**Methods**

**Network design**

UV-polymerization has been widely used for the synthesis of polymers due to its simplicity, controllability and low polymerization temperature. Thus, UV-polymerization was selected in this work to synthesize the acrylic-based dielectric elastomer. Firstly, a quantitative structure-property relationship (QSPR) approach was used to theoretically select the optimum monomer from 11 commonly-used acrylate monomers. Some critical physical parameters of these acrylate polymers including molar stiffness function (MSF), cohesive energy (CE) and glass transition temperature (*Tg*), which represent the flexibility, intermolecular interaction energy and minimum working temperature, respectively, were calculated by this QSPR method (Extended Data Fig. 1). The obtained results indicated that *n*-butyl acrylate (BA) monomer exhibited the optimum comprehensive properties for the synthesis of acrylic-based dielectric elastomer.

Moreover, the MSF and *Tg* of 18 commonly-used flexible long chain oligomers with polyester, polyether and silicone as repeat units as well as the compatibility between these oligomers and BA monomers were calculated (Extended Data Fig. 2). Considering the features of flexibility and solubility, the oligomers with polyether as repeat units was recommended for the optimization of the network of *n*-butyl acrylic-based elastomer. Additionally, the average molecular weight of oligomers should be located in the range of 104 g mol-1 to 105 g mol-1 since a structurally uniform polymer network can only be achieved in this molecular weight range (see Extended Data, Part: “Selection of number average molecular weight**”**). According to the calculated results and analysis discussed above, CN9021NS, a difunctional urethane acrylate compound composed of a flexible polyether diol segment and an aliphatic diisocyanate segment, was chosen for elastomer network optimization (see Extended Data, Part: “Selection of number average molecular weight**”**). Besides, another kind of *n*-butyl acrylate homopolymer, crosslinked by the equimolar (taking BAC2 as reference) polyethylene glycol (400) diacrylate, was also prepared for comparison.

**Selection of number average molecular weight**

According to the previously reported studies, crosslinking density of dielectric elastomers should be in the range of 10-5 mol cm-3 to 10-4 mol cm-3.42 The density of elastomers is usually around 1 g cm-3. Thus, the range of the crosslinking density could be derived as 10-5 mol g-1~10-4 mol g-1. Afterwards, the average molecular molar weight between adjacent crosslinking points should be in the range of 104 g mol-1 to 105 g mol-1.

Uniform distribution of crosslinking points is helpful to alleviate or eliminate the stress concentration inside elastomer network, which is usually regarded as a key issue that leads to the early failure of elastomer. Therefore, a difunctional oligomer with an average molecular molar weight in the range of 104 g mol-1 to 105 g mol-1 is desirable for the crosslinking of elastomer. Based on this principle, CN9021NS, a kind of macromolecular crosslinking agent, was specifically selected and its mean relative molecular weight distribution was characterized by gel permeation chromatography in tetrahydrofuran solvent (Shimadzu LC20/RID-20) (Fig. 2d).

**Preparation**

The dielectric elastomer was synthesized via a UV curing method. *n*-butyl acrylate (BA, 99.5 %) from J&K Scientific Ltd was selected as monomer. CN9021NS (difunctional acrylate esters resin) and polyethylene glycol (400) diacrylate (PEG(400)DA) were purchased from Sartomer Company as crosslinking agents. 2-hydroxy-2-methylpropiophenone from Shanghai Macklin Biochemical Co., Ltd was used as photo-initiator. Formulations composed of monomer, crosslinking agent and photo-initiator (as shown in Extended Data Table 1) were mixed thoroughly by vortex oscillator to form a homogeneous precursor solution.

A custom-built vessel composed of a silicone spacer sandwiched by release films and glass plates in a proper order was fabricated to create an oxygen-free condition. The precursor solution was injected into that vessel, which was subsequently followed by a vacuum degassing process for 15 min. After that, it was cured in a UV curing reaction chamber with a UV intensity of 2.5 W cm-2 for 3 min. Finally, the elastomer was placed into oven at 40 °C for 24 h to remove the remnant monomer. The film thickness can be easily adjusted by tuning the thickness of silicone spacer.

**Characterization**

**Gel experiment and light transmittance test**: All the samples with a thickness of 1mm were cut into 10 mm×10 mm squares and their original mass was subsequently measured. Then, they were immersed in the tetrahydrofuran solvent for a week. And the dimensions after swollen were measured to obtain the swelling ratio, which is defined as the cubic ratio of the length of a swollen elastomer to its original length. After drying the samples in oven at 40 °C for 24 h, the final mass was measured to obtain the gel fraction9. Light transmittance test with the wavelength ranging from 380 nm to 800 nm for the films with a thickness of 1 mm was conducted by Lambda™ 35 UV/VIS Spectrometer from PerkinElmer. Since each formulation exhibits high transparency, BAC2 was taken as a representative sample (Extended Data Fig. 3).

**Elemental analysis**: Firstly, the uncrosslinked chains extracted from BAC2 sample after swollen in the tetrahydrofuran solvent was dried in oven at 60 °C for at least 24 h to evaporate solvent completely. Then, CN9021NS and those free chains in BAC2 were measured on PERKIN ELMER CE-440. At least 3 samples were measured to obtain a reliable result.

**Dielectric properties**: Dielectric properties in the range of 100 Hz to 1 MHz were characterized by a Broadband Dielectric Spectroscopy (Novocontrol Technologies GmbH & Co. KG). The thickness of the samples used for dielectric characterization was 1 mm.

**Mechanical properties**: The measurement was conducted according to the standard of ISO 37. To eliminate the effect of thickness, all samples were in a fixed thickness of 1 mm. Type 1A dumbbell shaped samples were fabricated with a cutting die and stretched on Instron 3343 at the rate of 500 mm min-1 to obtain Young’s modulus. After that, samples were cut into a dumbbell shape with an effective area of 2 mm width by 12 mm length and stretched at the rate of 200 mm min-1 until fracture to obtain stress vs strain curves. Finally, samples were cut into a narrow strip of 10 mm width by 50 mm length (the gauge length was 30 mm) and stretched to triple at the rate of 100 mm min-1 to obtain stress vs strain loops. Each formulation was tested at least three times. Dynamic mechanical analysis (DMA) was carried out on a TA Q800 dynamic mechanical analyzer with a frequency of 1 Hz, < 2 % strain, and a temperature range from -45 °C to 50 °C with a ramping rate of 7 °C min-1.

**Actuation properties**: The nominal electric field was calculated by dividing the applied voltage by the initial thickness of elastomer film before electrically actuated. For the films free of pre-strain, the samples with a thickness of 1 mm were fixed on an annular PMMA rigid frame with an inner diameter of 20 mm (*d*=20 mm). And carbon grease (NyoGel 756G, Nye Lubricants) was coated onto both sides of film as the compliant electrodes. The voltage applied on the films was increased at a step of 1 kV until electric breakdown and each strain value was obtained by holding the constant voltage for 1 min. A laser sensor (HL-G105-S-J, Panasonic) was used to measure and record the invagination depth (*h*) in the center of actuator. Finally, actuation area strain (*S*) is derived from the equation: (Extended Data Fig. 5) Each formulation was tested at least three times. As for planar area strain, films were fixed on a PMMA rigid frame with an inner diameter of 120 mm after equiaxially stretched to 4×4 times (Supplementary Video 4). And the compliant electrodes with a 25-mm diameter were fabricated by coating carbon grease (NyoGel 756G, Nye Lubricants) onto both sides of films. Electric field was increased from 0 MV m-1 to 70 MV m-1 by a step of 10 MV m-1 and each step was held for 30 s. A digital SLR camera was used to record the actuation process. The actuation strain was obtained by a MATLAB script for video processing43. At least 3 samples for the same composition were tested.

**Dynamic response**: The experiment condition in this part was similar to that of the planar area strain characterization except that the electric strength applied on film was 40 MV m-1 and held for 10 min. Each formulation was tested at least three times.

**Fabrication and actuation performance of non-magnetic motor**

After equiaxially stretched to 4×4 times, films were fixed on a PMMA rigid frame with an inside diameter of 140 mm using a specially-designed stretching rig and the films were divided into four parts by electrodes. And carbon grease (NyoGel 756G, Nye Lubricants) was used for the fabrication of compliant electrodes. In the center, a circular ring with an inside diameter of 40 mm was used to maintain pre-strain and fix gears. Single chip machine (SCM) was written to control relays on and off. Thus, the frequency of driving voltage can be easily tuned by adapting the SCM script. Rotary motion was recorded by a digital SLR camera, and rotation property was obtained from the video frame-by-frame. The schematic diagram of non-magnetic motor driving system is shown in Extended Data Fig. 7. The motor was mounted with its axis aligned vertically and a string bound with the rotor and ran over the pully mounted on table to connect a basket6. Then, torque was measured by placing a fixed weight in the basket at each driving electric field and frequency. The weight was increased until the motor lost its synchronization with driving electric field. At last, output power was calculated by the product of torque and rotation speed.

**Toughness**

Apart from enhancing dielectric constant, lowering the elastic modulus is another feasible approach for the increase of the actuation sensitivity of elastomer films. However, materials with low moduli is usually delicate, consequently restricting the application of elastomers. Thus, toughness () was discussed here, which is defined as the area surrounded by the stress - strain curves from uniaxial stretch using equation (S1):

(S1)

where, is the stress (MPa) and is the strain (mm mm-1).

Compared with the pervious DEs with a low elastic modulus (< 1 MPa), polyacrylate dielectric elastomers synthesized in this work exhibits an overwhelming advantage on toughness (Extended Data Fig. 5 and Fig. 1e). Despite owning an ultralow Young’s modulus, BAC2 also has excellent stretchability and tensile strength due to the uniform crosslink network and the existence of trace amounts of hydrogen bonds44. The combination of low modulus and high toughness makes the synthesized elastomer desirable in the different applications.

**Calculation of transient time constant and simulation of electrically driving frequency**

The theoretical analysis of the charging process was carried out through an equivalent circuit model. Extended Data Fig. 8a displays the schematic of the equivalent circuit model of non-magnetic motor. Since the electromechanical response speed of DEAs not only depends on mechanical properties like viscoelasticity and stiffness but also is related to electrical parameters. The evolution of voltage applied to elastomer film with the time during the charging process will be systematically discussed as follows.

When a fixed bias voltage is applied between two electrodes, there is a transient process for voltage establishment on elastomer film.

According to Kirchhoff’s voltage law,

(S2)

where is the protective resistance, is the equivalent resistance of electrodes, is the output current of power supply, and and are the voltages of power supply and capacitor, respectively.

Next, according to Kirchhoff’s current law,

(S3)

where and are the resistance and capacitance of the equivalent parallel model of elastomer film as a plate capacitor. Since mechanical response is much slower than electrical response, the capacitance change of could be neglected. Thus, the following equation can be derived from Equation (S2) and (S3).

(S4)

It’s easy to obtain the solution of Differential Equation of the first-order.

(S5)

Time constant () can be gotten from Equation (S5).

(S6)

and , where , and are volume resistivity, permittivity of vacuum and relative dielectric constant, respectively. and are the surface area and thickness of film after stretched, respectively.

From the engineering point of view, it usually needs 3τ~5τ for the voltage to achieve the steady-state value (Equation S5). Therefore, the driving frequency is limited to such a narrow range (), where is the duty ratio of square wave voltage. As shown in Extended Data Fig. 9a, the frequency limitation is set to 500 Hz by adjusting the parameters of circuit elements. When the power frequency is well below 500 Hz, for example 50 Hz, the voltage on capacitor can reach the steady-state value (Extended Data Fig. 9b, S9c). On the contrary, when the power frequency exceeds 500 Hz, for example 5000 Hz, the voltage on capacitor is much lower than the steady-state value due to unfinished charging process within extremely short time (Extended Data Fig. 9d).

As for the non-magnetic motor, a thorough analysis and simulation were conducted. The high-field volume resistivity of BAC2 and VHBTM4910 were measured by two-probe method. The volume resistivity of BAC2 is of the order of magnitude of 1011 Ω m, which is two orders of magnitude smaller than that of VHBTM 4910. And the relative dielectric constant of BAC2 and VHBTM 4910 is around 5.75 and 4.4 at 1 kHz, respectively. After equiaxially stretched to 4×4 times, the film was cut into a circle with a radius of 70 mm and thickness of 62.5 m. Therefore, for BAC2, is 11.5 nF and is 4.43×108 Ω. For VHBTM4910, is 8.8 nF and is 2.2×1011 Ω.

However,

(S7)

where is of the order of the magnitude of kV and is less than 1 mA. In fact, the resistance of is of the order of the magnitude of 106 Ω, which is far less than the resistance of .

Therefore, equation S6 can be simplified into the following one,

(S8)

and τ is in the magnitude of 10-2 s.

Actually, the discussion above is greatly simplified. Supposing the capacitance is a constant value during actuation, we simulated voltage on film and current from power supply (Extended Data Fig. 10). The obtained results could be concluded as follows: (i) Current did not pass the limitation (1 mA) during the whole simulation; (ii) When driving frequency exceeded about 3 Hz, voltage on film of both VHBTM 4910 and BAC2 went down. As the driving frequency range of BAC2-based motor is much larger than 3 Hz, upper limit of rotation speed not only was determined by BAC2 elastomer itself but also was related to experimental device.

**Supplementary tables, figures and videos**

Extended Data Table 1 | Formulation of acrylic dielectric elastomers.

Extended Data Fig. 1 | Comparison of molar stiffness function (MSF), cohesive energy (CE) and glass transition temperature (*Tg*) of homopolymers of commonly used acrylic monomers.

Extended Data Fig. 2 | Comparison of MSF, *Tg* and solubility parameter (δ) of common flexible long chain oligomers containing polyester, polyether and silicone as repeat units.

Extended Data Fig. 3 | Light transmittance comparison between VHBTM 4910 and BAC2.

Extended Data Fig. 4 | Comparison of toughness among VHBTM 4910 and new polyacrylates.

Extended Data Fig. 5 | Dielectric and mechanical properties of VHBTM4910, BAP and BAC series samples.

Extended Data Fig. 6 | Schematic diagram showing the actuation performance test platform for elastomers without pre-strain.

Extended Data Fig. 7 | Time dependent behavior of actuation area strain of VHBTM 4910 and BAC2 with 4×4 equiaxial pre-strain at 40 MV m-1.

Extended Data Fig. 8 | The schematic diagram of non-magnetic motor driving system.

Extended Data Fig. 9 | Theoretical analysis of the frequency-dependent behavior of the charging process for a simplified capacitor.

Extended Data Fig. 10 | Simulation of soft motor.

Supplementary Video 1 | Revolve speed of BAC2-based motor at different driving frequencies under 48 MV m-1.

Supplementary Video 2 | Revolve speed of BAC2-based motor with transmission gears at different driving frequencies under 48 MV m-1.

Supplementary Video 3 | Revolve speed of VHBTM 4910-based motor at different driving frequencies under 48 MV m-1.

Supplementary Video 4 | Process of pre-stretching using a custom-built rig.

**Extended Data Table 1 | Formulation of acrylic dielectric elastomers**





**Extended Data Fig. 1 | Comparison of molar stiffness function (MSF), cohesive energy (CE) and glass transition temperature (*Tg*) of homopolymers of commonly used acrylic monomers.**



**Extended Data Fig. 2 | Comparison of MSF, *Tg* and solubility parameter (δ) of common flexible long chain oligomers containing polyester, polyether and silicone as repeat units.**



**Extended Data Fig. 3 | Light transmittance comparison between VHBTM 4910 and BAC2.** Samples are 1 mm in thickness. And transmittance at wavelength of 550 nm that human eye is the most sensitivity to, was highlighted.



**Extended Data Fig. 4 | Comparison of toughness among VHBTM 4910 and new polyacrylates.**



**Extended Data Fig. 5 | Dielectric and mechanical properties of VHBTM4910, BAP and BAC series samples.** **a**, Stress-strain curves of VHBTM4910, BAP and BAC series samples. BAP will not be considered for the following analysis due to its inferior property. **b**, Frequency dependence of dielectric constant and dissipation factor (Tan δ) of VHBTM4910 and BAC series samples. **c**, Comparison of actuation sensitivity, dielectric constant and Young’s modulus of VHBTM4910 and BAC series samples. BAC1 will also be ignored due to its sticky feature. **d**, **e**, Temperature dependence of storage modulus (**d**) and mechanical loss (**e**). **f**, Cyclic test of VHBTM4910 and BAC series samples under uniaxial stretching.



**Extended Data Fig. 6 | Schematic diagram showing the actuation performance test platform for elastomers without pre-strain.**



**Extended Data Fig. 7 | Time dependent behavior of actuation area strain of VHBTM 4910 and BAC2 with 4×4 equiaxial pre-strain at 40 MV m-1.**



**Extended Data Fig. 8 | The schematic diagram of non-magnetic motor driving system.**



**Extended Data Fig. 9 | Theoretical analysis of the frequency-dependent behavior of the charging process for a simplified capacitor.** **a**, Schematic of the equivalent circuit model of non-magnetic motor, among which *U*s, S1, *R*1, *R*s, *R*p and *C*p stands for the power supply during charging process, relay, protective resistor in series, total resistor of wires and electrodes, and resistance and capacitance of the equivalent parallel model of elastomer film as a plate capacitor, respectively. *R*s, *R*p and *C*p are variable values since the dimension of elastomer film changes when an electric field is applied. **b**, Evolutions of loop current and voltage on *C*p under the actuation of a periodic square wave voltage *U*s at 50 Hz calculated by the Matlab/Simulink. **c**, The enlarged curves of content encircled by dashed line in Extended Data Fig. 9b. *τ* is the time constant for a transient process. **d**, Evolutions of current and voltage when the frequency is increased to 5000 Hz.



**Extended Data Fig. 10 | Simulation of soft motor.** **a**, Equivalent circuit of soft motor. Control signal for switches (Relays) was not displayed here. **b**, At 0.833 Hz and 48 MV m-1 (3.2 kV), from top to down, these curves represent on-off state of Switch A1, current from power supply, current through Switch A1 and voltage on phase A part of BAC2. **c**, At 17.61 Hz and 48 MV m-1 (3.2 kV), these curves represent on-off state of Switch A1, current form power supply, current through Switch A1 and voltage on phase A part of BAC2. **d**, Frequency dependence of maximum field strength on films of VHBTM4910 and BAC2. Grey points denote the frequency where soft motor did not rotate.

**Reference**

1. Wu, J. et al. Vulcanization kinetics of graphene/natural rubber nanocomposites. *Polymer* **54**, 3314-3323, (2013).
2. Acome, E. et al. Hydraulically amplified self-healing electrostatic actuators with muscle-like performance. *Science* **359**, 61-65, (2018).
3. Kang, J. et al. Tough and Water-Insensitive Self-Healing Elastomer for Robust Electronic Skin. *Adv. Mater.* **30**, 1706846, (2018).