Supporting Information

Electroluminescent Soft Elastomer Actuators with Adjustable Luminance and Strain

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FEM simulation of the electric field in ZnS particles

Modeling: One 0.7 μm diameter ZnS:Cu particle was placed in the center of an Ecoflex cube (one side length of 3 μm). The actual size of ZnS is too large (average diameter: 9 μm) compared to the size of $BaTiO_3$ (average diameter: 50 nm), making simulation difficult. Therefore, it is set smaller than the actual size (diameter: 0.7 μm) for smooth simulation. The 50 nm diameter $BaTiO_3$ were arranged around the ZnS particle in the x, y, and z directions at intervals of 0.1 μm (Figure S3a).

Boundary condition: 100 V was applied between the two top and bottom surfaces of the cube. This corresponds to applying an electric field of 34 MV/m across the cube. This is similar to the electric field when 7.3 kV is applied to a 216 μm thick ELDEA.

Material parameter: the permittivity of Ecoflex, ZnS, and $BaTiO_3$ were set as 3, 10, and 300, respectively.

Software: COMSOL Multiphysics (COMSOL Inc, USA). The simulation was run in electrostatics application mode.

Result: The profile of the electric field applied to the ZnS when $BaTiO_3$ is added is as shown above in Fig S3c, and the profile before $BaTiO_3$ addition is as shown in Fig S3c below. As shown in Figure S3b, the electric field applied to the center of ZnS increased by 521 kV/m when $BaTiO_3$ was added. As a result, it can be expected that the luminance of ZnS becomes higher with $BaTiO_3$ addition.

Dielectric constant measurement

A parallel plate capacitor type device was fabricated by sandwiching a VHB tape and a $ZnS:Cu/BaTiO_3$/Ecoflex composite between copper tapes. The capacitance was measured with a HIOKI 3532-50 LCR meter. Using simple capacitor equation $C=\varepsilon_0 \varepsilon_r A/d$, the capacitance value was converted to relative permittivity using the thickness and width of the sample. At 1 kHz, relative permittivity of VHB tape and EL layer were 4.6 and 7.4, respectively.

Mechanical transient behavior of ELDEA

As the applied voltage increases, the dielectric elastomer behaves as a pure elastic material for a short time and expands instantaneously in response to the applied voltage increase. The elastomer then expands as a function of time due to the viscoelastic creep.[1] The ELDEA showed a very short response time with a strain increase due to AC amplitude increase at fixed DC offset within about 4 ms (Figure S5a) and a strain increase due to DC offset increase at fixed AC amplitude within about 50 ms (Figure S5b).

Estimation of Effective Modulus of Elastomer

The compressional strain in the thickness direction of the dielectric elastomer is

$$s_z = -\frac{\varepsilon\varepsilon_0}{Y}\left(\frac{V}{z_0}\right)^2, \qquad (1)$$

where ε is the dielectric constant, ε_0 is the permittivity of free space, Y is the modulus of the elastomer, V is the applied voltage, and z_0 is the initial thickness of the elastomer. At this time, the modulus of the elastomer significantly increases as the frequency of the applied voltage increases.[2] As a result, at high frequencies, the elastomer is hardly affected by the AC amplitude. Our estimation of the effective modulus of the elastomer says that the areal strain is below 0.01% under an AC voltage of 5 kV_{pp}.

First, it is assumed that the dielectric constants of the EL and dielectric elastomer layers are constant at 7 and 4.5. Considering that the initial thicknesses of the EL layer and the dielectric elastomer layer are 100 μm and 58 μm, a voltage of 0.89 kV_{pp} is applied to one layer of the dielectric elastomer when a voltage of 5 kV_{pp} is applied to the entire EL device.

The relationship between the thickness and area of the elastomer is as follows:

$$z = z_0(1 + s_z), \qquad (2)$$

$$Az = A_0 z_0 , \qquad (3)$$

where z is the thickness of the elastomer, A is the area of the elastomer, and A_0 is the initial area of the elastomer. Since the areal strain is $\frac{A - A_0}{A_0} \times 100\ (\%)$, substituting Eqs.(2) and (3), s_z is approximately -0.0001. Therefore, when this value and the remaining constants are substituted into Eqs.(1), the effective modulus is approximately 23MPa. That is, as the frequency of the voltage increases, if the modulus of the dielectric elastomer exceeds 23 MPa, the strain of the device hardly changes even if an AC voltage of 5 kV_{pp} amplitude is applied.

Supplementary Figures

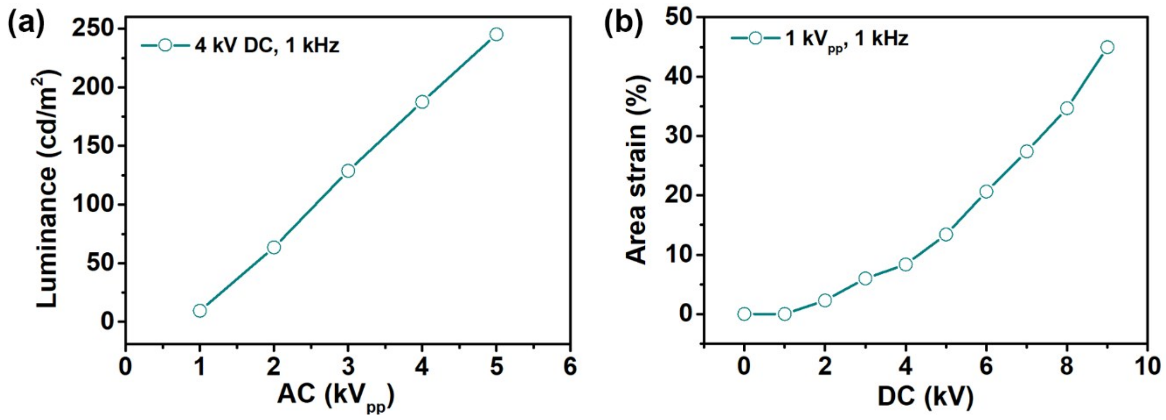

Figure S1. ELDEA performance without CNTs. (a) Luminance as a function of AC amplitude and (b) areal strain as a function of DC offset without CNT incorporation into the AgNW network.

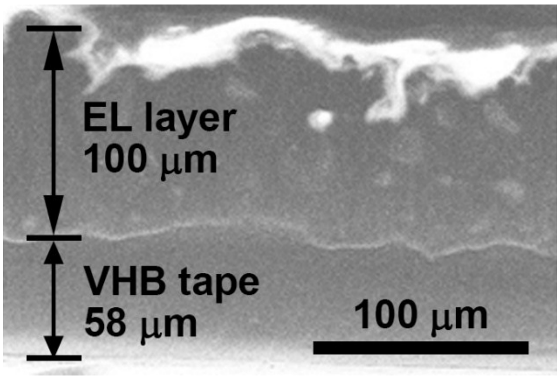

Figure S2. SEM image of the cross section of an ELDEA device. It shows the thickness of each layer.

60 Hz	AC (kV_{pp}) \ DC (kV)	0	1	2	3	4
	1	0.227	0.53	0.643	0.719	0.833
	2	4.204	4.545	4.659	4.924	5.379
	3	9.091	9.432	9.621	9.962	11.02
	4	14.35	14.5	14.73	15.34	16.62
	5	19.69	19.8	20.1	20.84	21.43

1 kHz	AC (kV_{pp}) \ DC (kV)	0	1	2	3	4
	1	13.28	14.05	15.33	18.06	19.84
	2	59.78	62.15	66.77	74.66	80.58
	3	122.1	126.6	133.2	146.9	156.5
	4	186.2	191	201	217.2	230.2
	5	248.3	253.5	265	282	299.8

3 kHz	AC (kV_{pp}) \ DC (kV)	0	1	2	3	4
	1	17.88	20.02	21.84	23.72	26.09
	2	92.87	102.9	107.3	120.6	128.8
	3	204.7	215.8	229.8	249.1	265.7
	4	327.6	339	359	384.6	406.8
	5	451.9	466.7	485.2	512.6	536.7

Table S1. Numerical data of ELDEA luminance at various DC offset, AC amplitude and frequency conditions shown in Figure 4c.

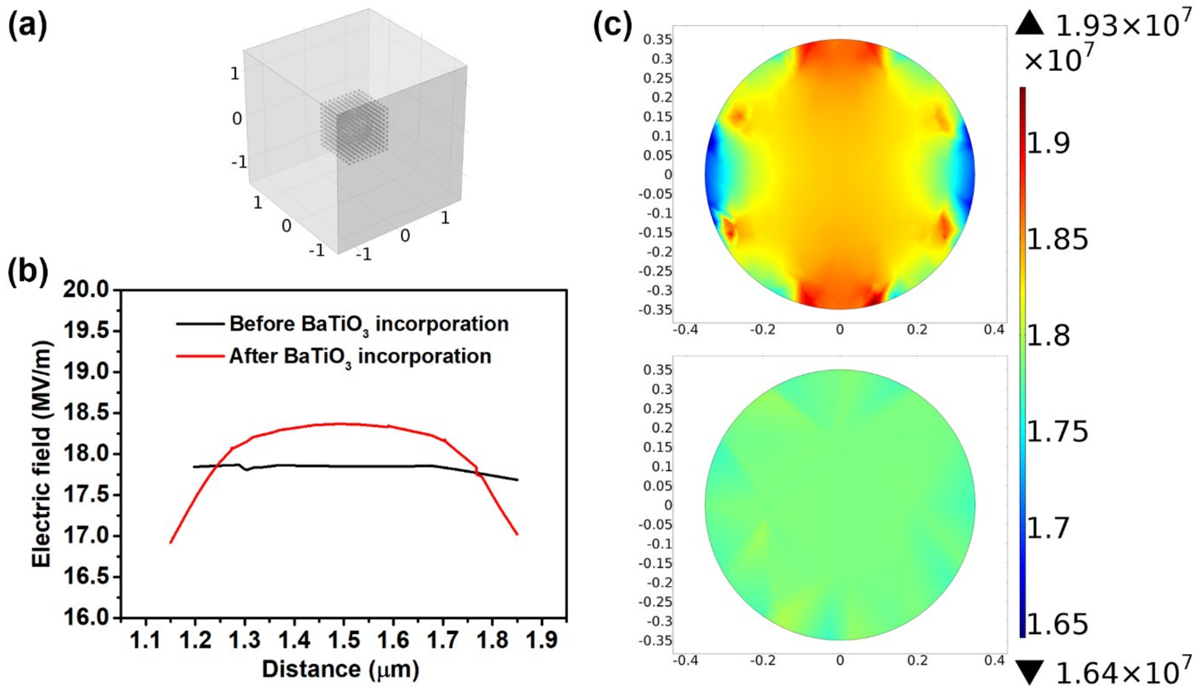

Figure S3. Finite element method simulation of $BaTiO_3$ incorporation effect into EL layer. (a) Geometry of simulation modeling. The dimensional unit is in μm. (b) Electric field across a line passing through the center of the ZnS particle. (c) Visualization of the electric field distribution before (below) and after (above) $BaTiO_3$ incorporation. The unit of color scale bar is V/m.

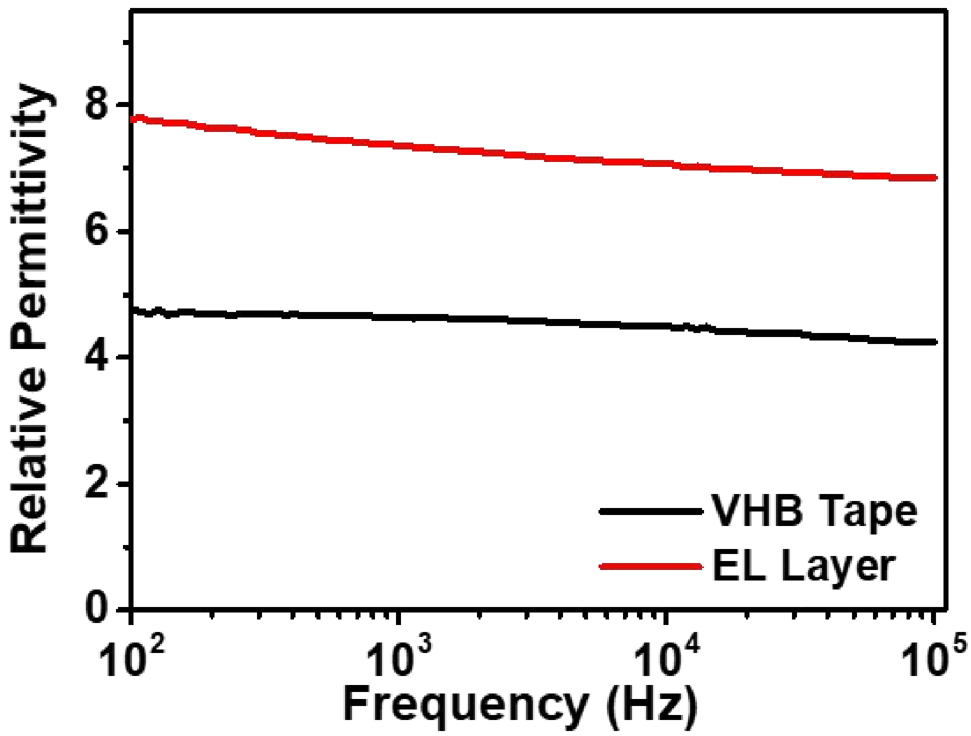

Figure S4. Relative permittivity of each polymer layer. The detailed measurement and calculation processes are described above, in supplementary methods.

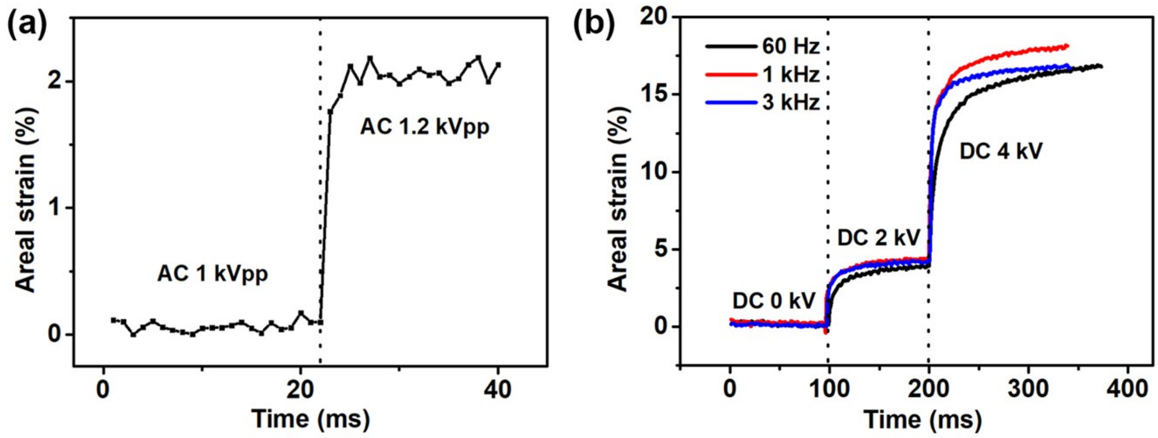

Figure S5. Mechanical transient behavior of actuator according to applied voltage change. (a) Strain change when increasing AC amplitude at fixed DC offset 2 kV and 60 Hz. (b) Strain change when increasing DC offset at fixed AC amplitude 1 kV_{pp}.

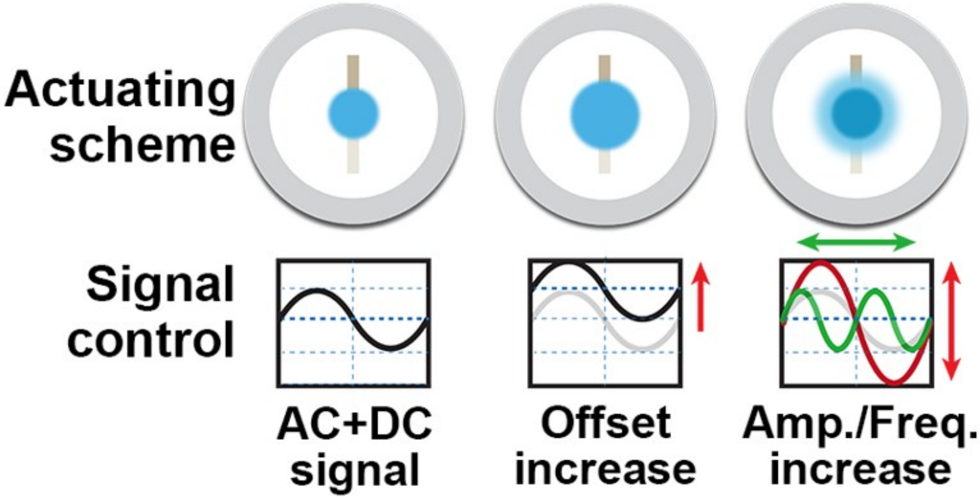

Figure S6. Luminance and strain characteristics of a representative ELDEA sample. Schematic diagram showing individual adjustment of luminance and strain by AC amplitude/frequency and DC offset control.

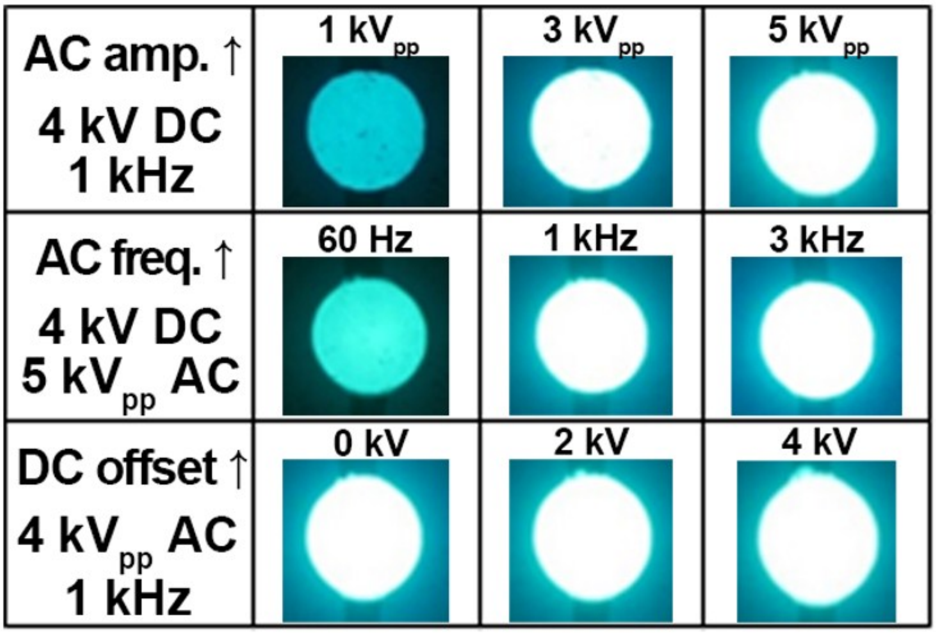

Figure S7. Photographs of the ELDEA under various different AC and DC conditions. All images are 2.84×2.84 cm^2 in size.

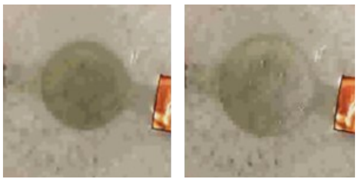

Figure S8. Photographs of the ELDEA when only DC voltage of (left) 1 kV and (right) 5 kV is applied. All images are 3.76×3.76 cm^2 in size.

Video

Video S1: Operation of ELDEA according to applied voltage. Video shows the operation of ELDEA with DC offset change at fixed frequency and AC amplitude.

Supporting Information Reference

(1) Keplinger, C., Sun, J.-Y., Foo, C. C., Rothemund, P., Whitesides, G. M., and Suo, Z., Stretchable, Science, 2013, **341**, 984-987.

(2) M. Molberg, Y. Leterrier, C. J. Plummer, C. Walder, C. Löwe, D. M. Opris, F. A. Nüesch, S. Bauer and J.-A. E. Månson, *J. Appl. Phys.*, 2009, **106**, 054112.