

DIELECTRIC ELASTOMER ACTUATOR AND METHOD OF FORMING THE SAME

CROSS REFERENCE TO RELATED APPLICATION

[0001] This application claims the benefit of priority of Singapore application No. 10202403600R filed November 18, 2024, the contents of it being hereby incorporated by reference in its entirety for all purposes.

TECHNICAL FIELD

[0002] Various embodiments of this disclosure may relate to a dielectric elastomer actuator. Various embodiments of this disclosure may relate to a method of forming a dielectric elastomer actuator.

BACKGROUND

[0003] Soft actuators are highly deformable materials that can be activated by external stimuli to generate desired motions, offering improved flexibility, adaptability and compliance over traditional rigid robots and actuators. Among various soft actuators, dielectric elastomer actuators (DEAs) can achieve large deformation, fast response and high energy density, showing great potential in soft robotics applications. Typically, DEAs are composed of a dielectric elastomer (DE) layer sandwiched by two compliant electrodes, where an external voltage is applied to generate Maxwell stress for mechanical shape change. Generally, the voltage used to drive DEAs is up to several kilovolts, which severely limits its applications. This is because state-of-art DEs suffer from low relative permittivity (also known as dielectric constant). For example, the dielectric constant of three most commonly used DEs are 2.8 for polydimethylsiloxane (PDMS), 4.7 for acrylic elastomer (very high bond (VHB) tape), and 7.0 for polyurethane (PU). To obtain satisfying strain under relatively low voltage, a DE material with large dielectric constant and low elastic modulus, or simply high electromechanical sensitivity ($\frac{\text{dielectric constant}}{\text{elastic modulus}}$) is paramount. However, it is worth noting that a low elastic modulus will result in low effective work for practical DEA applications, and DEs with large dielectric constant and high breakdown field are highly favored.

[0004] Tremendous efforts have been made to increase the dielectric constant of DEs. Many studies have shown that introducing conductive or high permittivity fillers (e.g., metal nanomaterials, carbon-based microparticles, conjugated polymers, liquid metal, ionic liquid, gel particles, barium titanate, titanium dioxide, boron nitride, etc.) can readily enhance the dielectric performance of DEs. Yet, the introduction of fillers will usually lead to decreased breakdown field and elevated stiffness, which are not desirable for DEA applications. Meanwhile, the dielectric properties of the composite are still dominated by the DE matrix, according to the percolation theory. To address this fundamental issue, polar functional groups have been introduced into DEs to increase dielectric constant via dipole polarization effect. For instance, a high-performance polar fluorinated DE with a high dielectric constant (10.23 measured at 1 kHz) for a fast-moving soft robot has been developed. Also, a plasticized polyvinyl chloride-based DE with an excellent dielectric constant of 18.9 (measured at 1 kHz) formed from interfacial polarization of $C\equiv N$ has been reported. Despite these inspiring outcomes, there is still a lack of a higher permittivity dielectric elastomer.

SUMMARY

[0005] Various embodiments may relate to a dielectric elastomer actuator. The dielectric elastomer actuator may include a dielectric elastomer layer including a dielectric elastomer. The dielectric elastomer may include a zwitterionic segment, and a soft segment joined to the zwitterionic segment. The dielectric elastomer actuator may also include a first electrode and a second electrode. The dielectric elastomer actuator may further include one or more structural members. The dielectric elastomer actuator may be configured such that a force is applied on the dielectric elastomer layer due to Maxwell stress in response to a potential difference applied between the first electrode and the second electrode, thereby moving the one or more structural members.

[0006] Various embodiments may provide a method of forming a dielectric elastomer actuator. The method may include forming a dielectric elastomer layer including a dielectric elastomer. The dielectric elastomer may include a zwitterionic segment, and a soft segment joined to the zwitterionic segment. The method may also include forming or providing a first electrode. The method may further include forming or providing a second electrode. The method may additionally include providing or forming one or more structural members. The dielectric elastomer actuator may be configured such that a force is applied on the dielectric

elastomer layer due to Maxwell stress in response to a potential difference applied between the first electrode and the second electrode, thereby moving the one or more structural members.

BRIEF DESCRIPTION OF THE DRAWINGS

[0007] In the drawings, like reference characters generally refer to the same parts throughout the different views. The drawings are not necessarily drawn to scale, emphasis instead is generally being placed upon illustrating the principles of various embodiments. In the following description, various embodiments of the invention are described with reference to the following drawings.

FIG. 1 shows a general illustration of a dielectric elastomer actuator according to various embodiments.

FIG. 2 shows a general illustration of a method of forming a dielectric elastomer actuator according to various embodiments.

FIG. 3 shows examples of (i) soft segment monomer/precursors, (ii) diisocyanate monomers, and (iii) a zwitterionic monomer which may be used to form or synthesize the zwitterionic dielectric elastomer according to various embodiments.

FIG. 4 shows (i) synthesis of two examples of zwitterionic monomers; and (ii) synthesis of one example of a zwitterionic dielectric elastomer according to various embodiments.

FIG. 5A shows a plot of dielectric constant as a function of frequency (in Hertz or Hz) illustrating the dielectric constant- frequency curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments.

FIG. 5B shows a plot of dielectric loss as a function of frequency (in Hertz or Hz) illustrating the dielectric loss-frequency curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments.

FIG. 5C shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments.

FIG. 5D shows a chart comparing the modulus (in mega-Pascals or MPa), dielectric constant, and electromechanical sensitivity (in per mega-Pascals or MPa^{-1}) of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments.

FIG. 5E shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of a pristine sample as well as self-healed samples of zwitterionic dielectric elastomer ZPU-50 according to various embodiments.

FIG. 5F shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of a pristine sample as well as recycled samples of zwitterionic dielectric elastomer ZPU-50 according to various embodiments.

FIG. 6 is a schematic illustrating the operation of a buckling-mode actuator without pre-stretch according to various embodiments.

FIG. 7 is a schematic illustrating the operation of a bending-mode actuator with uniaxially pre-stretch according to various embodiments.

FIG. 8A shows a plot of displacement (in millimeters or mm) as a function of electric field (volt per micrometer or V/ μ m) comparing the electric field-induced displacement of a bending-mode actuator based on zwitterionic dielectric elastomer ZPU-50 according to various embodiments with the electric field-induced displacements of conventional bending-mode actuators based on very high bond (VHB) tape and Thermoplastic Polyurethane (TPU).

FIG. 8B shows a plot of displacement (in millimeters or mm) as a function of electric field (volt per micrometer or V/ μ m) comparing the electric field-induced displacement of a 1-stack bending-mode dielectric elastomer actuator and the electric field-induced displacement of a 5-stack bending-mode dielectric elastomer actuator according to various embodiments, with the inset showing a schematic of a robotic gripper based on the 1-stack bending-mode dielectric elastomer actuator according to various embodiments.

FIG. 8C is a magnified schematic of the robotic gripper shown in the inset of FIG. 8B according to various embodiments.

FIG. 8D shows a plot of displacement (in millimeters or mm) as a function of cycles illustrating long-term stability of actuation performance of the bending-mode dielectric elastomer actuator according to various embodiments.

FIG. 8E shows schematics of the operation of the robotic gripper based on the bending-mode dielectric elastomer actuator according to various embodiments.

FIG. 9A shows a plot of area strain (in percent or %) as a function of electric field (in volts per micrometer or V/ μ m) comparing the area-strain performance of a buckling-mode actuator based on zwitterionic dielectric elastomer ZPU-50 according to various embodiments with the area-strain performances of conventional buckling-mode actuators based on very high bond

(VHB) tape and Thermoplastic Polyurethane (TPU), with the inset illustrating the operation of the buckling-mode actuator according to various embodiments.

FIG. 9B shows schematics illustrating the operation of a weight-lifting device based on the buckling-mode actuator according to various embodiments.

FIG. 10A shows a schematic illustrating the coupling of a robotic micro-gripper according to various embodiments for simultaneous sensing and actuation.

FIG. 10B shows a schematic illustrating the operation states of the robotic micro-gripper according to various embodiments.

FIG. 10C shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU), when moving up and down.

FIG. 10D shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU) , when turned on and turned off.

FIG. 10E shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU) at different states as illustrated in FIG. 10B.

DESCRIPTION

[0008] The following detailed description refers to the accompanying drawings that show, by way of illustration, specific details and embodiments in which the invention may be practiced. These embodiments are described in sufficient detail to enable those skilled in the art to practice the invention. Other embodiments may be utilized, and structural, logical, and electrical changes may be made without departing from the scope of the invention. The various embodiments are not necessarily mutually exclusive, as some embodiments can be combined with one or more other embodiments to form new embodiments.

[0009] Features that are described in the context of an embodiment may correspondingly be applicable to the same or similar features in the other embodiments. Features that are described in the context of an embodiment may correspondingly be applicable to the other embodiments, even if not explicitly described in these other embodiments. Furthermore, additions and/or combinations and/or alternatives as described for a feature in the context of an embodiment may correspondingly be applicable to the same or similar feature in the other embodiments. For instance, embodiments described in the context of one of the dielectric elastomer actuators are analogously valid for the other dielectric elastomer actuators. Similarly, embodiments described in the context of a method are analogously valid for a dielectric elastomer actuator, and vice versa.

[0010] In the context of various embodiments, the articles “a”, “an” and “the” as used with regard to a feature or element include a reference to one or more of the features or elements.

[0011] In the context of various embodiments, the term “about” or “approximately” as applied to a numeric value encompasses the exact value and a reasonable variance, e.g., within 10% of the specified value.

[0012] As used herein, the term “and/or” includes any and all combinations of one or more of the associated listed items.

[0013] By “comprising” it is meant including, but not limited to, whatever follows the word “comprising”. Thus, use of the term “comprising” indicates that the listed elements are required or mandatory, but that other elements are optional and may or may not be present.

[0014] By “consisting of” it is meant including, and limited to, whatever follows the phrase “consisting of”. Thus, the phrase “consisting of” indicates that the listed elements are required or mandatory, and that no other elements may be present.

[0015] FIG. 1 shows a general illustration of a dielectric elastomer actuator according to various embodiments. The dielectric elastomer actuator may include a dielectric elastomer layer 102 including a dielectric elastomer. The dielectric elastomer may include a zwitterionic segment, and a soft segment joined to the zwitterionic segment. The dielectric elastomer may alternatively be referred to as zwitterionic elastomer or zwitterionic polyurethane. The dielectric elastomer actuator may also include a first electrode 104a and a second electrode 104b. The dielectric elastomer actuator may further include one or more structural members 106. The dielectric elastomer actuator may be configured such that a force is applied on the dielectric elastomer layer 102 due to Maxwell stress in response to a potential difference

applied between the first electrode 104a and the second electrode 104b, thereby moving the one or more structural members 106.

[0016] In other words, various embodiments may relate to a dielectric elastomer actuator having a layer 102 made of or including a dielectric elastomer molecule having a zwitterionic segment and a soft segment. The dielectric elastomer actuator may also include a first electrode 104a, a second electrode 104b as well as one or more structural members 106. The layer 102 may be applied with a force (e.g., compressive force) arising from Maxwell stress in response to the potential difference applied between the first electrode 104a and the second electrode 104b, thereby actuating the one or more structural members 106.

[0017] For avoidance of doubt, FIG. 1 is intended to illustrate various components of a dielectric elastomer actuator according to various embodiments, and are not intended to limit, for instance, the shape, dimensions, sizes, arrangement, orientation etc. of the various components.

[0018] The incorporation of zwitterionic moieties may provide the dielectric elastomer layer 102 a large dielectric constant and a high breakdown field. The dielectric elastomer layer 102 may exhibit outstanding area strain and high energy density under a low electric field. The dielectric elastomer including the zwitterionic segment may exhibit significantly superior results compared to commercial dielectric elastomers, such as Very High Bond (VHB) elastomer and Thermoplastic Polyurethane (TPU), under the same testing conditions. The dielectric elastomer layer 102 may actuate over 1000 cycles, while maintaining over 80% of the initial displacement. The dielectric elastomer layer 102 may require a low driving voltage or potential difference, and the potential difference (applied to the dielectric elastomer layer 102 via the first electrode 104a and the second electrode 104b) may be a voltage equal to or lower than 800 V, e.g., equal to or lower than 500V.

[0019] The Maxwell stress may arise due to presence of an electrostatic or electric field resulting from the potential difference applied between the first electrode 104a and the second electrode 104b. In various embodiments, the potential difference (applied) may result in compression of the dielectric elastomer layer 102 due to Maxwell stress to cause strain of the dielectric elastomer layer 102, thereby resulting in deformation of the dielectric elastomer layer 102. In contrast, some conventional actuators may employ different working mechanisms, e.g., thermal induced shape memory effect, electrochemical capacitance effect where mobile ions

migrate across the membrane layer and induce different volumetric expansion at the electrodes, or chemical effect in the form of pH changes to induce molecular structural changes.

[0020] The dashed lines in FIG. 1 represent the direct or indirect coupling between the dielectric elastomer layer 102 and the one or more structural members 106. In various embodiments, the one or more structural members 106 may be in contact with the dielectric elastomer layer 102, or may be attached, coupled or joined to the dielectric elastomer layer 102 via the first electrode 104a, the second electrode 104b, or via any suitable intermediate component, member or layer.

[0021] In various embodiments, the first electrode 104a and the second electrode 104b may be in contact with the dielectric elastomer layer 102. In various embodiments, the first electrode 104a may be a flexible or compliant electrode. In various embodiments, the second electrode 104b may be a flexible or compliant electrode. In various embodiments, the first electrode 104a and the second electrode 104b may be on opposite sides of the dielectric elastomer layer 102. The first electrode 104a and/or the second electrode 104b may include any suitable electrically conductive material(s), for instance, graphene, carbon nanotubes, activated carbon, polyaniline and/or polypyrrole.

[0022] In various embodiments, the dielectric elastomer actuator may be a weight-lifting device, and the one or more structural members 106 may be or may include a load, or a hook or attachment member for suspending or carrying the load.

[0023] In various embodiments, the dielectric elastomer actuator shown may be a robotic gripper (e.g., a robotic micro-gripper), and the one or more structural members 106 may be or may include a pair of grips. The robotic gripper or micro-gripper may be able to achieve sensing and actuation simultaneously under low voltage, thanks to the stable signal from capacitive sensing.

[0024] In various embodiments, the dielectric elastomer layer 102 may be configured to act as a capacitive sensor, e.g., a fringe field effect capacitive sensor. A fringing electric field may be generated when a sensing voltage is applied between the first electrode 104a and the second electrode 104b. The first electrode 104a and the second electrode 104b may be in electrical connection with a sensing circuit (e.g., an inductance-capacitance-resistance (LCR) meter). In various embodiments, the sensing voltage may be provided or generated by a same voltage source configured to provide or generate the potential difference for actuation, while in various other embodiments, the sensing voltage may be provided or generated by a voltage source

different from a voltage source configured to provide or generate the potential difference for actuation. In various embodiments, the one or more structural members 106 may be electrically conductive and in electrical connection with the first electrode 104a and/or the second electrode 104b. The one or more structural members 106 may, for instance, be the pair of grips. The fringing electric field may be generated by the first electrode 104a, the second electrode 104b and/or the one or more structural members 106 when the sensing voltage is applied between the first electrode 104a and the second electrode 104b. When different objects having different dielectric constants are positioned within the fringing electric field, the capacitance measured by the sensing circuit may change.

[0025] In various embodiments, the dielectric elastomer may further include one or more diisocyanate-based segments. The soft segment may be joined or linked to the zwitterionic segment via the one or more diisocyanate-based segments. Each of the one or more diisocyanate-based segments may be formed from a diisocyanate monomer, for instance, hexamethylene diisocyanate (HDI), 1,4-diisocyanatobutane, isophorone diisocyanate (IPDI), toluylene diisocyanate, 1,3-phenylene diisocyanate, diphenylmethane 4,4'-diisocyanate, or 1,4-phenylene diisocyanate.

[0026] The zwitterionic segment may be formed from a zwitterion monomer, i.e., a molecule having both cationic (positively charged) and anionic (negatively charged) groups. In various embodiments, the zwitterionic monomer may have a branch having an alkyl (R_4) and a functional group including a sulfonate (SO_3^-), a carboxylate (CO_2^-) or a phosphite (PO_3^-). In various embodiments, the alkyl (R_4) may be bonded to a (cationic) group including a nitrogen atom and three R groups (i.e., R_1 , R_2 and R_3) joined to the nitrogen atom. R_1 and R_2 may each be a hydroxyalkyl, a vinylalkyl, or an acrylate etc. R_3 may be an alkyl, a hydroxyalkyl, a vinylalkyl, or an acrylate etc. The zwitterion monomer may, for instance, be sulfobetaine3-diol (SB3-diol) zwitterionic monomer or sulfobetaine4-diol (SB4-diol) zwitterionic monomer.

[0027] The soft segment may be formed from polytetramethylene ether glycol (PTMG), polycaprolactone diol, poly(propylene glycol) or polyethylene glycol.

[0028] FIG. 2 shows a general illustration of a method of forming a dielectric elastomer actuator according to various embodiments. The method may include, in 202, forming a dielectric elastomer layer including a dielectric elastomer. The dielectric elastomer may include a zwitterionic segment, and a soft segment joined to the zwitterionic segment. The method may

also include, in 204, forming or providing a first electrode. The method may further include, in 206, forming or providing a second electrode. The method may additionally include, in 208, providing or forming one or more structural members. The dielectric elastomer actuator may be configured such that a force is applied on the dielectric elastomer layer due to Maxwell stress in response to a potential difference applied between the first electrode and the second electrode, thereby moving the one or more structural members.

[0029] In other words, various embodiments may relate to forming a dielectric elastomer actuator as described herein. The dielectric elastomer actuator may include a dielectric elastomer layer, a first electrode, a second electrode and one or more structural members.

[0030] For avoidance of doubt, FIG. 2 is intended to illustrate various steps according to various embodiments, and is not intended to limit the sequence of the various steps. For instance, step 202 may occur before, after or at the same time as step 204.

[0031] In various embodiments, the dielectric elastomer may further include one or more diisocyanate-based segments. The soft segment may be joined or linked to the zwitterionic segment via the one or more diisocyanate-based segments.

[0032] In various embodiments, the dielectric elastomer may be formed via a polymerization process. In various embodiments, the dielectric elastomer may be formed by mixing a zwitterionic monomer, a soft segment monomer/precursor and a solvent (e.g., dimethylformamide (DMF), dimethyl sulfoxide, tetrahydrofuran and/or dimethylacetamide) to form a mixture, heating the mixture at a first predetermined temperature (e.g., 80 °C), and adding one or more diisocyanate monomers and a catalyst (e.g., dibutyltin dilaurate (DBTDL), triethylenediamine and/or stannous octoate) to the mixture after the mixture has cooled from the first predetermined temperature to a second predetermined temperature (e.g., room temperature or 20 °C).

[0033] The one or more diisocyanate monomers may be or may include any suitable diisocyanate monomers. The one or more diisocyanate monomers may, for instance, be selected from a group consisting of hexamethylene diisocyanate (HDI), 1,4-diisocyanatobutane, isophorone diisocyanate (IPDI), toluylene diisocyanate, 1,3-phenylene diisocyanate, diphenylmethane 4,4'-diisocyanate, and 1,4-phenylene diisocyanate.

[0034] The zwitterionic monomer may be any suitable zwitterionic monomer. In various embodiments, the zwitterionic monomer may have a branch having an alkyl (R_4) and a functional group including a sulfonate (SO_3^-), a carboxylate (CO_2^-) or a phosphite (PO_3^-). In

various embodiments, the alkyl (R_4) may be bonded to a (cationic) group including a nitrogen atom and three R groups (i.e., R_1 , R_2 and R_3) joined to the nitrogen atom. R_1 and R_2 may each be a hydroxyalkyl, a vinylalkyl, or an acrylate etc. R_3 may be an alkyl, a hydroxyalkyl, a vinylalkyl, or an acrylate etc. The zwitterion monomer may, for instance, be sulfobetaine3-diol (SB3-diol) zwitterionic monomer or sulfobetaine4-diol (SB4-diol) zwitterionic monomer.

[0035] The soft segment monomer/precursor may be any monomer/precursor suitable to react with the zwitterion monomer and optionally, the one or more diisocyanate monomers. The soft segment monomer/precursor may, for instance, be polytetramethylene ether glycol (PTMG), polycaprolactone diol, poly(propylene glycol) or polyethylene glycol.

[0036] In various embodiments, the zwitterionic monomer may be formed by mixing a first precursor, a second precursor and a further solvent (e.g., acetonitrile) to form a solution, and heating the solution to form the zwitterionic monomer. The first precursor and the second precursor may be any precursors suitable for forming the zwitterionic monomer required. For instance, the first precursor may be N-butyldiethanolamine, and the second precursor may be 1,3-propane sultone (for forming sulfobetaine3-diol (SB3-diol) zwitterionic monomer) or 1,4-butane sultone (for forming sulfobetaine4-diol (SB4-diol) zwitterionic monomer).

[0037] In various embodiments, the dielectric elastomer layer may be formed by depositing the dielectric elastomer using a suitable deposition process selected from blade coating, spin coating, dip coating, spray coating, screen printing, inkjet printing and three-dimensional (3D) printing. In various embodiments, the dielectric elastomer layer may be formed by depositing the dielectric elastomer on the first electrode or the second electrode. In various other embodiments, the dielectric elastomer layer may be formed by depositing the dielectric elastomer on a substrate, forming the first electrode in contact with the dielectric elastomer layer, removing the substrate, and forming the second electrode in contact with the dielectric elastomer layer.

[0038] In various embodiments, the potential difference may result in compression of the dielectric elastomer layer due to Maxwell stress to cause strain of the dielectric elastomer layer, thereby resulting in deformation of the dielectric elastomer layer.

[0039] In various embodiments, the dielectric elastomer layer may also be configured to act as a capacitive sensor.

[0040] In various embodiments, the one or more structural members may be or may include a pair of grips, and the dielectric elastomer actuator may be a robotic gripper, e.g., a robotic

micro-gripper. In various other embodiments, the one or more structural members may be or may include be a load, or a hook or attachment member for suspending or carrying the load, and the dielectric elastomer actuator may be a weight-lifting device.

[0041] Rather than depending on weak/moderate dipole moment to produce incremental increase in dielectric constant, the inherently strong dipole moment of zwitterions can lead to a drastic increase in the dielectric constant in dielectric elastomers (DEs). Zwitterions possess enormous dielectric constant (e.g., 200 – 270) amongst reported soft materials. Further, very high dielectric constant (~ 150 @ 1 kHz) has also been reported for the corresponding homopolymer. The copolymerizing of zwitterionic moieties into elastomers may confer the resultant zwitterionic dielectric elastomer (DE) a large dielectric constant, a high breakdown field, as well as a maximum electrostatic energy density higher than that of existing DEs. The dielectric elastomer actuator (DEA) according to various embodiments may exhibit outstanding area strain and high energy density under low electric fields, and may be significantly superior to commercial DEs such as VHB and TPU. Notably, the dielectric elastomer actuator (DEA) may actuate over 1000 cycles, while maintaining over 80% of the initial displacement under a low driving voltage of only 500 V, demonstrating superior results over existing dielectric elastomer actuators. The grippers according to various embodiments may achieve sensing and actuation simultaneously under low operating voltage, thanks to the stable signal generated from capacitive sensing.

[0042] The zwitterionic elastomer may have the following advantages: (i) exhibits good mechanical properties such as large stretchability, high tensile strength, low modulus and superior toughness; (ii) can self-heal and be recycled by hot-pressing or solution recycling; (iii) benefits from the large dipole moment of zwitterions; the dielectric constant of the zwitterionic elastomer is significantly larger than existing DE materials (for example, zwitterionic liquid has a dipole moment of 35 - 41 D, as compared to 3.6 D for $C\equiv N$ in acetonitrile, and 4.47 D for sulfone in dimethylsulfone; the dielectric constant may also be modulated by tuning the distance between the cation and anion in the zwitterion); (iv) may possess a breakdown field higher than most reported DE materials, as the zwitterionic elastomer may be a pure elastomer free of fillers; (v) can be actuated by a low voltage with satisfactory actuation performance, due to high dielectric constant and satisfactory elastic modulus; (vi) shows excellent stability when actuated, with no significant decrease in displacement from its low viscoelasticity; (vii) can show stable capacitive sensing signals when operated under a low voltage; and (viii) has

anti-fouling and anti-bacteria characteristics from its hydrophilicity and repulsion of fouling agents.

[0043] Different components such as soft segment monomer/precursors, diisocyanates, zwitterionic monomers, acrylates, etc. may be used to synthesize the zwitterionic dielectric elastomers. FIG. 3 shows examples of (i) soft segment monomer/precursors, (ii) diisocyanate monomers, and (iii) a zwitterionic monomer which may be used to form or synthesize the zwitterionic dielectric elastomer according to various embodiments. The soft segment monomer/precursor may, for instance, be polytetramethylene ether glycol (PTMG), polycaprolactone diol, poly(propylene glycol) or polyethylene glycol. The diisocyanate monomers may, for instance, be hexamethylene diisocyanate (HDI), 1,4-diisocyanatobutane, isophorone diisocyanate (IPDI), toluylene diisocyanate, 1,3-phenylene diisocyanate, diphenylmethane 4,4'-diisocyanate, or 1,4-phenylene diisocyanate. The zwitterionic monomer may have a branch having an alkyl (R_4) and a functional group including a sulfonate (SO_3^-), a carboxylate (CO_2^-) or a phosphite (PO_3^-). In various embodiments, the alkyl (R_4) may be bonded to a (cationic) group including a nitrogen atom and three R groups (i.e., R_1 , R_2 and R_3) joined to the nitrogen atom. R_1 and R_2 may each be a hydroxyalkyl, a vinylalkyl, or an acrylate etc. R_3 may be an alkyl, a hydroxyalkyl, a vinylalkyl, or an acrylate etc. The zwitterion monomer may, for instance, be sulfobetaine3-diol (SB3-diol) zwitterionic monomer or sulfobetaine4-diol (SB4-diol) zwitterionic monomer. For avoidance of doubt, FIG. 3 is intended to provide examples of component choices which may be used, and in various embodiments, materials or components not listed in FIG. 3 may be used to form or synthesize the zwitterionic dielectric elastomer according to various embodiments. Additionally, the following examples provided below are not intended to be limiting.

[0044] Example 1: Fabrication of zwitterionic monomers and zwitterionic dielectric elastomers (zwitterionic polyurethanes or ZPUs)

[0045] FIG. 4 shows (i) synthesis of two examples of zwitterionic monomers; and (ii) synthesis of one example of a zwitterionic dielectric elastomer according to various embodiments. The molecular ratio of zwitterion monomers and soft segment monomers/precursors may be tuned to obtain different zwitterionic elastomers with different dielectric and mechanical properties. With reference to (i) of FIG. 4, for the synthesis of sulfobetaine3-diol (SB3-diol) zwitterionic monomer, 1,3-propane sultone (11.966 g, 98 mmol), N-butyldiethanolamine (17.38 g, 108 mmol) and acetonitrile (20 mL) were dissolved together

in a round-bottom flask. The mixture was heated to reflux at 110 °C and stirred for 24 hours under argon atmosphere. After the reaction, the product was washed with diethyl ether for three times. For the sulfobetaine4-diol (SB4-diol) zwitterionic monomer, 1,4-butane sultone (13.344 g, 98 mmol), N-butyldiethanolamine (17.38 g, 108 mmol) and acetonitrile (20 mL) were dissolved together in a round-bottom flask. The mixture was heated to reflux at 110 °C and stirred for 24 hours under argon atmosphere. After the reaction, the product was washed with diethyl ether for three times.

[0046] Before polymerization, the soft segment precursor/monomer (e.g., polytetramethylene ether glycol (PTMG)) and zwitterion monomer (e.g., SB3-diol/SB4-diol) were vacuum dried overnight at 120 °C to remove any moisture. The PTMG and SB3-diol/SB4-diol may be mixed at different molecular ratios (e.g., 75 : 25, 50 : 50 and 25 : 75). The zwitterionic polyurethanes formed with different molecular ratios of PTMG : SB4-diol may be named as ZPU-25 (ratio of 75 : 25), ZPU-50 (ratio of 50 : 50), and ZPU-75 (ratio of 25 : 75). For example, to form ZPU-50, PTMG (1.0 g, 1.0 mmol), SB4-diol (0.297 g, 1.0 mmol) and dimethylformamide DMF (3.5 mL) were first added into a glass vial and heated at 80 °C with stirring and argon bubbling for 1 hour. The mixture was cooled down to room temperature, and diisocyanate monomers (e.g., isophorone diisocyanate IPDI (0.233 mL, 1.10 mmol), hexamethylene diisocyanate HDI (0.177 mL, 1.10 mmol)) as well as dibutyltin dilaurate DBTDL (40 µL) were added into the flask. The reaction mixture was stirred at 80 °C for 24 hours under argon atmosphere.

[0047] Example 2: Dielectric properties and mechanical properties of zwitterionic dielectric elastomers

[0048] In order to study the dielectric and mechanical properties of different zwitterionic polyurethanes (ZPUs), the dielectric constant, dielectric loss and stress-strain curves may be plotted based on measurements.

[0049] FIG. 5A shows a plot of dielectric constant as a function of frequency (in Hertz or Hz) illustrating the dielectric constant- frequency curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments. For the zwitterionic dielectric elastomers (i.e., ZPU-25, ZPU-50, ZPU-75) formed from SB4-diol with increasing zwitterion content (25, 50 and 75 atomic percentages (at. %)), the zwitterionic dielectric elastomers exhibited growing trend of high dielectric constant with increasing zwitterionic

content over all measured frequencies, with values of 14.2 for ZPU-25, 36.7 for ZPU-50 and 64.6 for ZPU-75 at 1 kHz.

[0050] FIG. 5B shows a plot of dielectric loss as a function of frequency (in Hertz or Hz) illustrating the dielectric loss-frequency curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments. Despite the high dielectric constant of the zwitterionic dielectric elastomers, dielectric loss was kept at a relatively low level (0.251 for ZPU-25, 0.393 for ZPU-50 and 0.646 for ZPU-75 at 1 kHz) albeit the increasing trend with higher zwitterion content.

[0051] FIG. 5C shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments. FIG. 5D shows a chart comparing the modulus (in mega-Pascals or MPa), dielectric constant, and electromechanical sensitivity (in per mega-Pascals or MPa^{-1}) of zwitterionic dielectric elastomers with different zwitterion content according to various embodiments. Due to the dynamic ionic interactions associated with the zwitterionic moieties, increasing zwitterion content led to higher Young's modulus (Y), lower stretchability and eventually lower toughness based on tensile measurements. In addition, all three zwitterionic dielectric elastomers (ZPUs) exhibited strain-hardening behavior, which may be required for high-performance DEAs.

[0052] The dynamic ionic interaction from zwitterion may provide ZPU-50 with outstanding self-healing characteristics and recyclability. The self-healing property of ZPU-50 was evaluated from the tensile test results of the original sample and healed samples (heated at 80 °C for different periods after bisecting). FIG. 5E shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of a pristine sample as well as self-healed samples of zwitterionic dielectric elastomer ZPU-50 according to various embodiments. FIG. 5E shows that as healing time increased, ZPU-50 may begin to restore its mechanical strength, and a 24h healing time may lead to an excellent 92.48%, 86.29% and 98.66% recovery in stretchability, tensile strength and toughness, respectively. The tensile stress-strain curves of ZPU-50 film before and after solution recycle process were measured to demonstrate the outstanding recyclability. FIG. 5F shows a plot of stress (in mega-Pascals or MPa) as a function of strain (in percent or %) illustrating the stress-strain curves of a pristine sample as well as recycled samples of zwitterionic dielectric elastomer ZPU-50 according to various embodiments. The tensile test showed ZPU-50 may retain similar

mechanical performance even after a third time of solution recycling, with only slight decrease in tensile strength and break elongation strain.

[0053] Example 3: Actuation performance of zwitterionic dielectric elastomer actuators

[0054] The actuation performance of the zwitterionic dielectric elastomer is characterized by two different kinds of actuators, which are buckling device without pre-stretch and bending device with uniaxially pre-stretch. FIG. 6 is a schematic illustrating the operation of a buckling-mode actuator without pre-stretch according to various embodiments. The device may include a stacked arrangement of the first electrode 604a, the second electrode 604b, and the dielectric elastomer layer 602 between two fixed frames or anchors 608a, 608b. In the absence of an applied potential difference between the first electrode 604a and the second electrode 604b, the first electrode 604a, the second electrode 604b, and the dielectric elastomer layer 602 between the first electrode 604a and the second electrode 604b may be in an unbuckled state. When the potential difference is applied between the first electrode 604a and the second electrode 604b, the opposing charges in the first electrode 604a and the second electrode 604b may compress the dielectric elastomer layer 602, causing the dielectric elastomer layer 602 and the electrodes 604a, 604b to be in a buckled state.

[0055] FIG. 7 is a schematic illustrating the operation of a bending-mode actuator with uniaxially pre-stretch according to various embodiments. In the absence of an applied potential difference between the first electrode 704a and the second electrode 704b, the first electrode 704a, the second electrode 704b, and the dielectric elastomer layer 702 between the first electrode 704a and the second electrode 704b may be in an unbent state. When the potential difference is applied between the first electrode 704a and the second electrode 704b, the opposing charges in the first electrode 704a and the second electrode 704b may compress the dielectric elastomer layer 702, causing the dielectric elastomer layer 702 and the electrodes 704a, 704b to be in a bent state.

[0056] For both devices shown in FIGS. 6 – 7, actuators based on zwitterionic dielectric elastomer exhibited significantly superior actuation performance compared to actuators based on commercial dielectric elastomers, such as TPU and VHB tape.

[0057] FIG. 8A shows a plot of displacement (in millimeters or mm) as a function of electric field (volt per micrometer or V/ μ m) comparing the electric field-induced displacement of a bending-mode actuator based on zwitterionic dielectric elastomer ZPU-50 according to various embodiments with the electric field-induced displacements of conventional bending-mode

actuators based on very high bond (VHB) tape and Thermoplastic Polyurethane (TPU). FIG. 8B shows a plot of displacement (in millimeters or mm) as a function of electric field (volt per micrometer or $V/\mu m$) comparing the electric field-induced displacement of a 1-stack bending-mode dielectric elastomer actuator and the electric field-induced displacement of a 5-stack bending-mode dielectric elastomer actuator according to various embodiments, with the inset showing a schematic of a robotic gripper based on the 1-stack bending-mode dielectric elastomer actuator according to various embodiments. FIG. 8C is a magnified schematic of the robotic gripper shown in the inset of FIG. 8B according to various embodiments. The robotic gripper may include a first electrode 804a, a second electrode 804b, and a dielectric elastomer layer 802 between the first electrode 804a and the second electrode 804b. The robotic gripper may also include a pair of grips 806a, 806b attached to the electrodes 804a, 804b. The first grip 806a may be attached to the first electrode 804a, while the second grip 806b may be attached to the second electrode 804b.

[0058] FIG. 8D shows a plot of displacement (in millimeters or mm) as a function of cycles illustrating long-term stability of actuation performance of the bending-mode dielectric elastomer actuator according to various embodiments. FIG. 8E shows schematics of the operation of the robotic gripper based on the bending-mode dielectric elastomer actuator according to various embodiments.

[0059] FIG. 9A shows a plot of area strain (in percent or %) as a function of electric field (in volts per micrometer or $V/\mu m$) comparing the area-strain performance of a buckling-mode actuator based on zwitterionic dielectric elastomer ZPU-50 according to various embodiments with the area-strain performances of conventional buckling-mode actuators based on very high bond (VHB) tape and Thermoplastic Polyurethane (TPU), with the inset illustrating the operation of the buckling-mode actuator according to various embodiments. FIG. 9B shows schematics illustrating the operation of a weight-lifting device based on the buckling-mode actuator according to various embodiments. The weight-lifting device may include a dielectric elastomer layer 902, a first electrode 904a on a first side of the dielectric elastomer layer 902, and a second electrode 904b on a second side of the dielectric elastomer layer 902 opposite the first side. The second electrode 904b may be attached to a load 912. When a potential difference is applied between the first electrode 904a and the second electrode 904b, the load 912 may be lowered. In the absence of the applied potential difference, the load 912 may be lifted up.

[0060] Example 5: Proprioceptive gripper based on zwitterionic dielectric elastomers

[0061] One way to achieve proprioception in grippers is via the use of fringe field effect capacitive sensors. Since dielectric elastomer actuators (DEAs) are capacitors in nature, it may be possible for DEAs to simultaneously sense and actuate during operation. In the case of micro-grippers, this simultaneous sensing and actuation capability may be even more important due to the size constraint of the micro-grippers, which makes installation of other sensors difficult. Moreover, capacitive sensing may be very sensitive to electromagnetic interference, particularly so in the presence of high voltage. FIG. 10A shows a schematic illustrating the coupling of a robotic micro-gripper according to various embodiments for simultaneous sensing and actuation. The robotic micro-gripper may include a dielectric elastomer layer 1002, a first electrode 1004a on a first side of the dielectric elastomer layer 1002, and a second electrode 1004b on a second side of the dielectric elastomer layer 1002 opposite the first side. The robotic micro-gripper may also include grips 1006a, 1006b attached to the electrodes 1004a, 1004b. The electrodes 1004a, 1004b may be electrically connected via electrical wires to a high voltage source 1008 for actuation. The resistance of the electrical wires may be represented by resistance elements 1010a, 1010b. The electrodes 1004a, 1004b may be electrically connected to an inductance-capacitance-resistance (LCR) meter 1012. The capacitor element 1014 may be connected to the electrodes 1004a, 1004b and the LCR meter 1012 to remove the direct current (DC) component of the sensing voltage before being measured by the LCR meter 1012.

[0062] FIG. 10B shows a schematic illustrating the operation states of the robotic micro-gripper according to various embodiments. The robotic micro-gripper may be at the “up” positions in operational states “1” and “4”, and may be at the “down” position in operational states “2”, “3”, “5” and “6”. Also, the sensing voltage may be turned on in operational states “1”, “2” and “6”, and turned off in operational states “3”, “4” and “6”. FIG. 10C shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU), when moving up and down. FIG. 10D shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU), when turned on and turned off. FIG. 10E

shows plots of capacitance (in arbitrary units or a.u.) as a function of time (in seconds or s) illustrating the capacitive sensing signals of a robotic micro-gripper based on the zwitterionic dielectric elastomer (ZPU-50) according to various embodiments and a robotic gripper based on commercial Thermoplastic Polyurethane (TPU) at different states as illustrated in FIG. 10B.

[0063] Table 1 below shows a comparison of dielectric elastomers and other means of actuation.

Table 1

	Advantages	Disadvantages
Miniaturized motors (not soft)	• Reliable • Robust • Large design	• Not very compatible with microfabrication • High cost
Piezoelectric	• Highly controllable movement • Fast response time • Low power consumption	• Limited deformation • High cost • Fragility
Magnetic	• Wireless control • High output force • Fast response time	• High cost • Interference of other magnetic fields • Limited deformation • Limited substrates
Thermal	• Simple design • Easy to manufacture	• Low power density • Sensitive to temperature fluctuations • Slow response time
Dielectric elastomer	• Large power density • High deformation and strain	• High voltage requirement • Short lifespan

	• Lightweight • Simple and low-cost design	
Electrostatic	• Fast response time	• Limited deformation • Easy to manufacture • Short lifespan • Sensitive to humidity
Chemical	• Low cost • High energy density	• Limited lifespan • Safety concerns • Impact on the environment
Optical	• Wireless control • Low power consumption • Fast response time	• Sensitivity to ambient light • High cost

[0064] Various embodiments based on zwitterionic dielectric elastomer may be able to address the shortcomings of dielectric elastomers. The high voltage requirement of dielectric elastomers may be drastically reduced, while the limited lifespan of dielectric elastomers may be improved. Typical DEAs usually require driving voltage of several kilovolts and have limited lifespan. However, various embodiments based on zwitterionic dielectric elastomer may be able to significantly reduce the driving voltage required, and may have improved cycling stability.

Claims

1. A dielectric elastomer actuator comprising:
 - a dielectric elastomer layer comprising a dielectric elastomer, the dielectric elastomer comprising a zwitterionic segment, and a soft segment joined to the zwitterionic segment;
 - a first electrode;
 - a second electrode; and
 - one or more structural members;wherein the dielectric elastomer actuator is configured such that a force is applied on the dielectric elastomer layer due to Maxwell stress in response to a potential difference applied between the first electrode and the second electrode, thereby moving the one or more structural members.
2. The dielectric elastomer actuator according to claim 1,
 - wherein the potential difference results in compression of the dielectric elastomer layer due to Maxwell stress to cause strain of the dielectric elastomer layer, thereby resulting in deformation of the dielectric elastomer layer.
3. The dielectric elastomer actuator according to claim 1 or claim 2,
 - wherein the one or more structural members comprise a pair of grips; and
 - wherein the dielectric elastomer actuator is a robotic gripper.
4. The dielectric elastomer actuator according to claim 1 or claim 2,
 - wherein the one or more structural members comprise a load; and
 - wherein the dielectric elastomer actuator is a weight-lifting device.
5. The dielectric elastomer actuator according to claim 1,
 - wherein the dielectric elastomer layer is also configured to act as a capacitive sensor.

6. The dielectric elastomer actuator according to claim 1,
wherein the dielectric elastomer further comprises one or more diisocyanate-based segments.
7. The dielectric elastomer actuator according to claim 1,
wherein the soft segment is formed from polytetramethylene ether glycol (PTMG), polycaprolactone diol, poly(propylene glycol) or polyethylene glycol.
8. The dielectric elastomer actuator according to claim 1,
wherein the zwitterionic segment is formed from a zwitterion monomer.
9. The dielectric elastomer actuator according to claim 8,
wherein the zwitterion monomer has a branch having an alkyl and a functional group including a sulfonate (SO_3^-), a carboxylate (CO_2^-) or a phosphite (PO_3^-).
10. The dielectric elastomer actuator according to claim 1,
wherein the potential difference is a voltage equal to or lower than 800 V.
11. A method of forming a dielectric elastomer actuator, the method comprising:
forming a dielectric elastomer layer comprising a dielectric elastomer, the dielectric elastomer comprising a zwitterionic segment, and a soft segment joined to the zwitterionic segment;
forming a first electrode;
forming a second electrode; and
providing one or more structural members;
wherein the dielectric elastomer actuator is configured such that a force is applied on the dielectric elastomer layer due to Maxwell stress in response to a potential difference applied between the first electrode and the second electrode, thereby moving the one or more structural members.

12. The method according to claim 11,
wherein the dielectric elastomer further comprises one or more diisocyanate-based segments.
13. The method according to claim 12,
wherein the dielectric elastomer is formed by:
mixing a zwitterionic monomer, a soft segment precursor and a solvent to form a mixture;
heating the mixture at a first predetermined temperature; and
adding one or more diisocyanate monomers and a catalyst to the mixture after the mixture has cooled from the first predetermined temperature to a second predetermined temperature.
14. The method according to claim 13,
wherein the zwitterionic monomer is formed by:
mixing a first precursor, a second precursor and a further solvent to form a solution; and
heating the solution to form the zwitterionic monomer.
15. The method according to any one of claims 11 to 14,
wherein the dielectric elastomer layer is formed by depositing the dielectric elastomer using a suitable deposition process selected from blade coating, spin coating, dip coating, spray coating, screen printing, inkjet printing and three-dimensional (3D) printing.