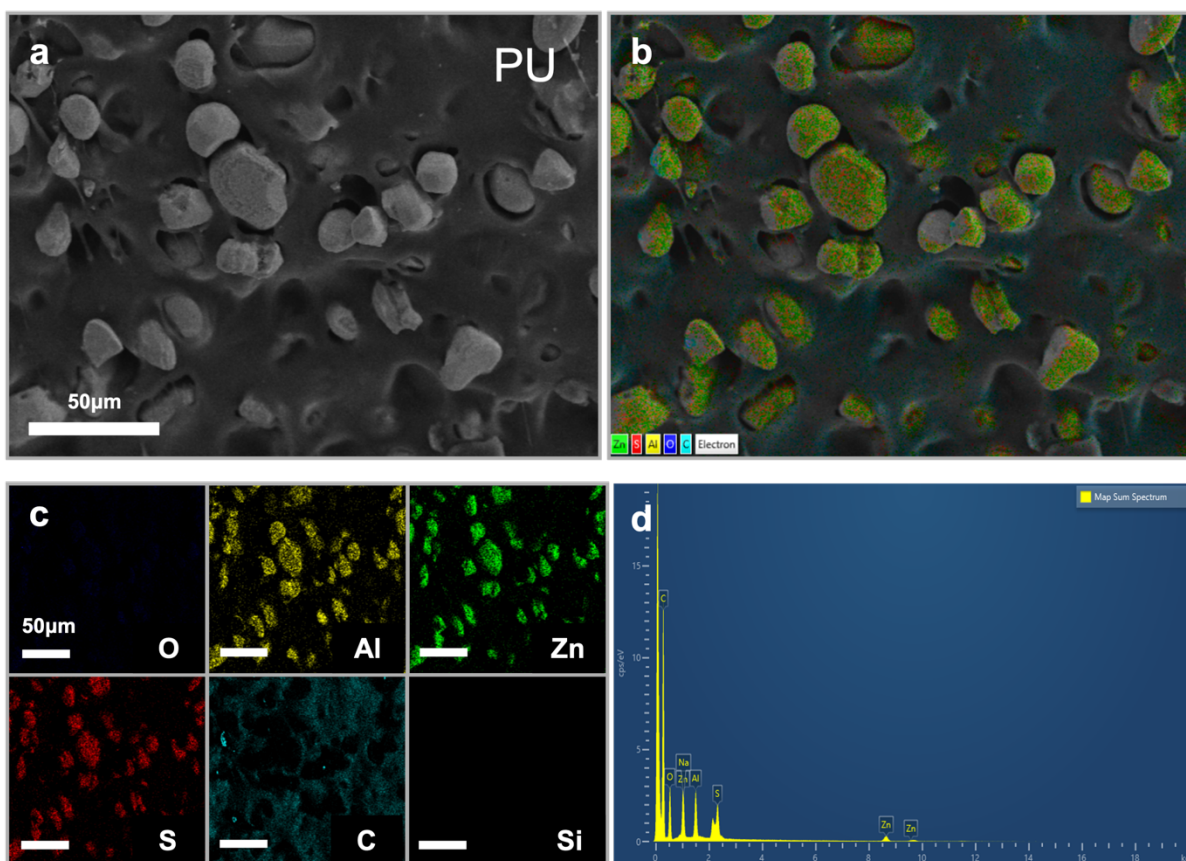
Photos of the durability test for the polymer matrix **a** without phosphor and **b** with phosphor.

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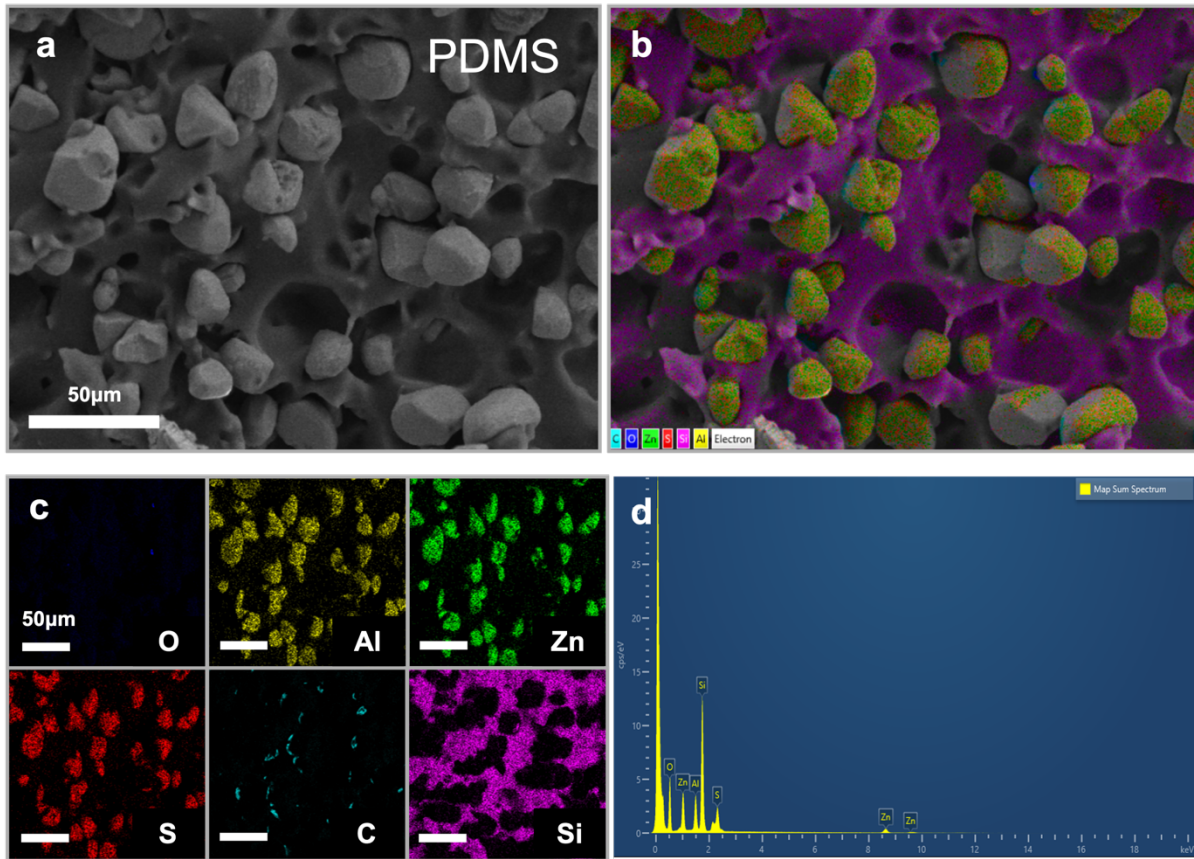
Supplementary Fig. 2 | Morphological properties of the ML platform with ER

a, b SEM and **c, d** EDX elements (O, Al, Zn, S, C, Si) mapping results of the ML platform with ER.



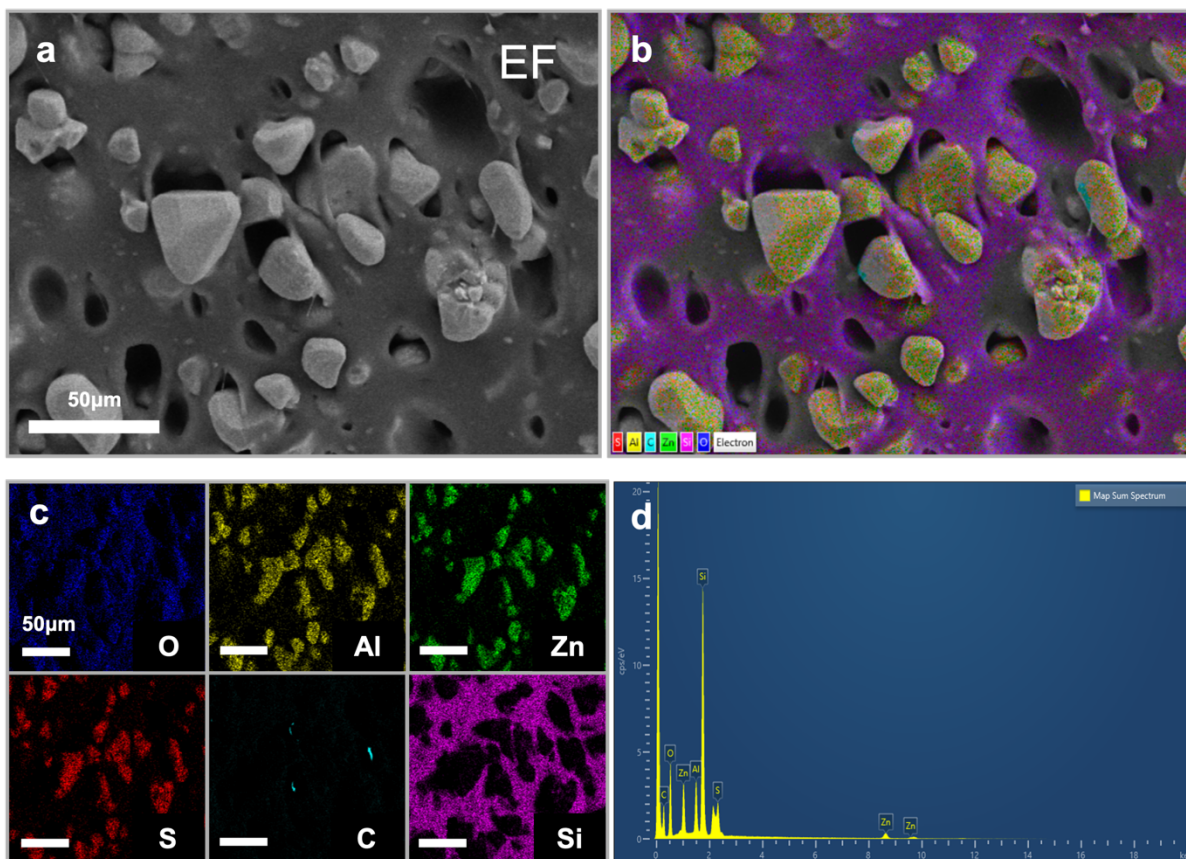
Supplementary Fig. 3 | Morphological properties of the ML platform with PU

a, b SEM and **c, d** EDX elements (O, Al, Zn, S, C, Si) mapping results of the ML platform with PU.



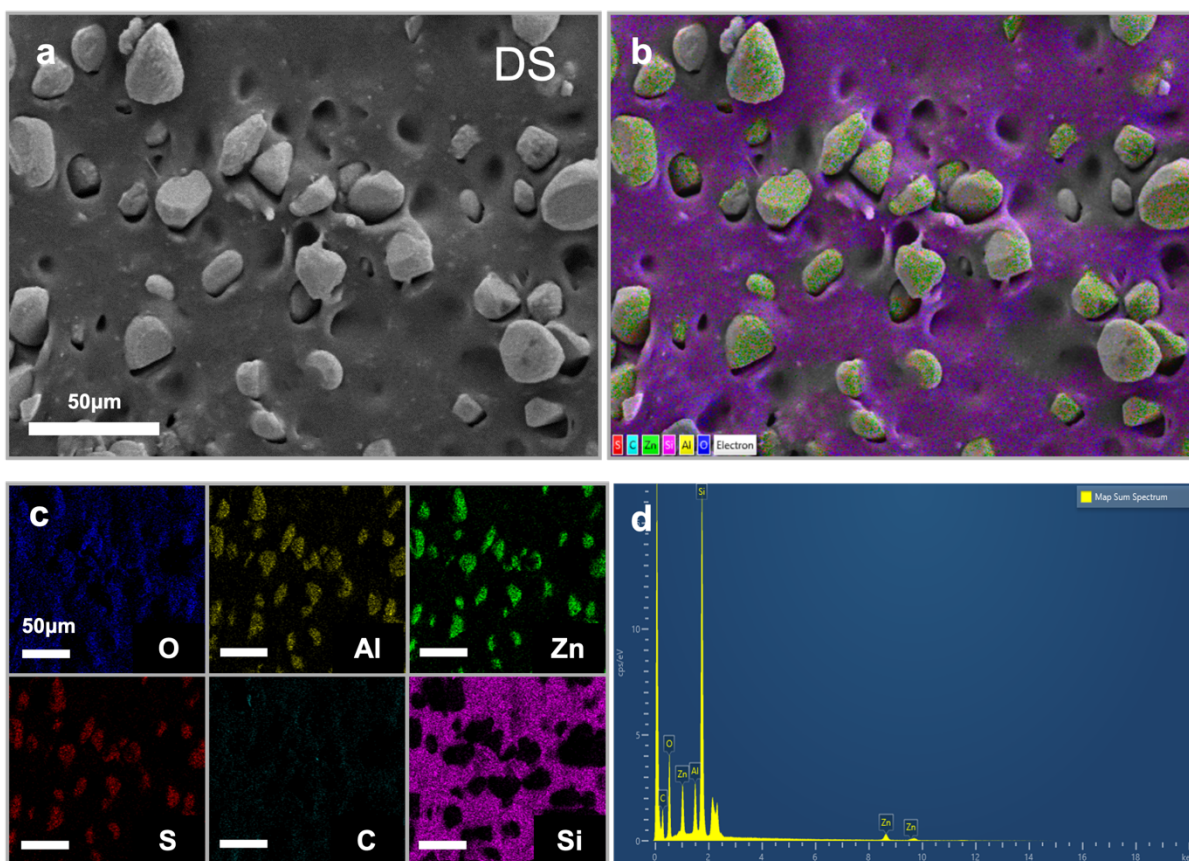
Supplementary Fig. 4 | Morphological properties of the ML platform with PDMS

a, b SEM and **c, d** EDX elements (O, Al, Zn, S, C, Si) mapping results of the ML platform with PDMS.



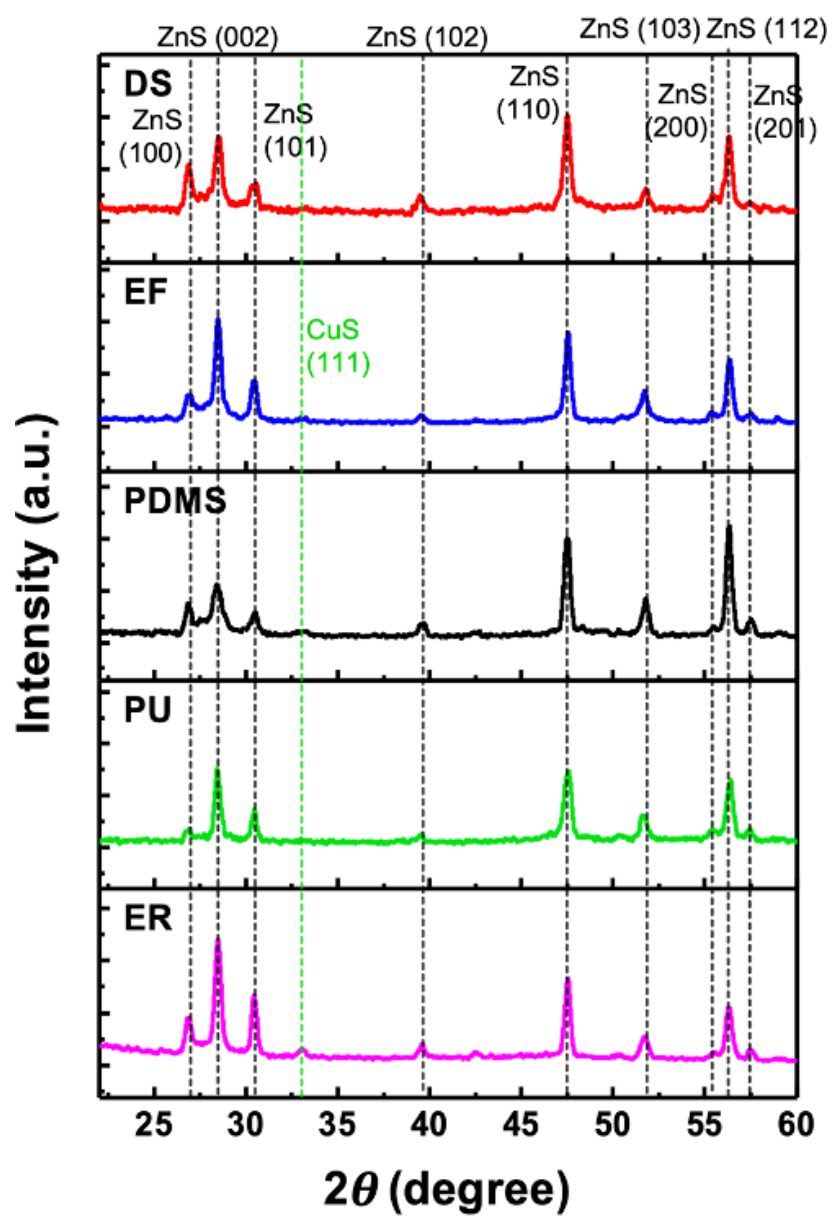
Supplementary Fig. 5 | Morphological properties of the ML platform with EF

a, b SEM and **c, d** EDX elements (O, Al, Zn, S, C, Si) mapping results of the ML platform with EF.

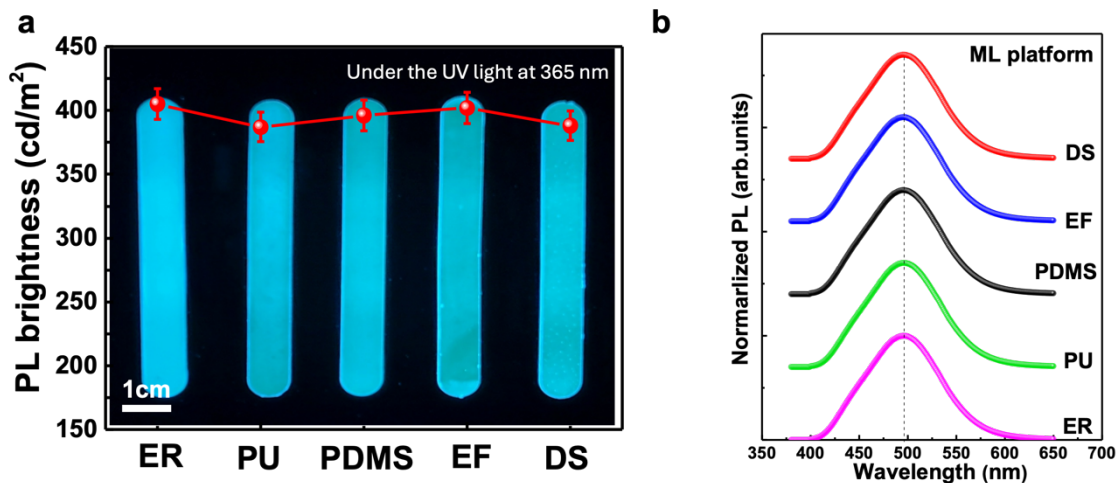


Supplementary Fig. 6 | Morphological properties of the ML platform with DS

a, b SEM and **c, d** EDX elements (O, Al, Zn, S, C, Si) mapping results of the ML platform with DS.

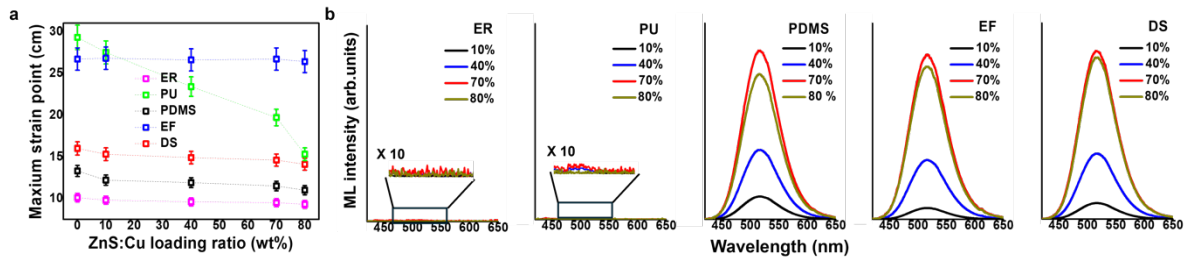


Supplementary Fig. 7 | Structural property of ML platform for various polymer matrices
XRD spectra of the ML platform with ER, PU, PDMS, EF and DS.



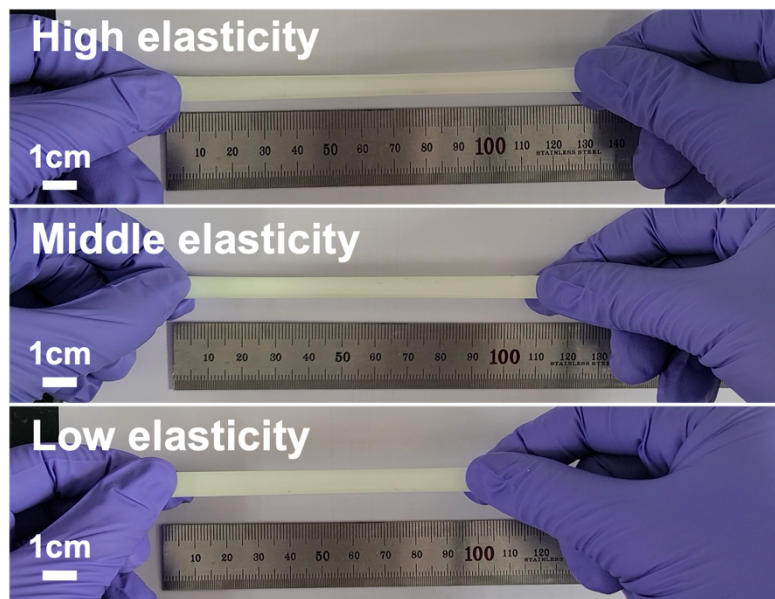
Supplementary Fig. 8 | Optical properties of ML platform for various polymer matrices

a PL brightness and **b** PL spectra of ML platform according to polymer matrix. Inset image in **a** shows the actual PL photo under the UV irradiation at 365 nm. Error bars in **a** represent 5% error.



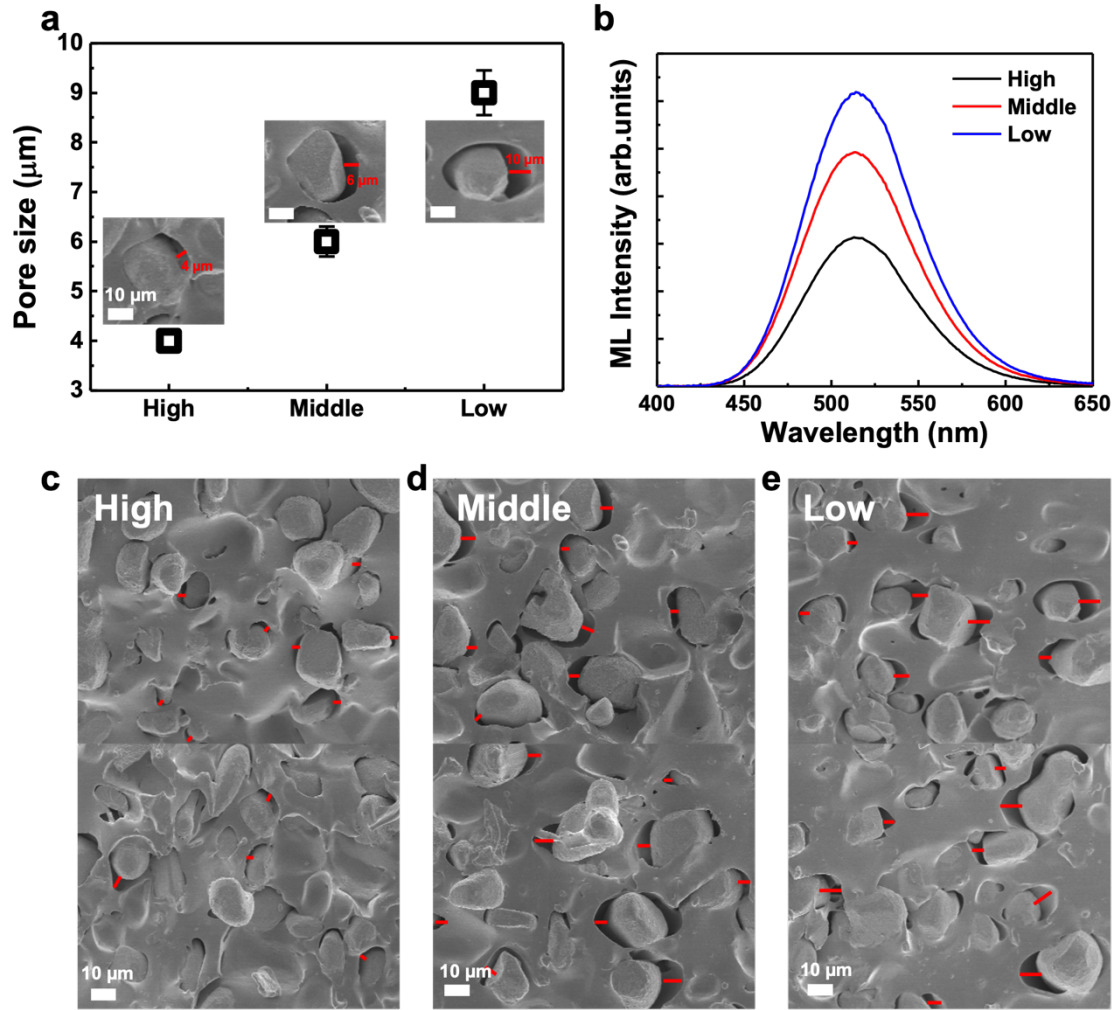
Supplementary Fig. 9 | ML performance depending on the ZnS:Cu loading ratio

a Maximum strain point and **b** relative ML intensity of the ML platforms with each polymer matrix depending on the ZnS:Cu loading ratio (wt%). Error bars in **a** indicate 5% error.

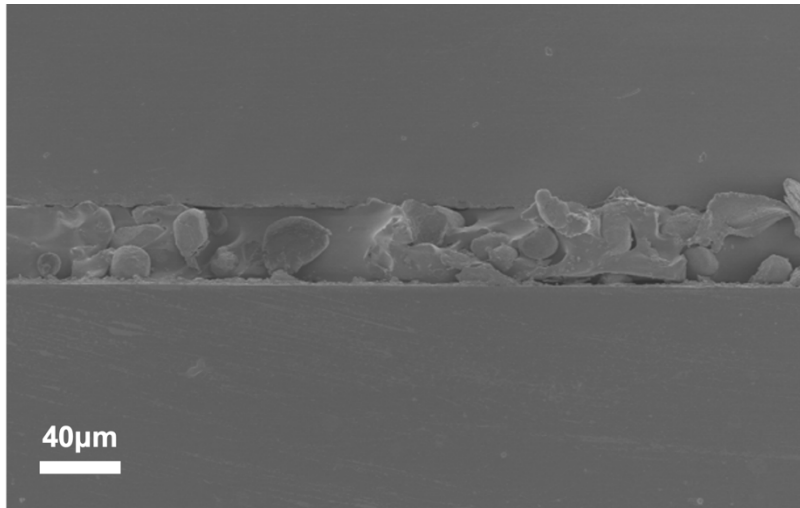


Supplementary Fig. 10 | Durability test of ML platform with PDMS according to the elasticity

The photo of strain test for ML platform with PDMS according to elasticity.



Supplementary Fig. 11 | ML performance and pore gap size with PDMS based on the elasticity
a Average pore size, and **b** relative ML intensity of ML platforms according to PDMS elasticity. Error bars in **a** indicate 8% error. Cross-sectional SEM image of ZnS:Cu in the PDMS matrix showing the average pore size depending on the **c** high, **d** middle and **e** low elasticity, respectively.



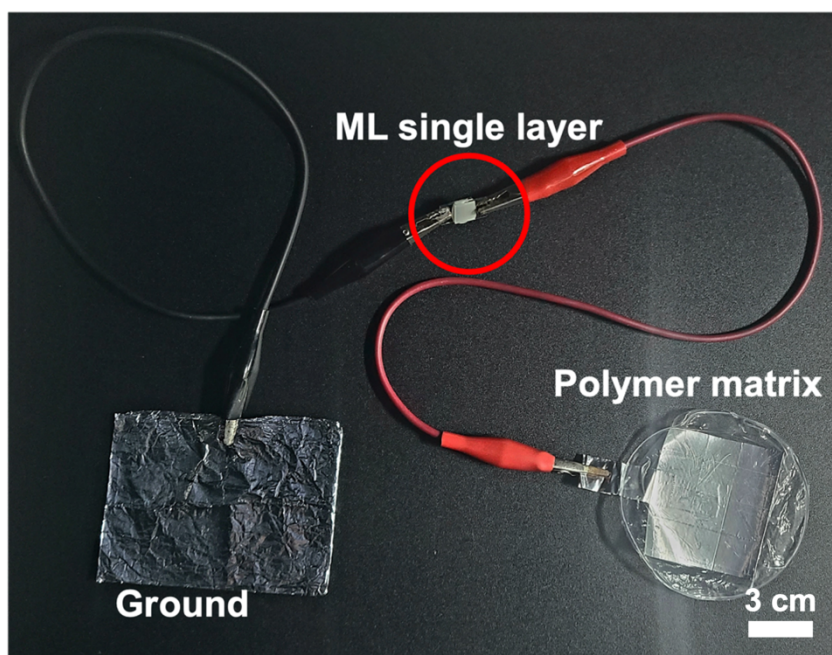
ITO glass

ML single layer

ITO glass

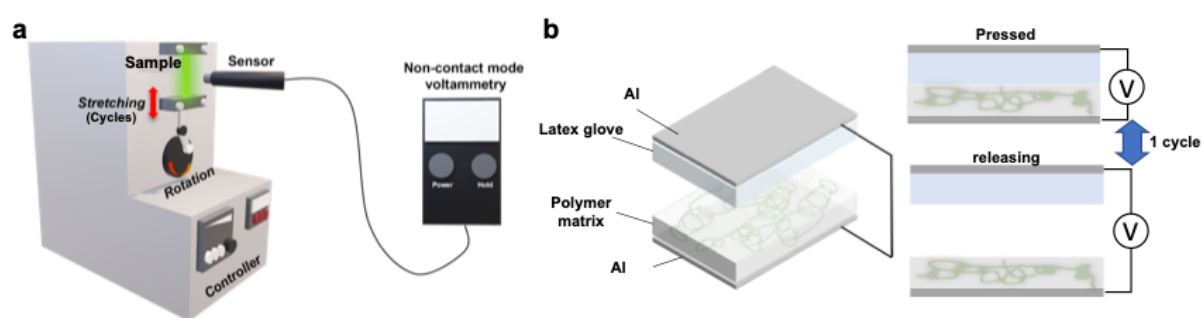
Supplementary Fig. 12 | Structure of the ZnS:Cu single layer

SEM image of the ZnS single layer between ITO electrodes.



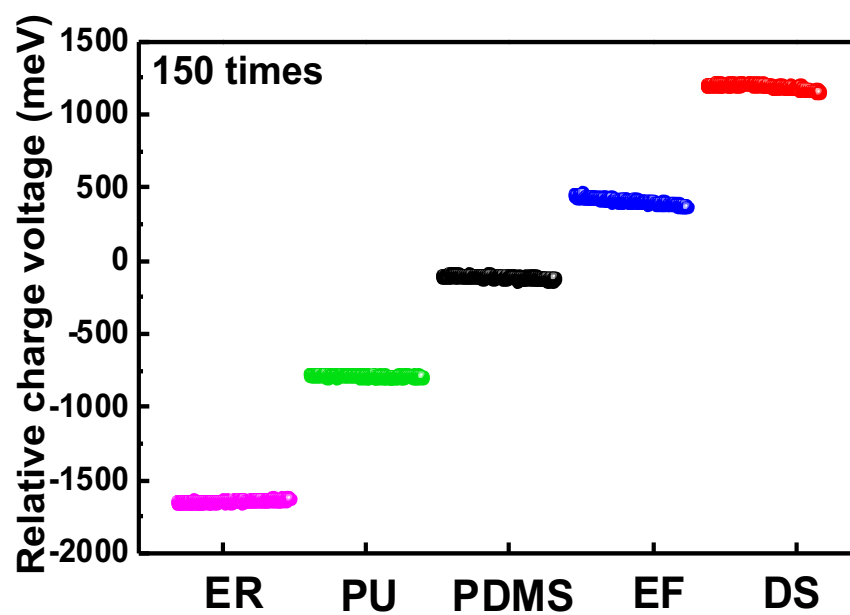
Supplementary Fig. 13 | Experimental setup for measuring ZnS:Cu luminescence induced by polymer triboelectricity

The photo shows the newly designed setup for measuring the ZnS:Cu luminescence from triboelectric effect of the polymer matrices.

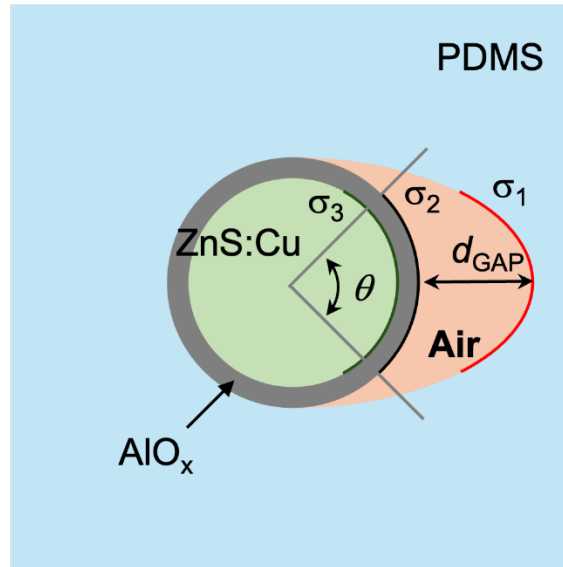


Supplementary Fig. 14 | Measurement methods for triboelectricity of polymer matrices

The schematics of the polymer sample for measuring **a** surface charge density and **b** surface output voltage.

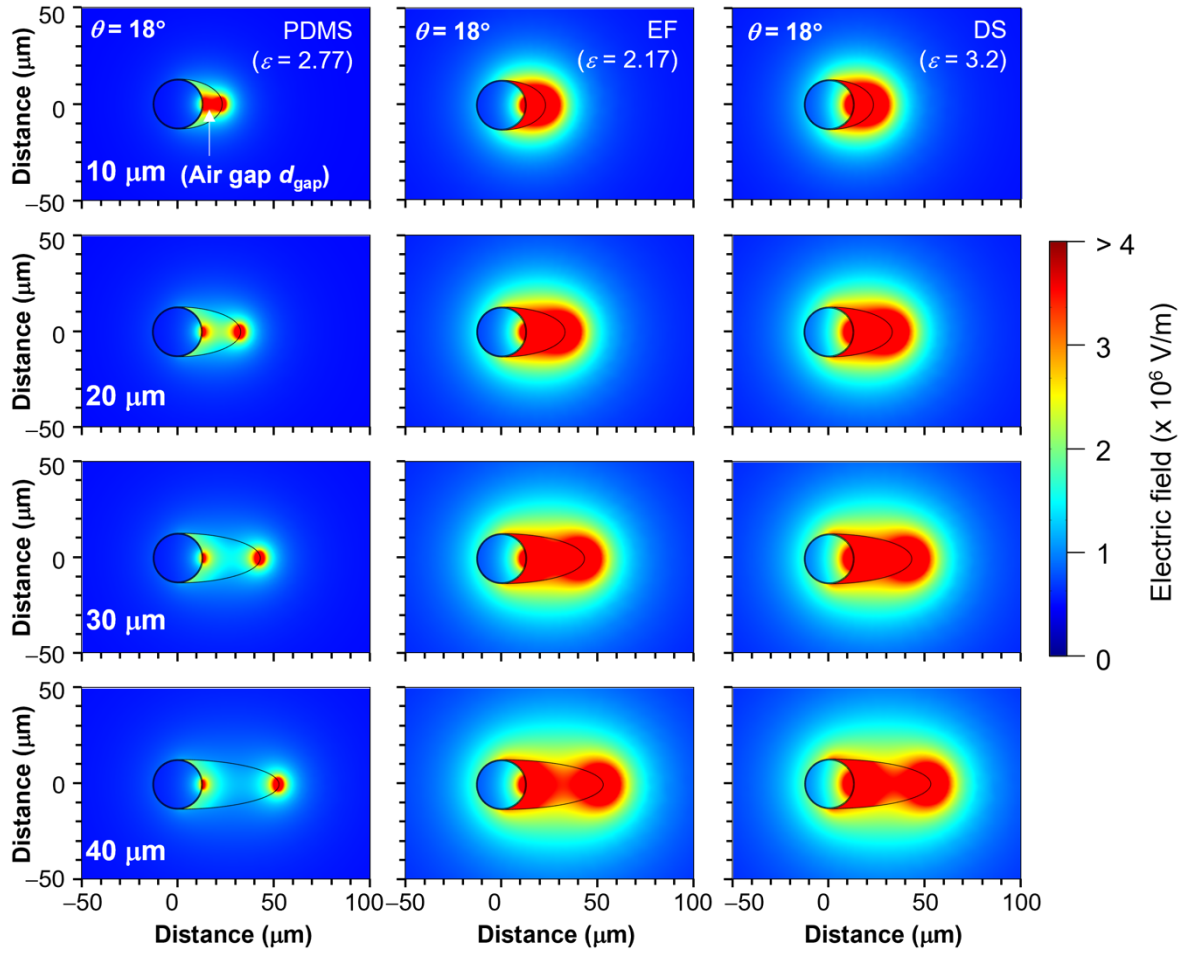


Supplementary Fig. 15 | Kelvin probe force microscopy (KPFM) results of each polymer matrix
Relative charge voltage on the surface of each polymer matrix during 150 sliding cycles.



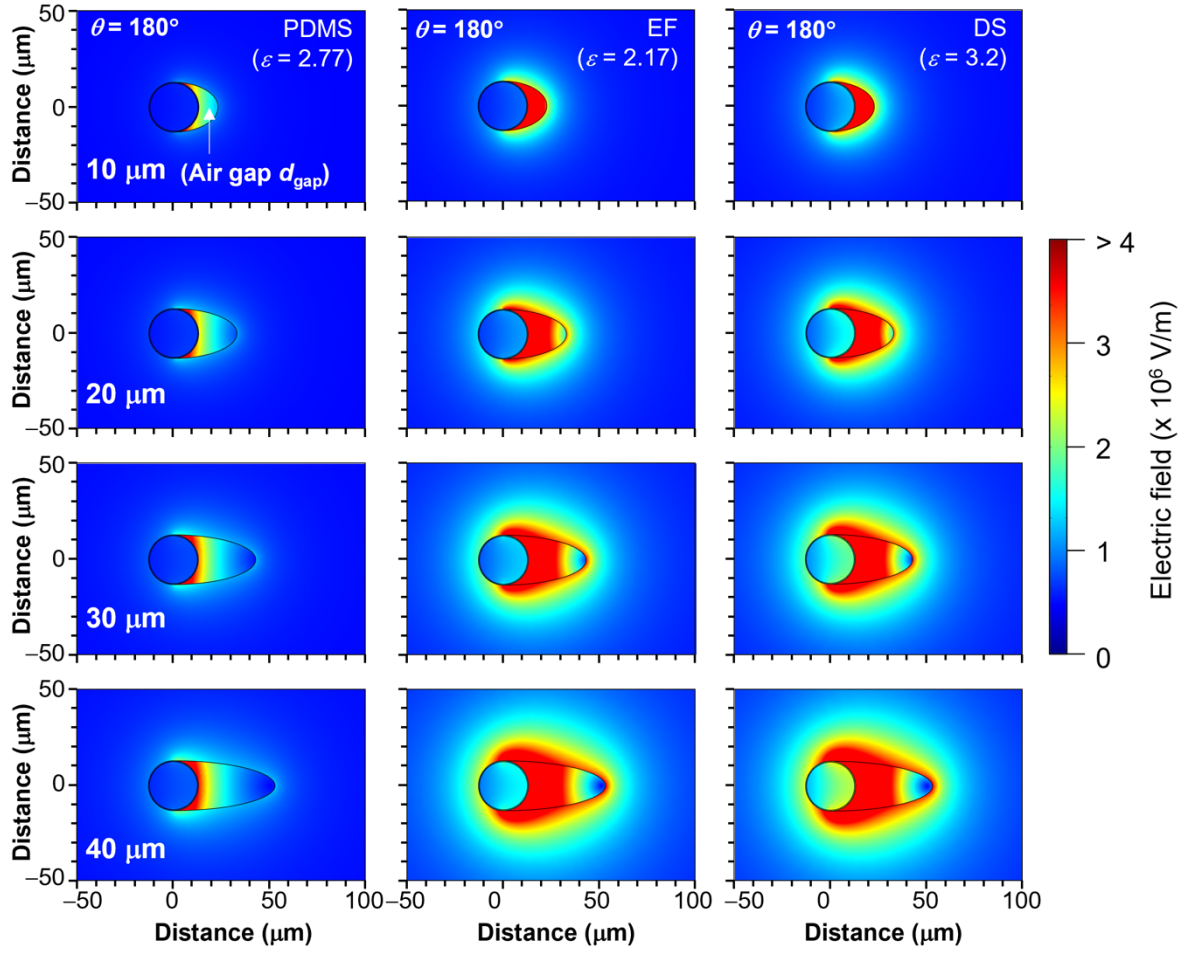
Supplementary Fig. 16 | The modeling of FDTD simulation for analysing the interface between ZnS:Cu and each polymer matrix

The simulation considers surface distributed charge density and pore gap size (d_{gap}), respectively.



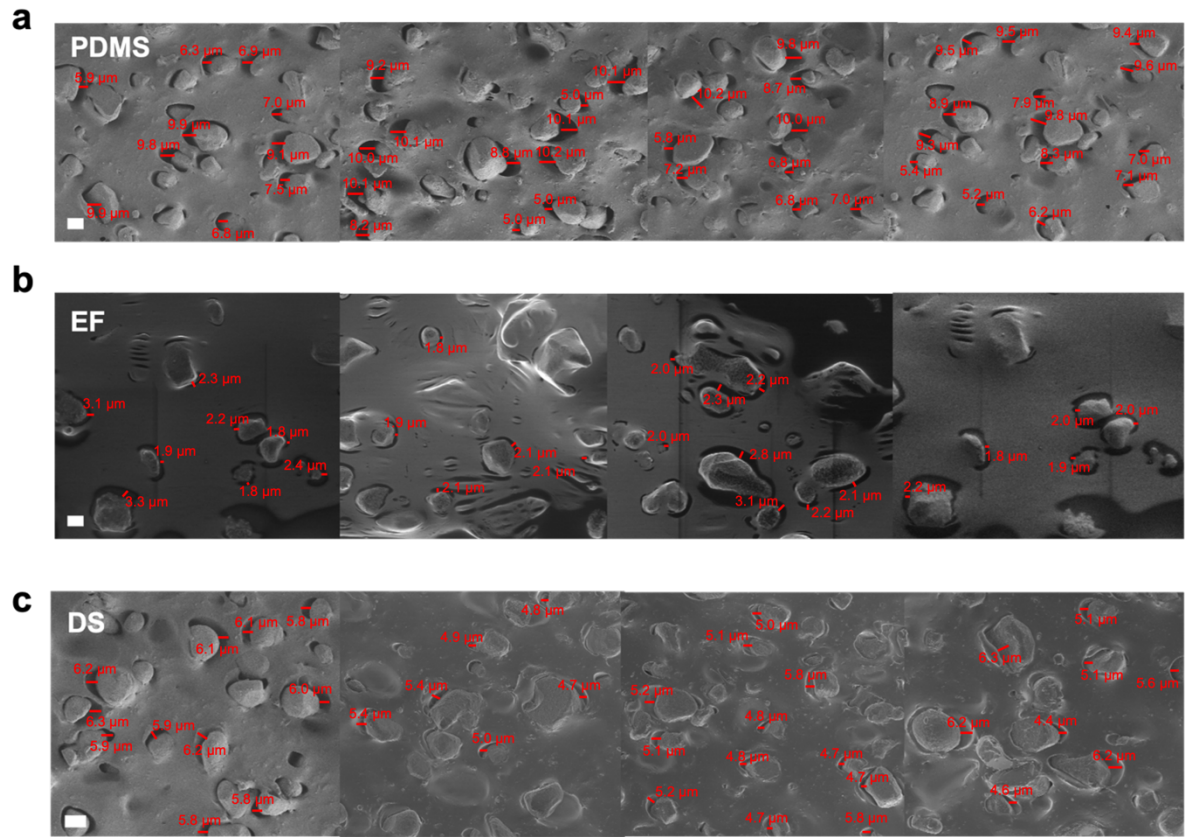
Supplementary Fig. 17 | Triboelectric field in interfacial gaps between ZnS:Cu and polymers

The simulated triboelectric field in the interfacial gap at 10 μm, 20 μm, 30 μm and 40 μm between ZnS:Cu and polymers as PDMS, EF and DS, respectively, where the surface charge density is distributed over one-tenth of the total area ($\theta = 18^\circ$).

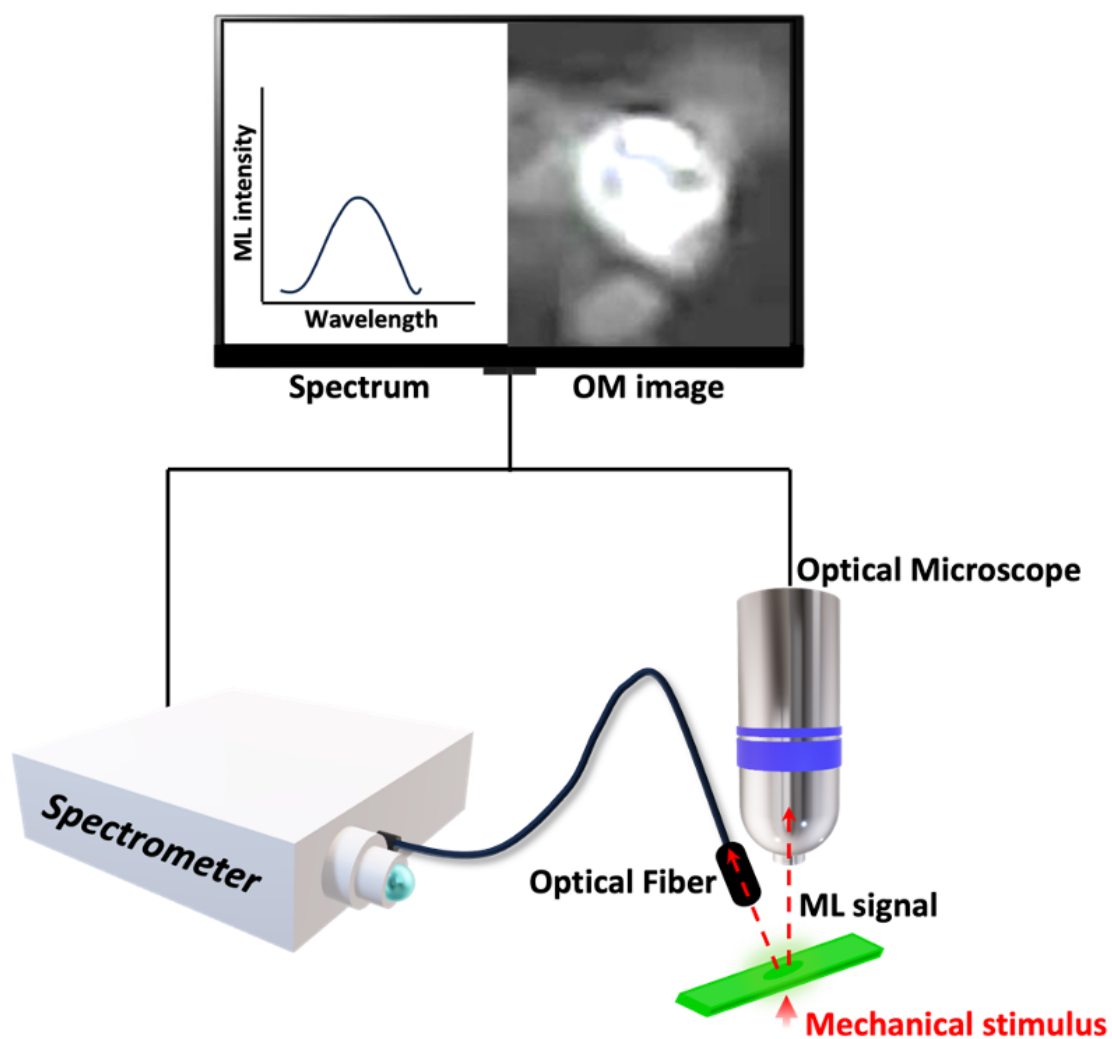


Supplementary Fig. 18 | Triboelectric field in interfacial gaps between ZnS:Cu and polymers

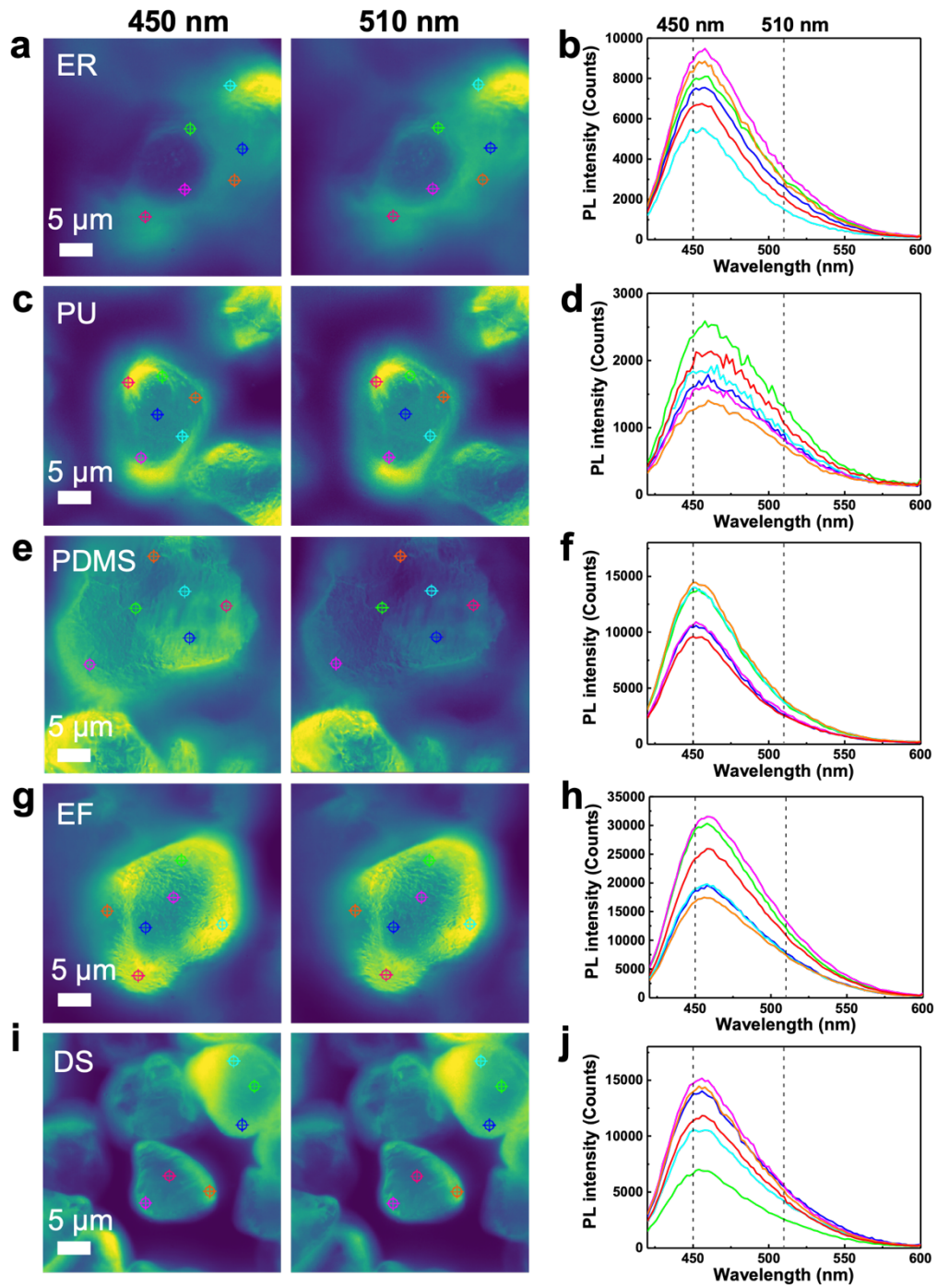
The simulated triboelectric field in the interfacial gap at 10 μm , 20 μm , 30 μm and 40 μm between ZnS and polymers as PDMS, EF and DS, respectively, where the surface charge density is distributed over half of the total area of ZnS ($\theta = 180^\circ$).



Supplementary Fig. 19 | Pore gap size of ML platform with PDMS, EF, and DS under the strain
Cross-sectional SEM images of the ML platform for **a** PDMS, **b** EF and **c** DS, respectively, under the same stretching motion.

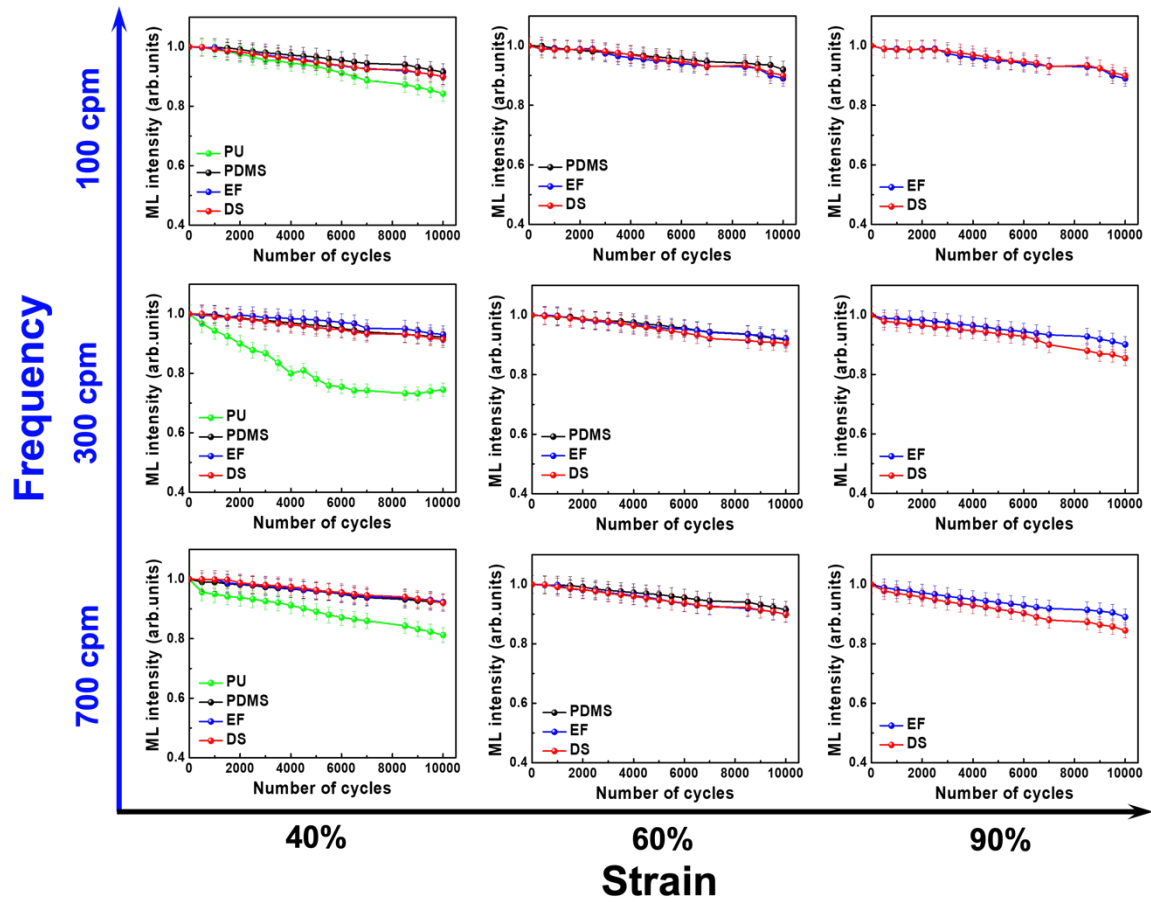


Supplementary Fig. 20 | Custom-made setup for measuring ML spectra and imaging
Schematic of the operando set-up for ML spectra and microscope imaging of the ZnS:Cu single particle.



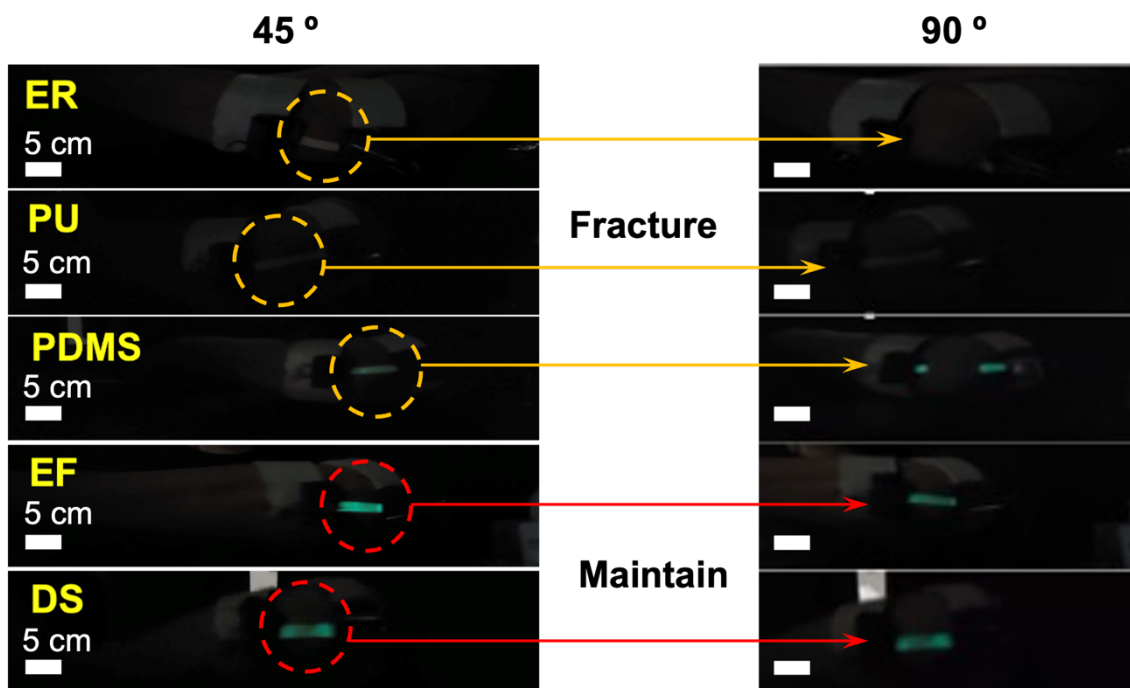
Supplementary Fig. 21 | PL performance of single particle in ML platform depending on the polymers

Hyperspectral PL maps and spectra of ML platform corresponding to each polymer matrices as **(a, b)** ER, **(c, d)** PU, **(e, f)** PDMS, **(g, h)** EF and **(i, j)** DS, respectively.



Supplementary Fig. 22 | ML stability test under continuous mechanical stimulus

Stability test of the ML single fiber under continuous mechanical cycles (up to 10,000 number of cycles (cpm)), different strain ranges (40%, 60%, 90% strain), and stretching-releasing frequencies (deformation speed: 100, 300, 700 cpm). Error bars represent a 5% error across all values of each ML intensity.



Supplementary Fig. 23 | ML performance based on polymer matrix during arm bending motion
 ML performance depending on the polymer matrices under the arm bending at 45 and 90 °.

Supplementary Table 1 | The durability test values of polymer matrices without phosphor and with phosphor.

	Without phosphor			With phosphor		
	Initial	Stretch	Release	Initial	Stretch	Release
ER	8.5	10.1	8.8	8.5	9.5	8.7
PU	8.5	29.3	13.4	8.5	19.7	9.8
PDMS	8.5	13.3	8.5	8.5	11.5	8.5
EF	8.5	26.7	8.5	8.5	26.7	8.5
DS	8.5	16	8.5	8.5	14.6	8.5

*The unit for all values is centimeters (cm).

Supplementary Table 2 | Maximum strain values of ML platform depending on the ZnS:Cu loading ratio (wt%).

ZnS loading ratio (wt%)	0	10	40	70	80
	Maximum strain (cm)				
ER	10.1	9.8	9.6	9.5	9.3
PU	29.3	27.5	23.4	19.7	15.3
PDMS	13.3	12.2	11.9	11.5	11.0
EF	26.7	26.8	26.6	26.7	26.4
DS	16	15.3	14.9	14.6	14.1

Supplementary Table 3 | Strain values of the ML platforms with PDMS depending on the different elasticity.

	Elongation (cm)	Strain (%)	Durability (%)
High	4.7	55	155
Middle	3.1	36	136
Low	2.0	24	124

Supplementary Table. 4 | Parameters of ZnS:Cu and polymer matrices for the FDTD simulation.

Polymer	Surface charge density		Dielectric constant (1MHz)	Thickness
PDMS	($\sigma_{1\text{-PDMS}}$)	-31.9 nC/cm ²	2.77	
EF	($\sigma_{1\text{-EF}}$)	-102.1 nC/cm ²	2.17	
DS	($\sigma_{1\text{-DS}}$)	-146.7 nC/cm ²	3.2	
AlOx	(σ_2)	10 nC/cm ²	9	500 nm
ZnS:Cu ²⁺	(σ_3)	-11.3 nC/cm ²	8.4	Diameter 25 μm
Air (Transmission medium)	-		1	d_{GAP} : 0 ~ 40 μm