Peer Review File

Polyvinyl chloride-based dielectric elastomer with high permittivity and low viscoelasticity for actuation and sensing

Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
In this manuscript, the authors produced a polyvinyl chloride (PVC)-based elastomer by the introduction of cyanoethyl cellulose (CEC) into a plasticized PVC. The PVC-based elastomer shows high permittivity, low viscoelasticity, but the dielectric loss is not satisfactory. Besides a few issues need to be clarify.

1、It is unclear how the plasticized PVC named by the authors is processable. It is confusing that the authors named the matrix as plasticized PVC (in line 92), pristine PVC (in line 94), PVC (in Fig.2e), and the author should provide a explicit concept. Besides, the mass fraction of DOP exceeds 50%, Why the authors provided no more explanation about the role of DOP in the performance of PVC-based elastomer?

2、Why the authors chose the electric field of 9.09 V/μm for actuation test? The electric strength of all new DEs in this work needs to be supplied.

3、In line 69-71, the authors claimed that "DE actuators with low dielectric permittivity often require high electric field to drive (> 20 V/μm), leading to the high risks of current leakage and electrical breakdown", but it is inaccurate. If the electric field to drive is far lower than the breakdown strength while just a large value, the current leakage and electrical breakdown may not happen.

4、According to the Supplementary Fig.7, the actuation test of DEs may be under prestrain condition, and the authors need to clarify this point. About the results of actuation test, the authors cited some references to highlight the driving properties of DEs in this work, while the results of actuation test in the references of 9, 22, 35, were obtained under non-prestrain condition. Besides, the work of reference 36, is about polyurethane dielectric elastomer, not about VHB.

5、About stress relaxations of DEs, in line 195 the constant strain is 200%, while in line 205 the constant strain is 100%?

6、Except of Fig 3, the quality of other Figures needs to be improved.

7、Some crucial references should be cited in order to make a good understanding to this work. They include Chen et al. Nature 2019, 575, 324-329; Chen et al. Chemical Engineering Journal 2021, 405, 126634; Cao et al. Extreme Mechanics Letters 2020, 35, 100619; Feng et al. Chemical Reviews 2022, 122, 3820-3878 and Yin et al. Nature Communications 2021, 12, 4517.

Reviewer #2:
Remarks to the Author:
Background: Dielectric elastomers show great promise as large strain artificial muscle. A challenge has been the high voltages and field strengths needed, with the devices operating close to breakdown and requiring kV level sources. If the voltage levels can be reduced to about 1 kV or less, the electronics is much less expensive. In order to achieve lower voltage operation, many investigators have sought to increase dielectric constant. This generally does not lead to improved strain, as voltage drops but so does breakdown strength, and elastic modulus rises. Another challenge is that the most commonly used elastomer - VHB - is highly viscoelastic in its behaviour, and so there is loss and creep. This is not the case in some silicones, which can achieve high bandwidth, relatively low loss, actuation - though silicones still requires high field strength. Kornbluh has achieved kHz frequency actuation of silicone, showing low loss.

The authors show that plasticized PVC with a high dielectric constant additive generate significant area strain (12%) at low field strength (9 kV/mm) compared to VHB, regular PVC, silicone and others. This is still a field strength well above the breakdown strength of air, but it is significantly lower than commonly used, and so is an important advance. The PVC additive used increases modulus, but produces a larger increase in dielectric constant, and reduces viscoelastic loss. Overall this leads to good low voltage actuation.

The authors compare creep and stress relaxation to that in the literature, for example in Figure 5d. How can a fair comparison be made, given the different loading conditions?

The authors have not expressed load in terms of stress, or described in the main text the amount of pre-strain used.
There has been substantial work on lowering voltage in dielectric elastomers by increasing dielectric constant, or reducing thickness. Early work on dielectrics was by Kofod I believe, and there has been a lot since, while a number of groups have sought to make thin elastomer layers. This work has not been properly summarized by the authors.
The improved sensor response compared to PVC is well presented. It is unclear how this performance compares to that of VHB and silicones.
What is the dielectric loss at typical operating frequencies/timescales - e.g. 1 Hz?

It is claimed that the actuators and sensors show fast response. I don't see a frequency response or time response analysis, or comparison with other materials.
What are the potential drawbacks of the approach? Do we expect a greater temperature dependence? Is breakdown strength reduced? Can large actuation be achieved, by applying higher voltages (why did the authors stop at the 9 V/micron)?

The sensor work is well presented. It it important to put this work put in context of other capacitive sensor work, and perhaps provide some detail in the supplementary. There are also commercial sensors (e.g. Stretchsense) to compare with.

Overall, the paper makes a significant contribution to the field of electroactive polymers. It is written clearly and concisely. I would recommend considering it for publication, following revisions.

Reviewer #3:
Remarks to the Author:
The authors proposed a polyvinyl chloride (PVC) -based dielectric elastomer with high permittivity and low viscoelasticity by introduction of cyanoethyl cellulose (CEC) into a plasticized PVC. The physicochemical and electromechanical coupling property of the as-synthesized CEC/PVC were carefully studied. This work improved the performance of traditional PVC. However, the demonstrated performance of actuation and sensing of the presented material are not convincing to me. The paper cannot meet the standard of Nature Communications unless the authors could clarify the advantage of the material compared to the existing material.

The strategy of increasing permittivity and reducing viscosity in the material synthesis part sounds reasonable and interesting. However, if the material is aimed to serve for dielectric elastomer applications, these two indexes are apparently not dominated material properties. As shown in Fig. 5, the actuation strain of the new material falls within the order of 10%. A lot of existing studies have shown that various of materials without careful optimization and complicated synthesis can easily achieve this actuation level. Similar concerns apply for the sensing demo in this work. In the introduction, the authors pointed out that the VHB material, widely used in literature, has a clear drawback in viscous property. However, the synthesized new material improves its viscous property compared to VHB but significantly sacrifices its ability of large actuation, which, to the reviewer’s opinion, is not satisfactory for dielectric elastomer applications.

Responses to Reviewer #1:

We thank the reviewer for her/his insightful report. We are delighted that the reviewer agreed that "*The PVC-based elastomer shows high permittivity, low viscoelasticity*". Here we address the comments and technical questions raised by the reviewer with new experimental results and analyses.

COMMENT 1#: *The PVC-based elastomer shows high permittivity, low viscoelasticity, but the dielectric loss is not satisfactory.*

Response: The heat generated due to dielectric losses of dielectric elastomers (DEs) would result in substantial increases in their temperature and conductivity over time, which would lower the breakdown strength and may lead to thermal or electrical breakdown [R1-R3]. Therefore, low levels of dielectric loss < 0.3 are highly preferred for DEA application as suggested by previous study [R4,R5]. However, the commonly used strategies for increasing dielectric permittivity, such as the addition of inorganic polar particles and conductive particles, are often associated with substantial increases in dielectric loss (*e.g.* > 0.5), leading to current leaking and/or electric breakdown. For instance, the addition of multiwalled carbon nanotube with 6-9 wt% loadings into PDMS precursors resulted in very high dielectric losses of 3.74-4.00 @ 1.0 kHz [R6]. By contrast, the dielectric loss of CEC/PVCg elastomers were maintained in a low value range 0.033-0.142 in this study (**Fig. R1a**). To address the reviewer's concern, we measured the conductivity and breakdown strength of CEC/PVCg elastomers. Because according to dielectric percolation theory, the increase of dielectric loss is resulted from the formation of a connected conductive network by the conductive filler materials. The results showed that CEC/PVCg elastomers with conductivity range of 3.40×10^{-9}-4.55×10^{-9} S/cm were in a highly insulating state (**Fig. R1b** and **R1c**). Moreover, it was found that the addition of CEC only slightly decreased the breakdown strength. The breakdown strength of CEC/PVCg elastomers were 18.22-20.06 V/μm (**Fig. R1a**). Although a low electric field of 9.09 V/μm is applied to drive the DEA in this study, a large actuation strain, which is much larger than commercial VHB 4910 based DEA has been achieved with our CEC/PVCg DEA. Therefore, we believe the dielectric loss of CEC/PVCg elastomers here is low enough to support the large actuation while preventing the device breakdown.

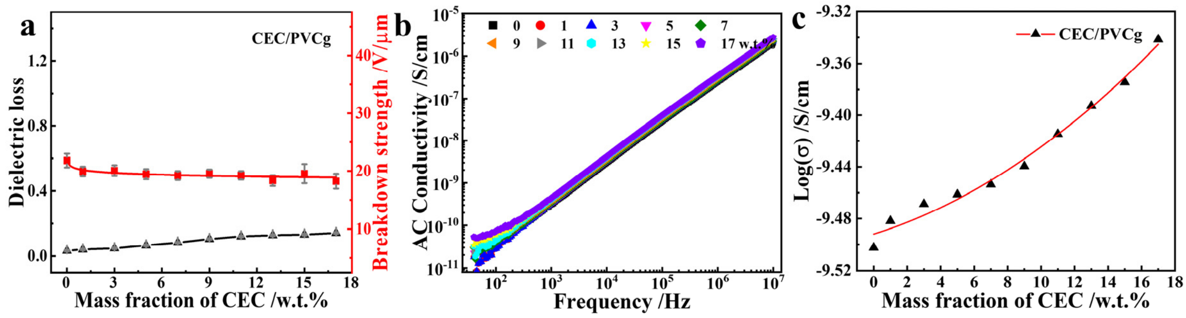


Fig. R1 Dielectric property tests. Dielectric losses and breakdown strengths of CEC/PVCg elastomers with CEC mass loadings of 0-17 wt% (a). AC conductivities of CEC/PVCg elastomers with varying CEC loadings under 40-10^7 Hz frequencies (b). Evolution of AC conductivity at 1 kHz for the CEC/PVCg elastomers (c).

The data has now been added into the revised manuscript as **Fig. 3c** and the relevant discussion at Page 7, Paragraph 3, as shown below:

"Dielectric loss is crucial for most DEs. Because the high levels of dielectric loss can result in substantial increases in both temperature and conductivity, which could potentially lead to thermal or electrical breakdown[43-45]. Unfortunately, the commonly used strategies of increasing dielectric permittivity, such as the addition of inorganic polar particles and conductive particles, were often associated with a substantial increase in the dielectric loss (*e.g.* > 0.5 at 1 kHz), which would lower the breakdown strength and reduce the lifetime of actuators[46,47]. According to dielectric percolation theory, the increase of dielectric loss is mainly due to the increase of conductive filler materials and the formation of a connected conductive network. For instance, the addition of 6-9 wt% multiwalled carbon nanotube into PDMS resulted in very high dielectric losses of 3.74-4.00 @ 1.0 kHz[48]. By contrast, the introduction of CEC into PVCg matrix in this study induced a marginal increase, resulting in dielectric losses of < 0.15 at 1 kHz for CEC/PVCg elastomers (**Fig. 3c**), which are still in a low level for DEs as suggested by previous study[49]. The limited increase of dielectric loss following the addition of CEC could be ascribed to the highly electrical insulating nature of CEC (**Supplementary Note 2**). The measurement of conductivity indicated that the CEC/PVCg elastomers (4.0×10^{-9} S/cm) were in a highly insulating state. Importantly, the breakdown strength was only slightly decreased by 10.4 % following the addition of CEC and kept almost constant for CEC/PVCg elastomers with different loading concentrations of CEC (**Fig. 3c** and **Supplementary Information Fig. S4**). The detected breakdown strength of CEC/PVCg elastomers with 9 wt% CEC was 19.53 V/μm. Thus, a much lower driving electric field of 9.09 V/μm was used in all of experiments in this study in order to prolong the working life of device and achieve a stable performance while generating a desired large area strain."

COMMENT 2#: *It is unclear how the plasticized PVC named by the authors is processable. It is confusing that the authors named the matrix as plasticized PVC (in line 92), pristine PVC (in line 94), PVC (in Fig.2e), and the author should provide an explicit concept. Besides, the mass fraction of DOP exceeds 50%, Why the authors provided no more explanation about the role of DOP in the performance of PVC-based elastomer?*

Response: We thank the reviewer for her/his suggestion. We have corrected the manuscript by naming pure PVC as pure PVC and naming plasticized PVC as PVC gel (PVCg) because of the plasticized PVC is in a gel-like form.

The major purpose of adding a plasticizer, *i.e.* DOP in this study, into PVC matrix is to increase the compliance and flexibility of PVC, which is critical for their actuation and sensing performances. As per the actuation strain equation of $S_z = \varepsilon_r \varepsilon_0 E^2/Y$, the matrix film with a lower modulus (Y) would generate a larger actuation strain (S) [R7-R9]. It has been suggested that DEs with a modulus less than 1 MPa is preferred for actuation application [R7]. The pure PVC has a very high modulus of 23.2 MPa (**Fig. R2a** and **R2c**), which is not suitable for actuation and sensing applications. The introduction of DOP would weaken the molecular interactions among PVC chains, leading to the transition of PVC matrix from glassy-state to hyperelastic-state with the high flexibility [R10]. Our results showed that the addition of 50-80 wt% DOP into PVC matrix significantly reduced the moduli to 0.02-0.54 MPa and increased the elongation at break to 200-560 % (**Fig. R2b** and **R2c**), making the matrix more favorable for actuation and sensing applications.

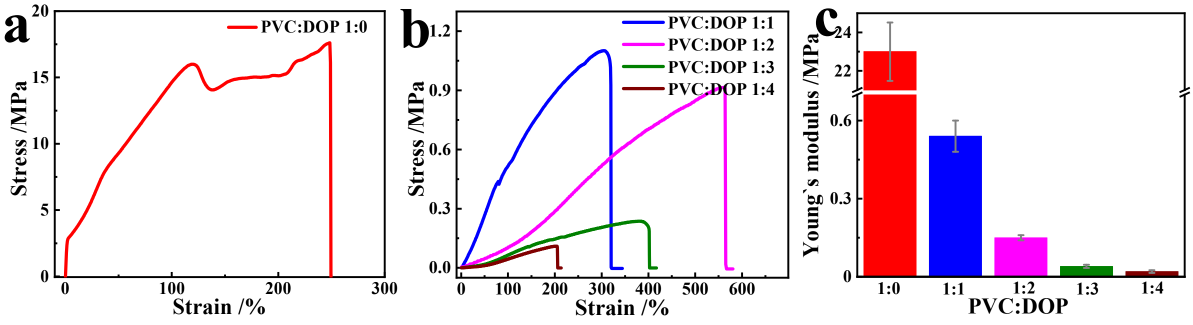


Fig. R2 Mechanical property tests. Stress-strain curves of the self-casting (a) PVC plastics and (b) PVCg elastomers with PVC: DOP mass ratios of 1:1, 1:2, 1:3, 1:4, and (c) their Young's moduli.

Although the introduction of plasticizer is a common strategy to generate large actuation of PVC-based DEs, the plasticized PVC suffers from inherent strong viscoelastic effects, which results in evident mechanical loss, stress relaxation, and viscoelastic hysteresis, eventually leading to instability and attenuation of output signals over time as well as delayed response in actuation and sensing applications [R11, R12]. Both dynamic and static viscoelasticity of plasticized PVC, *i.e.* PVCg, as a function of the loading concentrations of DOP were measured and shown in the following **Fig. R3** and **Fig. R4**, respectively. The dynamic viscoelasticity, *i.e.* mechanical loss of PVCg (tan $\delta = G''/G'$), was increased by 15.50 folds by increasing the loading concentration of DOP from 50 wt% (1:1) to 80 wt% (1:4) (**Fig. R3**). Under constant load of 60 *g* for 6 hours, the creep-induced elongation increased from 25 % for PVCg (1:1) to 60 % for PVCg (1:4) (**Fig. R4a-4d**). Similarly, under constant strain of 100 % for 10 min, the recorded stress attenuation increased from 11.41 % for PVCg (1:1) to 33.99 % for PVCg (1:4) (**Fig. R4e** and **4f**). Notably, currently existing PVC matrix used as DEA or DES often contain much larger concentrations of plasticizer (> 90 wt%) than this study [R13], leading to even stronger viscoelastic effects than what we demonstrated here. In addition, the introduction of DOP decreased the breakdown strength of PVC matrix due to the percolation effect of DOP, as shown in the following **Fig. R5a**.

To conclude, the increase of plasticizer content in PVCg resulted in lower elastic modulus (*i.e.* higher flexibility), higher viscoelastic effects, and a lower breakdown strength. Therefore, we choose the mass ratio of 1: 2 (PVC: plasticizer) to balance these properties of PVCg. More importantly, the introduction of CEC into the plasticized PVCg can address this long-standing challenge by significantly reducing its viscoelastic effects and concurrently achieving the high permittivity.

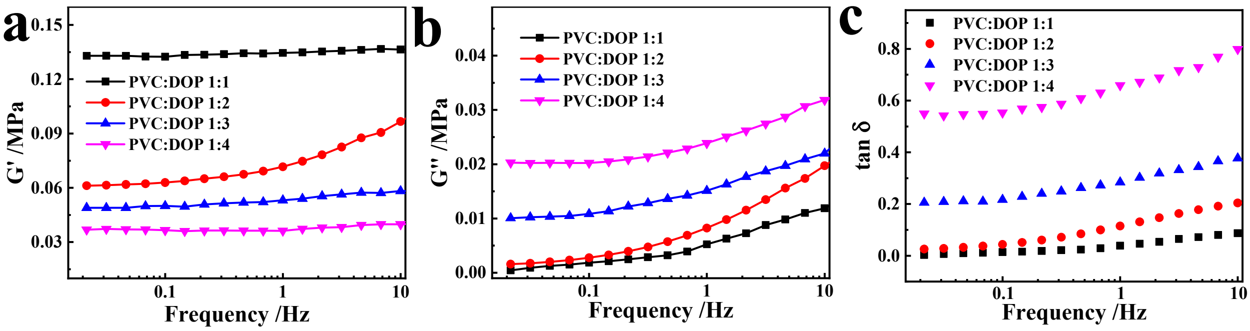


Fig. R3 Dynamic viscoelasticity tests of PVCg elastomers with varying DOP concentrations. Evolutions of (a) storage moduli (G'), (b) loss moduli (G''), and (c) the counted mechanical losses (tan $\delta = G''/G'$) under frequencies of 0.01-10 Hz.

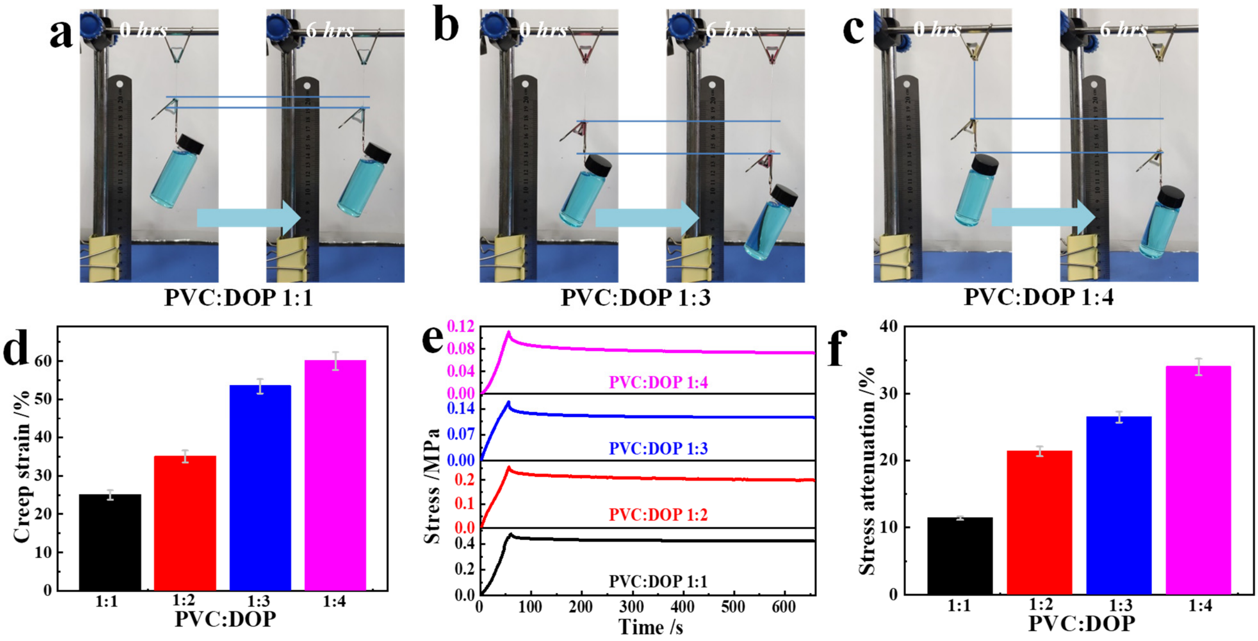


Fig. R4 Static viscoelasticity of PVCg elastomers with varying DOP concentrations. Creep behaviors for PVCg elastomers with PVC: DOP mass ratios of (a) 1:1, (b) 1:3, (c) 1:4 under a constant load of 60 *g* for 6 hours (data of the elastomer of mass ratio of 1:2 was shown formerly). Quantifications of (d) creep strains, (e) stress relaxations, and (f) percentages of stress attenuation of PVCg elastomers with PVC: DOP mass ratios of 1:1, 1:2, 1:3, 1:4.

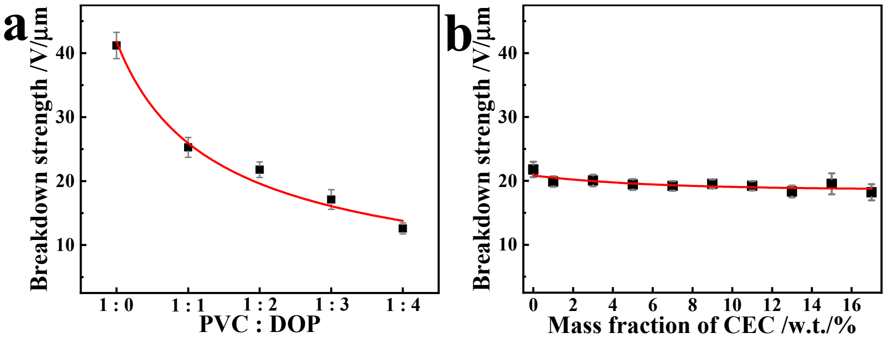


Fig. R5 Measurement of breakdown strengths. (a) Evolution of breakdown strengths for PVCg elastomers with varying DOP concentrations. (b) Evolution of breakdown strengths for CEC/PVCg elastomers with varying CEC concentrations.

We have now added the new data into the revised supplementary materials and relevant discussion about the rationale and impacts of the introduction of DOP into PVC in the **Supplementary Note 1** and the revised manuscript at Page 4, Paragraph 2 and Page 19 and Paragraph 2, as shown below:

"The plasticizers are often introduced into PVC matrices in order to produce highly flexible PVCg elastomers with high flexibility by weakening the interaction forces among PVC chains[29] (**Supplementary Information Fig. S1**). However, the plasticized PVCg suffer from low breakdown strength and inherent strong viscoelastic effects[30], which leads to time-dependent change of internal stress and strain[31], *i.e.* creep (**Supplementary Information Fig. S3**)."

"The loading concentration of DOP, *i.e.* 2:1 mass ratio of PVC: DOP, was chose to balance the flexibility, viscoelastic effects, and breakdown strength of the resulting PVCg (**Supplementary Note 1**).”

COMMENT 3#: *Why the authors chose the electric field of 9.09 V/μm for actuation test? The electric strength of all new DEs in this work needs to be supplied.*

Response: Existing DEAs often requires high driving electrical field to achieve large strain, which, however, could potentially lead to polymer creep, current leakage, and electrical breakdown. In addition, it also needs a bulky, high-voltage power supply system, hampering its wide-spread applications [R14]. Therefore, generation of a large strain under low driving voltages is highly desirable for DEAs, which remains a challenge. In this study, we have measured the electric breakdown strength of all DEs, as shown in the **Fig. R5**. The breakdown strength of plasticized PVCg (1:2) was about 20 V/μm. The introduction of CEC with various loading concentrations have limited impacts on the breakdown strength while it dramatically increased permittivity. Thus, a relative low working voltage of 9.09 V/μm was selected in order to prolong the working life of the device, achieve a stable performance while generating a desired large area strain [R15-18]. Notably, such strain of 12.22 % was achieved under a very small pre-strain of 25 %, which was negligible when compared to other pre-strains such as 540 % and 400 % [R19, R20]. We have now provided the new data of breakdown strength measurement in the revised manuscript as **Fig. 3c** and discussions on the reason for choosing the low electric field of 9.09 V/μm at Page 8, Paragraph 1, as shown below:

“Importantly, the breakdown strength was only slightly decreased by 10.4 % following the addition of CEC and kept almost constant for CEC/PVCg elastomers with different loading concentrations of CEC (**Fig. 3c** and **Supplementary Information Fig. S4**). The detected breakdown strength of CEC/PVCg elastomers with 9 wt% CEC was 19.53 V/μm. Thus, a much lower driving electric field of 9.09 V/μm was used in all of experiments in this study in order to prolong the working life of device and achieve a stable performance while generating a desired large area strain.”

COMMENT 4#: *In line 69-71, the authors claimed that “DE actuators with low dielectric permittivity often require high electric field to drive (> 20 V/μm), leading to the high risks of current leakage and electrical breakdown”, but it is inaccurate. If the electric field to drive is far lower than the breakdown strength while just a large value, the current leakage and electrical breakdown may not happen.*

Response: We agree with the reviewer that when the driving electric field is far lower than the breakdown strength, the current leakage and electrical breakdown should not happen. Unfortunately, the existing DE actuators often required the high driving electric fields that are close to their breakdown strength in order to achieve large strain because of their low permittivity. For example, as we measured in this study, VHB-based actuators produced rather small area strain of 3.45 % under driving electric field of 22.5 V/μm, which is close to their breakdown strength of 28.4 V/μm [R15] (**Supplementary Table S2**). PDMS (Gelest OE^{TM} Extended Cure)-based actuator required 30 V/μm, which was the measured breakdown strength, to achieve the area strain of 4.63 % [R21]. In addition, even the driving voltage is far lower than breakdown strength, a high value of several kilovolts arises safety issues and brings a problem of using a bulky high-voltage power supply system. To address the reviewer’s concern, we have updated our description and added more discussions on this issue in the revised manuscript Page 3 and Paragraph 1, as shown below:

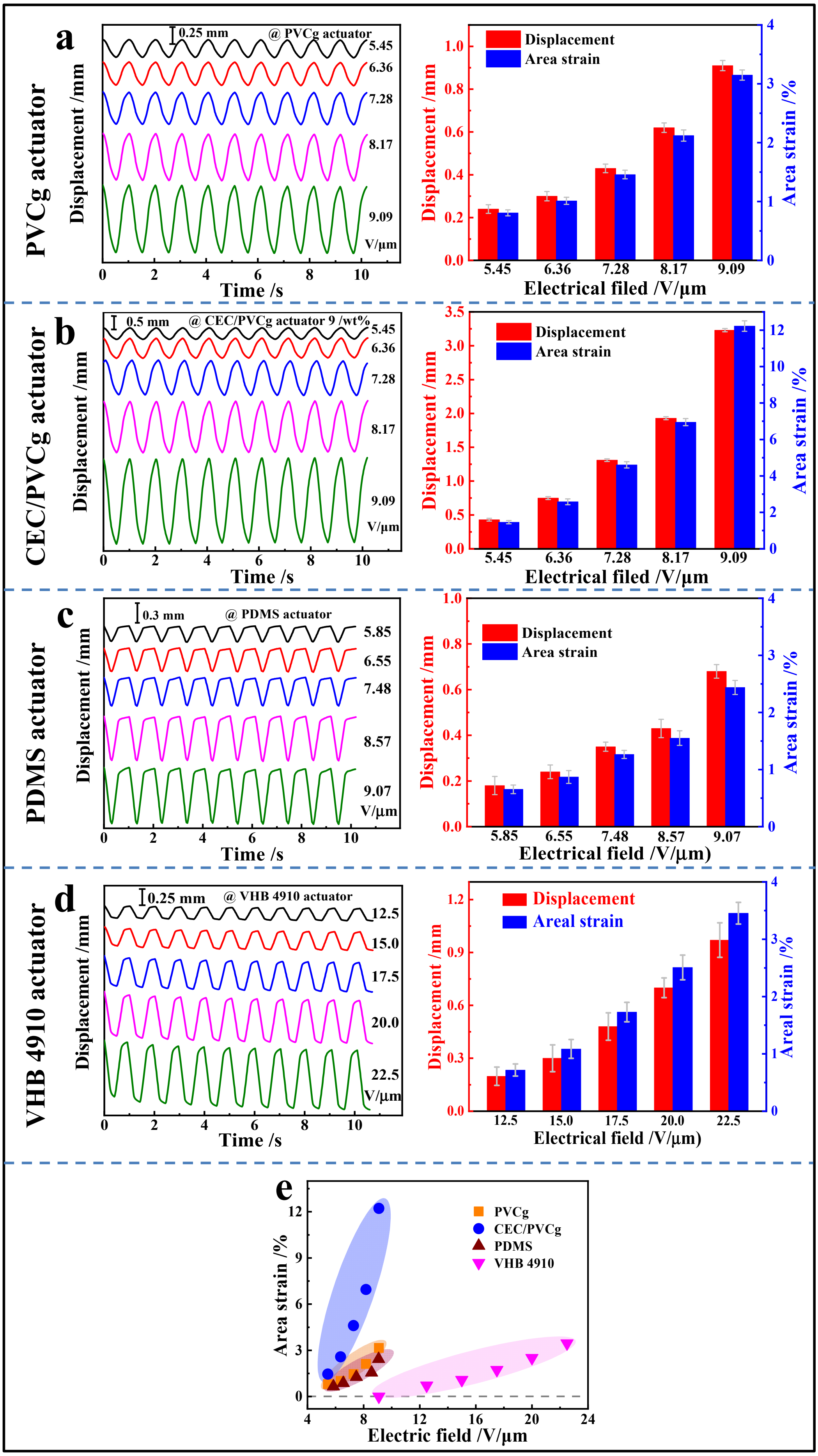
“DE actuators with low dielectric permittivity, such as PDMS and VHB materials, often require high driving electric fields (> 20 V/μm) to achieve large actuations, which would lead to the high risks of current leakage[15] and electrical breakdown[16] when such high driving electric fields are close to their breakdown strength. In addition, a high value of several kilovolts arises safety issues and brings about the problem of using a bulky high-voltage power supply system[17].”

COMMENT 5#*: According to the Supplementary Fig.7, the actuation test of DEs may be under prestrain condition, and the authors need to clarify this point. About the results of actuation test, the authors cited some references to highlight the driving properties of DEs in this work, while the results of actuation test in the references of 9, 22, 35, were obtained under non-prestrain condition. Besides, the work of reference 36, is about polyurethane dielectric elastomer, not about VHB.*

Response: We thank the reviewer for bringing up this important issue. We have now clarified that the DEAs were under 25 % pre-strain in this study in the revised manuscript. We have corrected our citation of Reference 36. Moreover, we have prepared the PDMS and VHB 4910-based DEAs and measured their actuation performances under the exact same conditions in order to make a fair comparison to our CEC/PVCg-based actuators. As shown in the following **Fig. R6**, the detected area strains were 1.46-12.22 % for the CEC/PVCg actuators under the driving electrical fields of 5.45-9.09 V/μm, 0.81-3.15 % for the PVCg actuators under 5.45-9.09 V/μm, 0.65-2.44 % for the PDMS actuators under 5.85-9.07 V/μm, while VHB 4910 actuators cannot be activated under the driving electrical field < 12.5 V/μm, *i.e.* strain of 0 %. The area strains of VHB 4910 actuators were 0.72-3.45 % when the driving electrical fields were further increased to 12.5-22.5 V/μm. To conclude, our CEC/PVCg actuators produced the largest actuation strains under low driving voltages as compared to the commonly used PDMS and VHB-based actuators. We have now provided the new data as **Fig. 6b** and relevant discussions in the revised manuscript at Page 11, Paragraph 3, and Page 20, Paragraph 1, as shown below:

"According to the strain model shown in **Supplementary Information Fig. S12**, the counted area strain generated by the CEC/PVCg actuators was 12.22 % (with 9 wt% CEC, under driving voltage of 9.09 V/μm, pre-strain of 25 %), which represents 3.9-fold increase as compared to the PVCg actuators (**Fig. 5b**). In addition, we prepared the commonly used PDMS and VHB-based actuators and measured their actuation strains under the same conditions as the comparison (**Fig. 5b, Supplementary Table S2,** and **Supplementary Information Fig. S15**). PDMS actuators produced strains of 0.65-2.44 % under electrical fields of 5.85-9.07 V/μm. VHB 4910 actuators could not be triggered, *i.e.* 0 % area strain, under electrical fields < 12.5 V/μm and produced only 0.72-3.45 % strains by further increasing electrical fields to 12.5-22.5 V/μm. Therefore, our CEC/PVCg actuators generated significantly larger actuation strains, *i.e.* > 5 times, than commonly used PDMS and VHB 4910 actuators, which was largely attributed to the augmentation of the electromechanical coupling sensitivity *k* of CEC/PVCg (**Fig. 3e**)."

"The initial flection amplitude of 12 mm was generated by using the elastic spring as a pre-load, which corresponded to a pre-strain of 25 %."


a
PVCg actuator
Displacement /mm
0.25 mm
@ PVCg actuator
5.45
6.36
7.28
8.17
9.09
V/μm
Time /s
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Area strain
Displacement /mm
Area strain /%
Electrical filed /V/μm
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VHB 4910
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Fig. R6 Evaluation of actuation properties of (a) PVCg, (b) CEC/PVCg, (c) PDMS, (d) VHB 4910 actuators by measuring their flection displacements and area strains under various driving voltages. (e) The comparison of area strains (mean values) that were generated by different actuators.

***COMMENT 6#:** About stress relaxations of DEs, in line 195 the constant strain is 200 %, while in line 205 the constant strain is 100 %?*

Response: 200 % was a typo and we have corrected the value of constant strain to 100 % in the revised manuscript.

***COMMENT 7#:** Except of Fig 3, the quality of other Figures needs to be improved.*

Response: We have now improved the quality and resolution of all the figures. We have now submitted all figures in the format of .TIFF instead of word file in the initial submission, which reduce the resolution of figures.

***COMMENT 8#:** Some crucial references should be cited in order to make a good understanding to this work. They include Chen et al. Nature 2019, 575, 324-329; Chen et al. Chemical Engineering Journal 2021, 405, 126634; Cao et al. Extreme Mechanics Letters 2020, 35, 100619; Feng et al. Chemical Reviews 2022, 122, 3820-3878 and Yin et al. Nature Communications 2021, 12, 4517.*

Response: We thank the reviewer for bringing these important articles to our attention. We have now added and discussed the following references in the revised manuscript as shown below:

“DE actuators (DEAs) are attractive artificial muscles due to their high energy density[7] and conversion efficiency[8], and fast response[9].”

“For example, the dielectric permittivity of widely used DEs are 2.2-3.0 for polydimethylsiloxane (PDMS)[11], 4.4-4.7 for VHB acrylic elastomer (3M)[12,13], and 4.0 for pure polyvinyl chloride (PVC)[14].”

“In addition, a high value of several kilovolts arises safety issues and brings about the problem of using a bulky high-voltage power supply system[17].”

7. Chen, Y. F. et al. Controlled flight of a microrobot powered by soft artificial muscles. *Nature* **575,** 324-329 (2019).
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12. Yin, L. J. et al. Soft, tough, and fast polyacrylate dielectric elastomer for non-magnetic motor. *Nat. Commun.* **12,** 4517 (2021).
17. Chen, Z. Q. et al. Ultrasoft-yet-strong pentablock copolymer as dielectric elastomer highly responsive to low voltages. *Chem. Eng. J.* **405,** 126634 (2021).

References

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Responses to Reviewer #2:

We are grateful for reviewer's detailed and positive comments on our manuscript. We thank the reviewer for his/her very encouraging remarks that "*Overall, the paper makes a significant contribution to the field of electroactive polymers. It is written clearly and concisely. I would recommend considering it for publication, following revisions.*" We are also delighted that the reviewer agreed that the viscoelastic effects of our device is "*significantly lower than commonly used, and so is an important advance*", "*The PVC additive used increases modulus, but produces a larger increase in dielectric constant, and reduces viscoelastic loss. Overall, this leads to good low voltage actuation*" Here we address the comments and the technical questions from the reviewer with the new experimental data and analysis.

COMMENT #1: *The authors compare creep and stress relaxation to that in the literature, for example in Figure 5d. How can a fair comparison be made, given the different loading conditions?*

Response: To address reviewer's concern, we have prepared PDMS and VHB 4910-based actuators and evaluated their actuation performances under the exact same conditions as our CEC/PVCg actuators. First, the actuation displacements over 1000 cycles, *i.e.* 1000 seconds of four types of actuators were measured and the relative displacement shifts (RDS) were calculated to quantify their viscoelastic drifts. As shown in the following **Fig. R1a-1e**, the RDS values of VHB 4910, PDMS, PVCg, and CEC/PVCg actuators were 136.09 %, 5.70 %, 59.40 %, and 7.78 %, respectively. Our CEC/PVCg actuators showed a very low shift of displacement over 1000 cycles of actuation, which was 94 % and 87 % reductions as compared to VHB 4910 and PVCg actuators. Second, the area strains of four types of DEAs were measured (**Fig. R1f**). The detected area strains of PDMS, PVCg, and CEC/PVCg actuators were 2.44 %, 3.15%, and 12.22 %, respectively under the driving electric field of 9.09 V/μm. VHB 4910 actuators cannot be activated under the driving electrical field < 12.5 V/μm, *i.e.* strain of 0 %, and showed area strains of 0.72 %-3.45 % when the driving electrical fields were further increased to 12.5-22.5 V/μm. Our CEC/PVCg actuators showed > 4-fold increases in area strains as compared to other three types of actuators. To conclude, Our CEC/PVCg actuators showed low viscoelastic effects and large area strains, demonstrating the significant improvement in actuation performances as compared to the existing dielectric elastomer actuators, such as PDMS and VHB 4910.

We have now provided the updated data of area strains and displacement shifts as **Fig. 5b** and **5d** in the revised manuscript. The relevant descriptions and discussions were provided in the revised manuscript at Page 11, Paragraph 3 and Page 12, Paragraph 2, as shown below:

"According to the strain model shown in **Supplementary Information Fig. S12**, the counted area strain generated by the CEC/PVCg actuators was 12.22 % (with 9 wt% CEC, under driving voltage of 9.09 V/μm, pre-strain of 25 %), which represents 3.9-fold increase as compared to the PVCg actuators (**Fig. 5b**). In addition, we prepared the commonly used PDMS and VHB-based actuators and measured their actuation strains under the same conditions as the comparison (**Fig. 5b, Supplementary Table S2,** and **Supplementary Information Fig. S15**). PDMS actuators produced strains of 0.65-2.44 % under electrical fields of 5.85-9.07 V/μm. VHB 4910 actuators could not be triggered, *i.e.* 0 % area strain, under electrical fields < 12.5 V/μm and produced only 0.72-3.45 % strains by further increasing electrical fields to 12.5-22.5 V/μm. Therefore, our CEC/PVCg actuators generated significantly larger actuation strains, *i.e.* > 5 times, than commonly used PDMS and VHB 4910 actuators, which was largely attributed to the augmentation of the electromechanical coupling sensitivity *k* of CEC/PVCg (**Fig. 3e**)."

"The displacements of the PVCg, CEC/PVCg, PDMS, and VHB 4910 actuators were measured and recorded over actuation for 1000 cycles, *i.e.* 1000 seconds (**Fig. 5c** and **Supplementary Information Fig. S16**). The CEC/PVCg and PDMS actuators produced remarkably stable displacement profiles. By contrast, apparent displacement shifts were observed over time for PVCg and VHB 4910 actuators. Relative displacement shifts (RDS) were calculated to quantify the viscoelastic effects (**Fig. 5d**). It was found that RDS values increased with time, *i.e.* number of actuation cycles, for VHB 4910 and PVCg actuators, while remaining almost constant for CEC/PVCg actuators. The relative shifts over 1000 cycles of CEC/PVCg actuators (7.78 % of RDS) represented 87 % and 94 % reductions as compared to PVCg (59.40 % of RDS) and VHB 4910 actuators (136.09 % of RDS). PDMS actuators (5.70 % of RDS) displayed similar viscoelastic drifts to CEC/PVCg actuators."

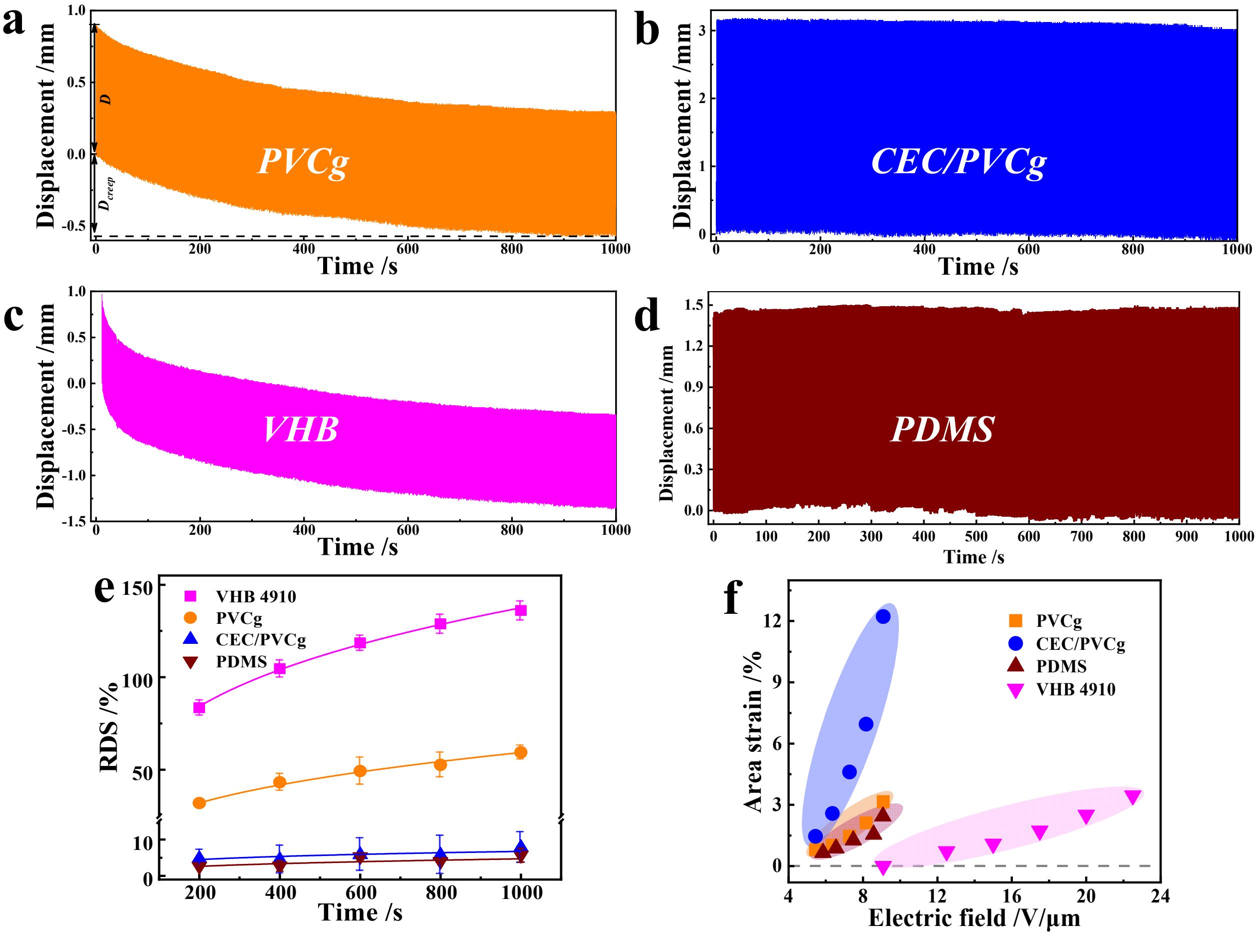

Fig. R1 Actuation stability and area strains of four types of actuators. Duration tests for the (a) PVCg, (b) CEC/PVCg, (c) VHB 4910, and (d) PDMS actuators over 1000 cycles of actuation, *i.e.* 1000 seconds. (e) Evolutions of relative displacement shift (RDS) of the four types of actuators. The actuation tests of PVCg, CEC/PVCg, and PDMS were performed under 9.09 V/μm electrical field and 1 Hz frequency, while the VHB actuator was triggered under 22.5 V/μm electrical field and 1 Hz frequency. $\mathrm{RDS} = \frac{|D-D_{creep}|}{D} \times 100\%$, where D is the amplitude of displacement and D_{creep} is the shift of the displacement as shown in (a). (f) The mean values of area strains that were generated by four types of actuators as functions of driving electric fields.

COMMENT #2: *The authors have not expressed load in terms of stress, or described in the main text the amount of pre-strain used.*

Response: In the evaluation of actuation performance, the elastomer film was carefully coated on a circle frame (ϕ = 50 mm) to make a concise and facile pump diaphragm. By using an elastic spring as the normal preload, the diaphragm center was pulled down 12 mm each time and the counted area pre-strain was 25 % according to the spherical crown model shown in **Supplementary Information Fig. S12**. We have now provided the description of pre-strain in the revised manuscript at Page 20, Paragraph 1 and relevant figure captions, as shown below:

"The initial flection amplitude of 12 mm was generated by using the elastic spring as a pre-load, which corresponded to a pre-strain of 25 %."

COMMENT #3: *There has been substantial work on lowering voltage in dielectric elastomers by increasing dielectric constant, or reducing thickness. Early work on dielectrics was by Kofod I believe, and there has been a lot since, while a number of groups have sought to make thin elastomer layers. This work has not been properly summarized by the authors.*

Response: Following the reviewer's suggestions, we have now summarized and discussed the earlier work on increasing dielectric permittivity and reducing thickness of dielectric elastomers in the revised manuscript at Page 3, Paragraph 2, as shown below:

"Numerous efforts have been devoted to increase the dielectric permittivity and mechanical flexibility to generate a large actuation under relatively low driving voltages[11-13,16-19]. For instance, the seminal work from Kofod's group enhanced the relative permittivity of the PDMS elastomer from 3.0 to 5.9 and decreased the elastic modulus from 1900 to 550 kPa by grafting small molecules with high dipole moment to the elastomer matrix, leading to significant improvement of their electromechanical performances[19]. In addition, the reduction of the film thickness is an alternative method to improve the actuation performance[20-23]. For example, Shea and his co-workers demonstrated that the actuation strain of 7.5 % could be generated with a 3 μm thick film under a driving voltage of 245 V[20]. By contrast, it required much higher driving voltage of 3.3 kV to generate the same actuation strain with the 30 μm thick film. Despite these positive outcomes, thin film actuators often require complicated fabrication processes and are associated with high prevalence of an electromechanical instability[24]."

COMMENT #4: *The improved sensor response compared to PVC is well presented. It is unclear how this performance compares to that of VHB and silicones.*

Response: To address the reviewer's concern, we made PDMS and VHB 4910 based sensors and evaluated their sensing performances, *i.e.* sensitivity and stability, under the exact same conditions as PVCg and CEC/PVCg sensors. First, sensitivity tests showed that our CEC/PVCg sensors demonstrated the highest sensitivity than other sensors, as shown in the following **Fig. R2**. Specifically, the sensitivity (S) of CEC/PVCg sensors were 3.1-fold and 1.5-fold higher than PDMS and VHB 4910 sensors in the displacement ranges of 7-14 mm, respectively. Second, the stability of sensing performance was evaluated over 1440 cycles of flection (2.5 s per cycle and 60 min in total), as shown in **Fig. R3**. The profiles of relative capacitance over time (**Fig. R3a-3d**) indicated the stable performance for CEC/PVCg and PDMS sensors while apparent shift over time for PVCg and VHB 4910 sensors. The relative standard deviation (RSD) of relative capacitances over 1440 cycles was calculated to quantify the stability of their sensing performance (**Fig. R3e**). The results showed that our CEC/PVCg and PDMS sensors displayed much lower RSD values (5.75 % and 3.67 %), *i.e.* higher stability, than PVCg (9.84 %) and VHB 4910 (8.20 %) sensors. Altogether, our CEC/PVCg sensors demonstrated the superior overall sensing performances regarding of high sensitivity and stability over currently existing PVCg, PDMS, and VHB 4910 sensors. The new data has now been added as **Fig. 6c** and **6e** in the revised manuscript and **Supplementary Note 5**. The relevant discussion has been added in the revised manuscript at Page 14, Paragraphs 2 and 3, as shown below:

"To demonstrate the sensing application of the CEC/PVCg elastomers, the periodic strain driven by a linear reciprocating actuator was applied to the prepared DES devices, including CEC/PVCg, PVCg, PDMS, and VHB 4910-based sensors (**Fig. 6a** and **Supplementary Information Fig. S17**). The profiles and periods of the capacitance signals (output) that were generated from both PVCg and CEC/PVCg sensors were identical to the strain signals (input) (**Fig. 6b** and **Supplementary Video S5**), suggesting that the mechanical signal can be accurately converted into the electric signal by the prepared sensors (**Supplementary Note 5)**. The CEC/PVCg sensors showed the fast response time, *e.g.* 1.0, 0.5, and 0.25 seconds under frequencies of 0.5-2.0 Hz (**Supplementary Information Fig. S19**). Notably, the CEC/PVCg sensors generated a significantly higher signal/noise ratio, baseline capacitance and capacitance width (*i.e.* ΔC, difference between peak capacitance C and baseline capacitance C_0) than the PVCg sensors because of the higher permittivity of CEC/PVCg matrix[36]. Moreover, the CEC/PVCg sensors showed the highest sensitivity (S) among four types of sensors that we studied here. For instance, the sensitivity of CEC/PVCg sensors was 3.1-fold, 1.5-fold, and 1.7-fold higher than PDMS, VHB 4910, and PVCg sensors, respectively in the displacement range of 7-14 mm (**Fig. 6c** and **Supplementary Information Fig. S18**)."

"Notably, the capacitance generated by PVCg and VHB 4910 sensors showed an apparent drift over the recording time of 60 min (*i.e.* 1440 cycles) (**Fig. 6d** and **Supplementary Information Fig. S20**), which is in line with the previous report[28]. By contrast, CEC/PVCg and PDMS sensors produced remarkably stable capacitance signals without visible drift over at least 60 min, which was resulted from the low viscoelasticity and the inhibition on the rearrangement of their polar functions by the multiple molecular interactions. The relative standard deviation (RSD) of capacitances over 1440 cycles was analyzed to quantify the stability of sensors (**Fig. 6e**). The results showed that our CEC/PVCg and PDMS sensors displayed much lower RSD values (5.75 % and 3.67 %) of relative capacitances, *i.e.* higher stability, than PVCg (9.84 %) and VHB 4910 (8.2 %) sensors. Altogether, our CEC/PVCg sensors demonstrated the superior overall sensing performances regarding of high sensitivity and stability compared to existing PVCg, PDMS, and VHB 4910 sensors."

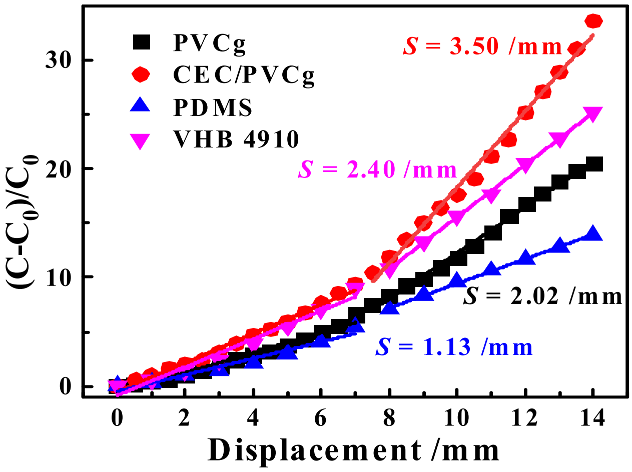

Fig. R2 Sensitivity tests. The relative capacitance ($C\text{-}C_0/C_0$) that was generated by PVCg, CEC/PVCg, PDMS, and VHB 4910 sensors as a function of the displacement. The tangential slope of the curve was defined as the sensitivity (S) of the sensors. The values of sensitivity in the displacement ranges of 7-14 mm were marked in the figure.

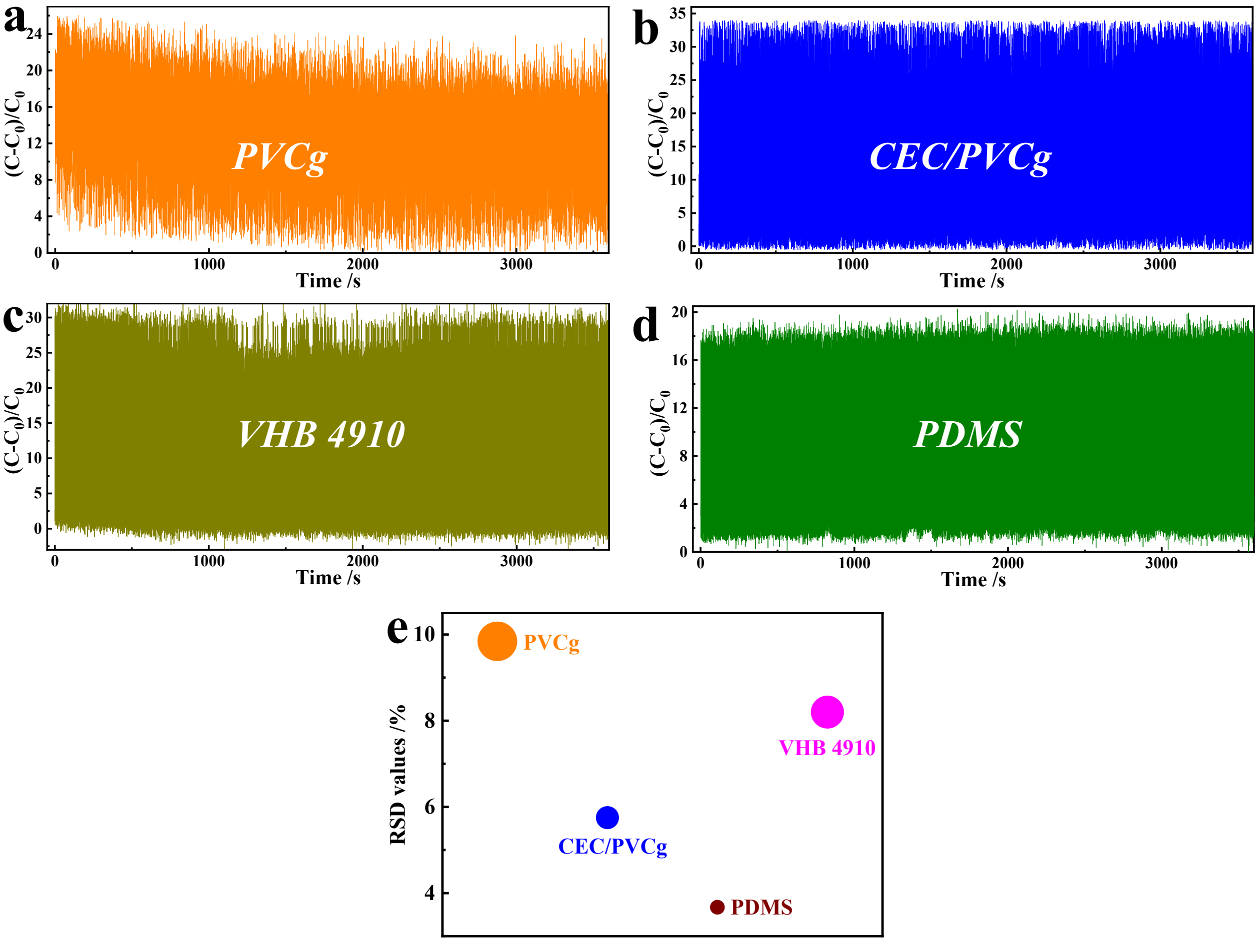

Fig. R3 Sensing stability tests. Duration tests within 1440 flection cycles (T = 2.5 s per cycle and 60 min in total) for the (a) PVCg, (b) CEC/PVCg, (c) VHB 4910, and (d) PDMS sensors. (e) The calculated relative standard deviation (RSD = standard deviation of relative capacitance / mean of relative capacitance) values of four types of sensors.

***COMMENT #5:** What is the dielectric loss at typical operating frequencies/timescales - e.g. 1 Hz?*

Response: We thank the reviewer for bringing up this issue. In the original manuscript, we used an impedance analyzer (4294A, Agilent, USA), which is the most commonly used system in the literature [R1-R6], to measure the dielectric properties of our elastomers. The testing frequency range of this machine is 40~10^7 Hz. Samples cannot be tested at 1Hz with this machine. To answer the reviewer's question, we tested the dielectric properties of our elastomers using a new machine (Concept 80 system, Novocontrol, Germany) in the frequency range of 0.1~10^7 Hz. The results showed that the dielectric losses of the CEC/PVCg elastomers with 0, 1, 9, 17 wt% loading concentrations of CEC were 20.84, 22.98, 22.98, 39.67 @ 1Hz, and 0.026, 0.031, 0.031, 0.047@ 1kHz, respectively (**Fig. R4**). The dielectric loss is highly frequency dependent, *i.e.* decrease at higher frequency, which is in line with previous reports [R9-R12]. In addition, the values of dielectric losses at 1kHz that were measured by traditional machine (4294A, Agilent) and the new machine (Concept 80 system) were different, *e.g.* 0.105 *vs* 0.031 at 1 kHz. Since most previous reports used the traditional machine (4294A, Agilent) to measure the dielectric properties at 1 kHz, we kept the original data that were acquired using traditional machine (4294A, Agilent) in the revised manuscript in order to have a better comparison to the previous literatures and avoid potential confusions.

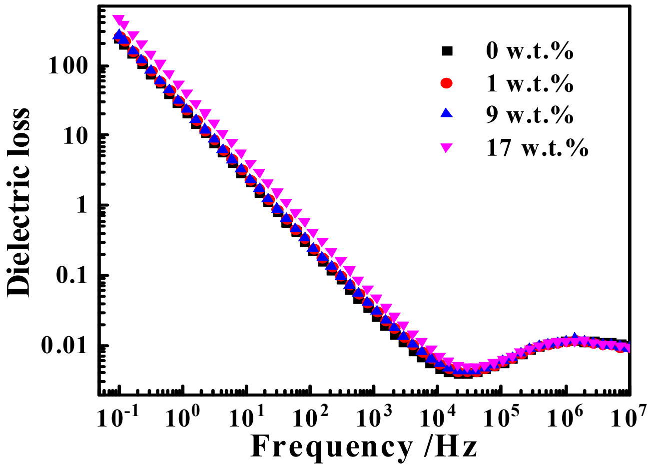

Fig. R4 Dielectric losses of the CEC/PVCg elastomers with 0, 1, 9, 17 wt% loading concentrations of CEC under frequencies of 0.1-10 MHz, that were measured using the new machine (Concept 80 system).

***COMMENT #6**: It is claimed that the actuators and sensors show fast response. I don't see a frequency response or time response analysis, or comparison with other materials.*

Our response: Most of creep driven actuators, such as high viscoelastic PVCg, take 5-20 seconds to complete one actuation cycle [R13-R16]. By contrast, our CEC/PVCg actuators take only 0.2-1 second to finish one actuation cycle (as shown in the following **Fig. R5a**). The CEC/PVCg actuators presented a characteristic Maxwell field driven actuator, where the flection amplitude decreases with the increase of frequency.

The capacitance sensors always showed a fast response from the previous literature. For instance, the capacitive strain sensor reported by Liu's group had a fast response time less than 140 ms [R17]. The flexible capacitive pressure sensor reported by Lee and Kim's group showed a response time of 0.578-1.04 seconds [R18]. **Fig. R5b** recorded typical capacity/frequency curves for the CEC/PVCg elastomers under a constant flection and different driving frequencies, including 0.5, 1.0, and 2.0 Hz. The typical response time of our CEC/PVCg sensors are 1.0, 0.5, and 0.25 seconds. In addition, the fast response of our CEC/PVCg sensors was also demonstrated by their faithful recording of the leg motion during fast running, where the response time is about 0.2 second, as shown in the **Fig. 6i in the manuscript** and **Supplementary Video S6**.

We have now added the relationship between the flection displacement and the driven frequency in **Supplementary Note 4**, and relevant discussions in the revised manuscript at Page 11, Paragraph 3 and Page 14, Paragraph 2 as shown below:

"In addition, the amplitude of flection displacement of our CEC/PVCg actuators was decreased with the increase of the driving frequency with fast response time (0.1-0.5 seconds) (**Supplementary Information Fig. S14**), which represented a characteristic electromechanical behavior of Maxwell field driven actuators. By contrast, it often took 5-20 seconds per cycle for creep-driven actuators, such as PVCg actuators[26]."

"The CEC/PVCg sensors showed the fast response time, *e.g.* 1.0, 0.5, and 0.25 seconds under frequencies of 0.5-2.0 Hz (**Supplementary Information Fig. S19**)."

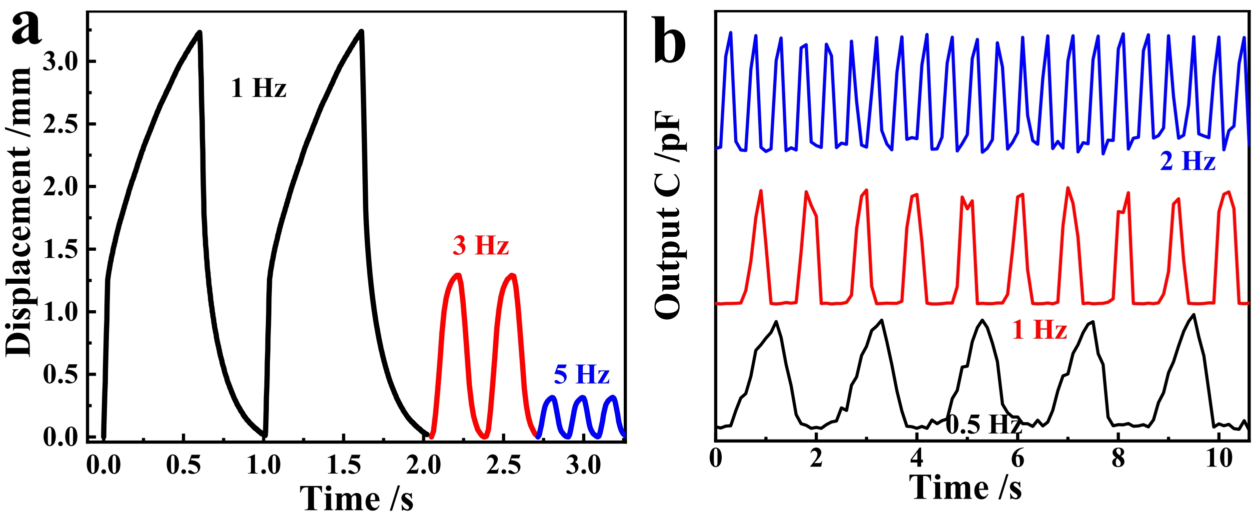

Fig. R5 Time response tests. (a) The flection displacements that were generated by our CEC/PVCg (9 wt% CEC) actuators under different driving frequencies and 9.09 V/μm electrical field. (b) The output capacity of our CEC/PVCg (9 wt% CEC) sensors under different driving frequencies and 14.0 mm flection displacement (input), which was generated from a commercial linear actuator.

COMMENT #7: *What are the potential drawbacks of the approach? Do we expect a greater temperature dependence? Is breakdown strength reduced? Can large actuation be achieved, by applying higher voltages (why did the authors stop at the 9 V/micron)?*

Response: We thank the reviewer for bringing up these important issues to discuss. A potential limitation of current device is the instinct low breakdown strength of the traditional PVCg, which was not improved by the addition of the CEC. The major reason is that the small plasticizer molecule *i.e.* DOP would percolate through the elastomer and generate the premature breakdown strength. The low breakdown strength limits the use of high driving voltage to achieve larger actuation. However, the introduction of CEC in the plasticized PVCg significantly increase their permittivity by 2.5 folds while maintaining the similar breakdown strength. Therefore, the CEC/PVCg can achieve large actuation under low driving electric field. Another limitation is the current film fabrication method, *i.e.* mold casting, which is very straightforward while producing a relative thick (~470 μm) film of the dielectric elastomer to ensure the uniformity of the film and large force output. The decrease of film thickness would be expected to lower the required driving electric field and increase the breakdown strength [R19,R20]. Thus, we are currently working on the fabrication of the thinner film, *e.g.* ~100 μm by using spray coating method, or down to ~50 μm by using spinning coating method, in order to further improve the actuation performances of our devices under low driving electric field. We have now provided the discussion about the limitations of this study in the revised manuscript at Page17, Paragraph 3, as shown below:

"One limitation of current devices is the intrinsic low breakdown strength, *i.e.* 21.79 V/μm, of the traditional plasticized PVCg, which has not been improved by the addition of CEC in this study. The low breakdown strength prevents the use of high driving electric field to achieve larger actuation. However, the introduction of CEC in the plasticized PVCg indeed significantly augmented their actuation by increasing their permittivity by 2.5 folds while maintaining the similar breakdown strength. Another limitation of this study is that the current film fabrication method, *i.e.* mold casting, produced a relative thick film (~470 μm) of DEs although it is very straightforward to use. It has been demonstrated that the decrease of film thickness would lower the required driving electric field and increase the breakdown strength[51,52]. Therefore, we are currently working on the fabrication of the thinner film, *e.g.* ~100 μm by using spray coating method, or down to ~50 μm by using spinning coating method, to further improve the actuation performances of DEAs under low driving electric field."

For the issue of temperature dependence of DEA/DES, the temperature is an important factor to consider for its influences on the electromechanical performance, especially when the devices would be used under environments with large temperature fluctuations. For example, Zhang's study demonstrated that the breakdown voltage of VHB 4910 decreased by increasing temperature [R21]. Michel *et al* and Vu-Conga *et al* reported that the elastic moduli of DEs were significantly decreased at elevated temperature, facilitating the generation of larger strains [R22,R23]. All the evaluations of this study were performed at room temperature. The investigation of temperature dependence is needed when our DEA and DES are applied at different environments in the future. To address the reviewer's concern, we have added the discussions in the revised manuscript at Page18, Paragraph 1, as shown below:

"In addition, the evaluation of all the actuation and sensing performances of our devices were evaluated under room temperature. It has been reported that both breakdown strength and elastic modulus of DEs were sensitive to variations of the temperature depending on materials[53]. The temperature dependence needs to be evaluated if our DEA and DES are used at different environments in the future."

According to the following performance figures of merit, $S_z = \varepsilon_r \varepsilon_0 E^2 / Y$, a larger actuation strain would be achieved by increasing the applied electric field. As demonstrated by our results (as shown in the following **Fig. R6**), the PVCg-based actuators produced 1.7-fold higher area strain by increasing the driving voltage from 9.26 to 19.48 V/μm. Existing DEAs often requires high driving electrical fields to achieve large strain, which, however, could lead to the increased risks of polymer creep, current leakage, and electrical breakdown. In addition, it also needs a bulky, high-voltage power supply system, hampering its wide-spread applications [R24]. Therefore, a low driving voltage actuation is preferred for DEAs. In this study, the breakdown strength of plasticized PVCg was about 20 V/μm. The introduction of CEC and its loading concentration have limited impacts on the breakdown strength while it dramatically increased permittivity. Thus, a relative low working voltage of 9.09 V/μm was selected in order to prolong the working life of the device, achieve a stable performance while generating a desired large area strain [R25-R28]. Notably, such strain of 12.22 % was achieved under a very small pre-strain of 25 %, which was negligible when compared to other pre-strains such as 540 % and 400 % [R29, R30]. To address the reviewer's comments, we have added the relevant discussions in the revised manuscript at Page 8, Paragraph1, as shown below:

"Importantly, the breakdown strength was only slightly decreased by 10.4 % following the addition of CEC and kept almost constant for CEC/PVCg elastomers with different loading concentrations of CEC (**Fig. 3c** and **Supplementary Information Fig. S4**). The detected breakdown strength of CEC/PVCg elastomers with 9 wt% CEC was 19.53 V/μm. Thus, a much lower driving electric field of 9.09 V/μm was used in all of experiments in this study in order to prolong the working life of device and achieve a stable performance while generating a desired large area strain."

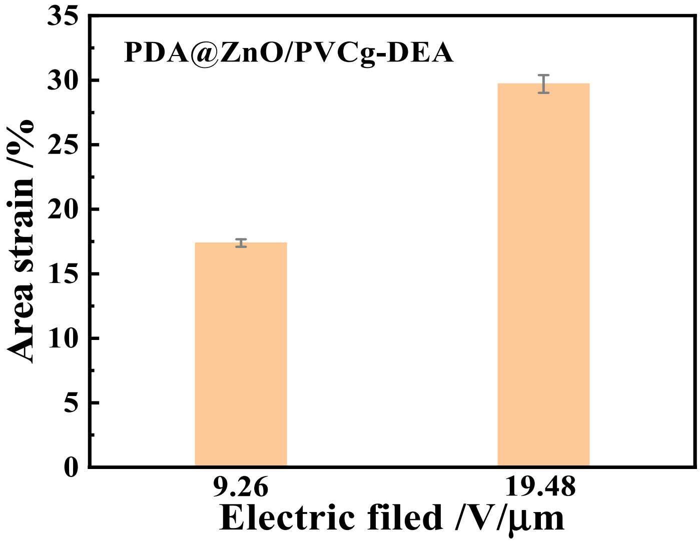

Fig. R6 Actuation strains of the dopamine (PDA) coating ZnO particles hybrid PVCg (PDA@ZnO/PVCg) actuators under driving electrical fields of 9.26 and 19.48 V/µm.

COMMENT #8: *The sensor work is well presented. It is important to put this work put in context of other capacitive sensor work, and perhaps provide some detail in the supplementary. There are also commercial sensors (e.g. Stretchsense) to compare with.*

Response: We thank the reviewer for her/his great suggestion. We have now compared our CEC/PVCg sensors with three different types of currently existing sensors, as shown in the following **Table R1**. One of the major drawbacks of existing sensors is that they cannot achieve high sensitivity over a wide range of deformation. For instance, the commercial strain gauge sensor has a very high sensitivity, but it was limited to a narrow range of distance *e.g.* 0-0.12 mm [R31]. By contrast, Our CEC/PVCg sensors can achieve a high sensitivity of 3.08 pF/mm over a much wider range of deformation, *i.e.* 0-14 mm. In addition, the relative high sensitivity of existing sensors has been heavily relying on the sophisticated structural design and micro/nano-manufacturing technologies [R32], such as micro-electromechanical systems (MEMS), resulting in the high complexity and high cost. By contrast, our CEC/PVCg sensor was fabricated by a simple mold-casting method and it costs ~ $ 2.00 per sensor. To address the reviewer's comment, we have added the following Table with new data as **Supplementary Table. S3** to compare our DEA against other commercially available sensors and relevant discussions in the revised manuscript at Page15, Paragraph 2, as shown below:

"In addition, our CEC/PVCg sensors showed a high sensitivity in a wide range of deformation, which cannot be achieved with commercially available sensors (**Supplementary Table S3)**. The high sensitivity of existing commercial sensors has been heavily relying on the sophisticated and complex design of their structures, which requires the costly and time-consuming micro-/nano-manufacturing techniques, such as micro-electromechanical systems (MEMS). By contrast, our CEC/PVCg sensors were fabricated by a very simple and highly accessible mold-casting method with an estimated cost of ~ $ 2.00 per sensor."

Table R1 Comparison of magnetic, strain, and capacitive sensors

	Magnetic sensors[R31]	Strain gauge sensors[R33]	Capacitive sensor[R31]	CEC/PVCg Sensor
Stiffness	Rigid	Flexible	Flexible and stretchable	Flexible and stretchable
Range	100nm-70mm	0-0.12 mm	10nm-10μm	0-14 mm
Sensitivity	1.68 V/mm	Very high	0.038-5.3 pF/mm	3.08 pF/mm
Linearity (R^2)	0.9994	0.98-0.99	0.97-0.9975	0.988-0.992
Cost	Expensive	Expensive	Moderate	Cheap
Complexity	Complex	Complex	Complex	Simple

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Response to Reviewer #3:

We thank the reviewer for her/his constructive comment. We are delighted that the reviewer found *"the physicochemical and electromechanical coupling property of the as-synthesized CEC/PVC were carefully studied*" and "*This work improved the performance of traditional PVC."* Here we address the comments and technical questions raised by the reviewer with new experimental results and analyses.

***COMMENT #1:** However, the demonstrated performance of actuation and sensing of the presented material are not convincing to me. The paper cannot meet the standard of Nature Communications unless the authors could clarify the advantage of the material compared to the existing material.*

Response: To address reviewer's concern, we have prepared the actuators and sensors using the most commonly used dielectric elastomers, *i.e.* PDMS and VHB 4910, and made a direct comparison of actuation and sensing performances between theirs and our CEC/PVCg. For actuation performance, the generated area strains of four types of actuators under the same pre-strain of 25 % and driving electric fields were first measured. As shown in the following **Fig. R1a**, our CEC/PVCg actuators produced the largest area strains among all actuators studied here. Our CEC/PVCg actuators showed 3.9-fold and 5-fold increase in area strain as compared to PVCg and PDMS actuators, respectively. Specifically, the area strains were 1.46-12.22 % for the CEC/PVCg actuators under the driving electrical fields of 5.45-9.09 V/µm, 0.81-3.15 % for the PVCg actuators under 5.45-9.09 V/µm, 0.65-2.44 % for the PDMS actuators under 5.85-9.07 V/µm, while VHB 4910 actuators cannot be activated under the driving electrical field < 12.5 V/µm, *i.e.* strain of 0 %. The area strains of VHB 4910 actuators were 0.72-3.45 % when the driving electrical field was further increased to 12.5-22.5 V/µm. Second, the actuation stability of actuators was evaluated by recording the displacements over 1000 cycles, *i.e.* 1000 seconds, and analyzing their relative displacement shifts (RDS) to quantify the stability. As shown in the following **Fig. R1b**, the RDS values of VHB 4910, PDMS, PVCg, and CEC/PVCg actuators over 1000 cycles were 136.09 %, 5.70 %, 59.40 %, and 7.78 %, respectively. Our CEC/PVCg actuators showed a very low shift of displacement, which was 94 % and 87 % reductions as compared to VHB 4910 and PVCg actuators. Therefore, our CEC/PVCg actuators in this study produced the largest actuation strain with the extremely low viscoelastic effects, demonstrating the significant improvements in actuation performances as compared to the existing DEAs, such as PDMS and VHB 4190.

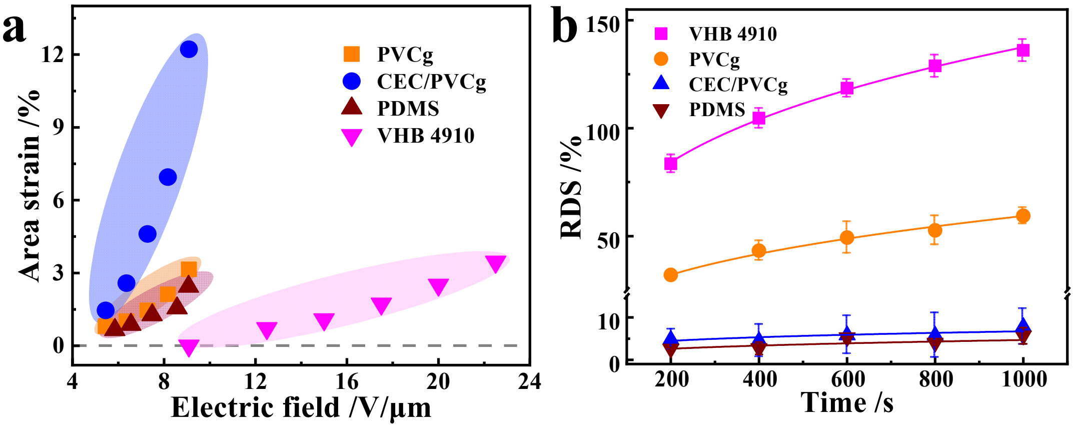

Fig. R1 The actuation performances of CEC/PVCg actuators as compared to existing PVCg, PDMS, and VHB 4910 actuators as measured by area strain (mean values) and actuation stability through the quantification of the relative displacement shift (RDS) over different number of cycles or seconds (1 second per cycle). The

RDSs were calculated by the following equation RDS = $\frac{|D-D_{creep}|}{D} \times 100\%$, where D is the amplitude of displacement and D_{creep} is the shift of the displacement.

For sensing performance (**Fig. R2**), the sensitivity of four types of sensors was first measured by calculating the slope of relative capacitance (output) as functions of displacement (input). As shown in **Fig. R2a**, our CEC/PVCg sensors demonstrated the highest sensitivity among all four types of sensors. For instance, the sensitivity (*S*) of CEC/PVCg sensors were 3.1-fold and 1.5-fold higher than PDMS and VHB 4910 sensors, respectively in the displacement ranges of 7-14 mm. Moreover, the stability of sensing performance was evaluated and quantified by analyzing the relative standard deviation (RSD) of relative capacitances over 1440 cycles of flection (2.5 s per cycle and 60 min in total), as shown in **Fig. R2b**. The results showed that our CEC/PVCg and PDMS sensors displayed much lower RSD values (5.75 % and 3.67 %), *i.e.* higher stability, than PVCg (9.84 %) and VHB 4910 (8.20 %) sensors. Altogether, our CEC/PVCg sensors demonstrated the superior overall sensing performances regarding of high sensitivity and stability over currently existing PVCg, PDMS, and VHB 4910 sensors.

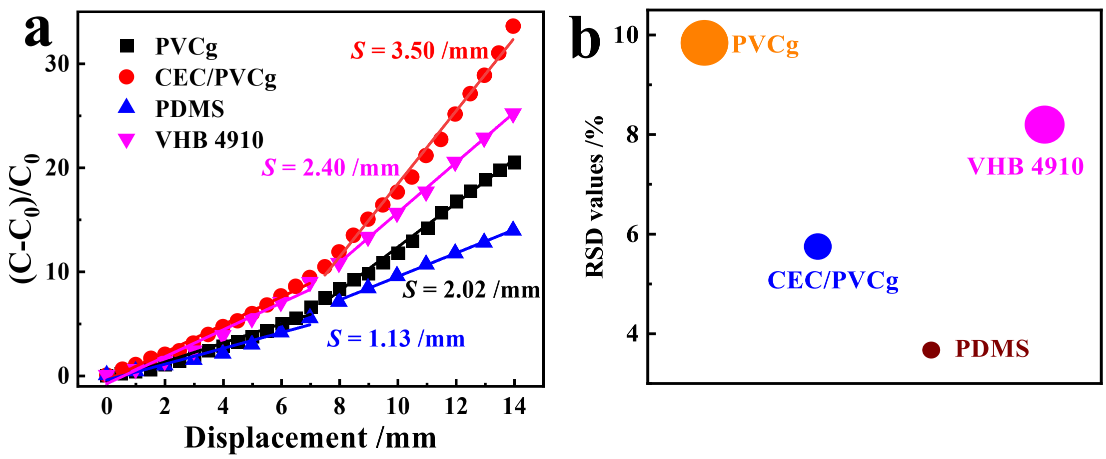

Fig. R2 (a) The relative capacitance (C-C_0/C_0) that was generated by PVCg, CEC/PVCg, PDMS, and VHB 4910 sensors as a function of the displacement. The tangential slope of the curve was defined as the sensitivity (*S*) of the sensors. The values of sensitivity in the displacement ranges of 7-14 mm were marked in the figure. (b) The relative standard deviation (RSD) of relative capacitances over 1440 cycles of flection (T = 2.5 s per cycle and 60 min in total). RSD = standard deviation of relative capacitance change / mean of relative capacitance change.

In summary, it is fair to make a conclusion that the CEC/PVCg dielectric elastomers developed in this study demonstrated the significant advantages over currently existing materials, *e.g.* PDMS and VHB 4910, for both actuation and sensing applications. We have now provided these new data and discussions in the revised manuscript at Page 11, Paragraph 3 as shown below:

"According to the strain model shown in **Supplementary Information Fig. S12**, the counted area strain generated by the CEC/PVCg actuators was 12.22 % (with 9 wt% CEC, under driving voltage of 9.09 V/µm, pre-strain of 25 %), which represents 3.9-fold increase as compared to the PVCg actuators (**Fig. 5b**). In addition, we prepared the commonly used PDMS and VHB-based actuators and measured their actuation strains under the same conditions as the comparison (**Fig. 5b, Supplementary Table S2,** and **Supplementary Information Fig. S15**). PDMS actuators produced strains of 0.65-2.44 % under electrical fields of 5.85-9.07 V/µm. VHB 4910 actuators could not be triggered, *i.e.* 0 % area strain, under electrical fields < 12.5 V/µm and produced only 0.72-3.45 % strains by further increasing electrical fields to 12.5-22.5 V/μm. Therefore, our CEC/PVCg actuators generated significantly larger actuation strains, *i.e.* > 5 times, than commonly used PDMS and VHB 4910 actuators, which was largely attributed to the augmentation of the electromechanical coupling sensitivity *k* of CEC/PVCg (**Fig. 3e**). In addition, the amplitude of flection displacement of our CEC/PVCg actuators was decreased with the increase of the driving frequency with fast response time (0.1-0.5 seconds) (**Supplementary Information Fig. S14**), which represented a characteristic electromechanical behavior of Maxwell field driven actuators. By contrast, it often took 5-20 seconds per cycle for creep-driven actuators, such as PVCg actuators[26]."

Page 12, Paragraph 2, as shown below:

"The displacements of the PVCg, CEC/PVCg, PDMS, and VHB 4910 actuators were measured and recorded over actuation for 1000 cycles, *i.e.* 1000 seconds (**Fig. 5c** and **Supplementary Information Fig. S16**). The CEC/PVCg and PDMS actuators produced remarkably stable displacement profiles. By contrast, apparent displacement shifts were observed over time for PVCg and VHB 4910 actuators. Relative displacement shifts (RDS) were calculated to quantify the viscoelastic effects (**Fig. 5d**). It was found that RDS values increased with time, *i.e.* number of actuation cycles, for VHB 4910 and PVCg actuators, while remaining almost constant for CEC/PVCg actuators. The relative shifts over 1000 cycles of CEC/PVCg actuators (7.78 % of RDS) represented 87 % and 94 % reductions as compared to PVCg (59.40 % of RDS) and VHB 4910 actuators (136.09 % of RDS). PDMS actuators (5.70 % of RDS) displayed similar viscoelastic drifts to CEC/PVCg actuators."

Page 14, Paragraph 2, as shown below:

"To demonstrate the sensing application of the CEC/PVCg elastomers, the periodic strain driven by a linear reciprocating actuator was applied to the prepared DES devices, including CEC/PVCg, PVCg, PDMS, and VHB 4910-based sensors (**Fig. 6a** and **Supplementary Information Fig. S17**). The profiles and periods of the capacitance signals (output) that were generated from both PVCg and CEC/PVCg sensors were identical to the strain signals (input) (**Fig. 6b** and **Supplementary Video S5**), suggesting that the mechanical signal can be accurately converted into the electric signal by the prepared sensors (**Supplementary Note 5)**. The CEC/PVCg sensors showed the fast response time, *e.g.* 1.0, 0.5, and 0.25 seconds under frequencies of 0.5-2.0 Hz (**Supplementary Information Fig. S19**). Notably, the CEC/PVCg sensors generated a significantly higher signal/noise ratio, baseline capacitance and capacitance width (*i.e.* ΔC, difference between peak capacitance C and baseline capacitance C_0) than the PVCg sensors because of the higher permittivity of CEC/PVCg matrix[36]. Moreover, the CEC/PVCg sensors showed the highest sensitivity (S) among four types of sensors that we studied here. For instance, the sensitivity of CEC/PVCg sensors was 3.1-fold, 1.5-fold, and 1.7-fold higher than PDMS, VHB 4910, and PVCg sensors, respectively in the displacement range of 7-14 mm (**Fig. 6c** and **Supplementary Information Fig. S18**)."

Page 14, Paragraph 3, as shown below:

"Notably, the capacitance generated by PVCg and VHB 4910 sensors showed an apparent drift over the recording time of 60 min (*i.e.* 1440 cycles) (**Fig. 6d** and **Supplementary Information Fig. S20**), which is in line with the previous report[28]. By contrast, CEC/PVCg and PDMS sensors produced remarkably stable capacitance signals without visible drift over at least 60 min, which was resulted from the low viscoelasticity and the inhibition on the rearrangement of their polar functions by the multiple molecular interactions. The relative standard deviation (RSD) of capacitances over 1440 cycles was analyzed to quantify the stability of sensors (**Fig. 6e**). The results showed that our CEC/PVCg and PDMS sensors displayed much lower RSD values (5.75 % and 3.67 %) of relative capacitances, *i.e.* higher stability, than PVCg (9.84 %) and VHB 4910 (8.2 %) sensors. Altogether, our CEC/PVCg sensors demonstrated the superior overall sensing performances regarding of high sensitivity and stability compared to existing PVCg, PDMS, and VHB 4910 sensors."

COMMENT #2: *The strategy of increasing permittivity and reducing viscosity in the material synthesis part sounds reasonable and interesting. However, if the material is aimed to serve for dielectric elastomer applications, these two indexes are apparently not dominated material properties.*

Response: There are two major factors in materials properties of dielectric elastomers (DEs), *i.e.* permittivity and mechanical properties, which are critical for their actuation and sensing applications according to the following performance figures of merits:

$$\textbf{Actuation strain}: \boldsymbol{S_Z = -\frac{\varepsilon_0\, \varepsilon_r\, E^2}{Y} = -kE^2}$$

$$\textbf{Sensing Capacitane}: \boldsymbol{C = \frac{\varepsilon_0\, \varepsilon_r\, A}{d}}$$

Where ε_0 and ε_r are the permittivity of free space and the relative permittivity of the elastomer matrix, respectively, Y is the Young's modulus, E is the applied electrical field, d is the thickness of the matrix film, k is electromechanical coupling sensitivity (ε/Y), and A is the area of electrodes.

Numerous studies have proposed and demonstrated that increasing dielectric permittivity and mechanical flexibility of DE matrix are critical and very effective strategies to improve their actuation and sensing performances [R1-R5]. For actuation applications, Pei *et a*l reviewed materials innovations and technological progress of DEAs, and concluded that "a high-performance DE should have sufficiently high elastic strains, a large dielectric permittivity, high dielectric strength, and an actuation stability without premature failure [R6,R7]". For example, the acrylate copolymer containing 4 vol% Al nanoparticles has a high dielectric permittivity of 8.4, which was increased by 78 % compared to the pure acrylate polymer, leading to significant increase in their breakdown strength and actuation pressure [R8]. Recently, Opris and co-workers successfully increased the dielectric permittivity of PDMS up to 18 by introducing the dipolar cyan group in PDMS precursor, which leads to an actuation strain of 5.4 % at very low electric field of 3.2 V/μm [R9]. For sensing applications, the magnitude of output capacitance is proportional to the dielectric permittivity of the elastomer [R5]. Increase of the DE permittivity would enlarge the magnitude of detected capacitance, resulting in higher signal/noise ratio and sensitivity.

On the other hand, existing methods, such as the introduction of plasticizer into DEs, for lowering the Young's moduli and increasing flexibility of DEs are often associated with the increase of viscoelastic effects. High viscoelastic effects of DEs would result in evident mechanical loss, stress relaxation, and viscoelastic hysteresis, leading to instability of output signals over time as well as delayed response [R10,R11]. For instance, the creep-driven PVCg actuators showed more frequent strain drifts and the delayed electromechanical response as compared to actuators that are primarily driven by the Maxwell field [R12]. As a sensor, the viscoelastic PVCg are often associated with a large signal shift of bulk permittivity and output signals over time because of the random rearrangement of polar groups of PVC chain during stretching [R5]. However, such detrimental impacts of high viscoelasticity have been overlooked in the filed for long time.

In this study, it was found that the introduction of CEC into the commonly used PVCg elastomers not only significantly increased the dielectric permittivity, leading to dramatic enhancement of actuation strain and sensitivity, but also effectively mitigated their viscoelastic effects, resulting in highly stable actuation and sensing performance over long time. Importantly, we have demonstrated the superior performance of our CEC/PVCg-based DEA and DES in both actuation and sensing as compared to existing and commonly used PDMS and VHB 4910-based actuators (**Fig. R1**) and sensors (**Fig. R2**). Altogether, we believe the concurrent increase of dielectric permittivity and reduction of viscoelastic effects of DEs are critical and effective strategy for the improvement of DE in actuation and sensing applications.

To address the reviewer's concern, we have now provided our new data about the direct comparison of actuation and sensing performances between our CEC/PVCg and commonly used dielectric elastomers, *e.g.* PDMS and VHB 4910, which directly demonstrated the efficiency of our strategy, in the revised manuscript as **Fig. 5b**, **5d**, and **Fig. 6c**, **6e**. We have also stressed the importance of increasing dielectric permittivity and reducing viscoelasticity in DEA and DES applications by adding more discussion in the revised manuscript, at Page 3, Paragraph 2 and Page 4, Paragraph 2, as shown below:

"Numerous efforts have been devoted to increase the dielectric permittivity and mechanical flexibility to generate a large actuation under relatively low driving voltages[11-13,16-19]. For instance, the seminal work from Kofod's group enhanced the relative permittivity of the PDMS elastomer from 3.0 to 5.9 and decreased the elastic modulus from 1900 to 550 kPa by grafting small molecules with high dipole moment to the elastomer matrix, leading to significant improvement of their electromechanical performances[19]. In addition, the reduction of the film thickness is an alternative method to improve the actuation performance[20-23]. For example, Shea and his co-workers demonstrated that the actuation strain of 7.5 % could be generated with a 3 μm thick film under a driving voltage of 245 V[20]. By contrast, it required much higher driving voltage of 3.3 kV to generate the same actuation strain with the 30 μm thick film. Despite these positive outcomes, thin film actuators often require complicated fabrication processes and are associated with high prevalence of an electromechanical instability[24]."

"Notably, such viscoelastic effects are widely presented in other elastomers such as VHB[12,32], polyurethane (PU)[33], and polyurethane acrylate (PUA)[34]. Although the creep could be utilized to trigger different mechanisms of deformation, such as bending, contracting, and crawling, it results in evident mechanical loss, stress relaxation, and viscoelastic hysteresis, leading to instability of output signals over time as well as delayed response[35]. For instance, the creep-driven PVCg actuators[25] show more frequent jump of output signals and the delayed electromechanical response as compared to actuators that are primarily driven by the Maxwell force. The viscoelasticity of PVCg sensors often leads to a large drift of bulk permittivity and output signals over time because of the random rearrangement of polar groups of PVC chain during stretching[36]. However, the viscoelastic effects of PVCg-based DEs have been largely over-looked. The mitigation of their viscoelastic effects without compromising their electromechanical functions remains warranted."

COMMENT #3: *As shown in Fig. 5, the actuation strain of the new material falls within the order of 10 %. A lot of existing studies have shown that various of materials without careful optimization and complicated synthesis can easily achieve this actuation level. Similar concerns apply for the sensing demo in this work.*

Response: For actuation applications, the generated actuation strains can have very large differences when they are measured under different settings, *e.g.* different driving electric fields and pre-strains. The large area strains often require high driving electric fields and large pre-strains. For instance, the commercial VHB elastomer generated area strain of 215 % under very high electrical field of 239 V/μm and large pre-strain of 540 % [R13] while producing much smaller area strain of 34 % under electrical field of 70 V/μm and pre-strain of 400 % [R14]. However, the high driving electric field (> 20 V/μm) could lead to the high risks of current leakage [R15] and electrical breakdown [R16]. In addition, a high value of several kilovolts arises safety issues and brings about the problem of using a bulky high-voltage power supply system [R17]. The generation of large pre-strains can add extra complexity in the fabrication process of devices, reduce the reproducibility, and increase risks of mechanical damage of the films. Therefore, the generation of large actuation under low driving electric field and pre-strain is highly desirable for DEA application while it has been a long-standing challenge. In this study, we evaluated the actuation performances of actuators under a low driving electric field of 9.09 V/μm and a small pre-strain of 25 %, which was negligible when compared to other actuators [R13,R14]. To address the reviewer's concern, we have fabricated actuators using the most commonly used dielectric elastomers, including PDMS and VHB 4910, and evaluated their actuation performance under the same settings as our CEC/PVCg. As shown in **Fig. R1a**, our CEC/PVCg actuators generated remarkably larger area strains (12.22 %) than PDMS (2.44 %) and VHB 4910 (no area strain, *i.e.* 0 %) actuators under low driving electric field of 9.09 V/μm and small pre-strains of 25 %. Notably, VHB 4910 actuators generated only 3.45 % area strain by further increasing the driving electric field to 22.5 V/μm. Moreover, our CEC/PVCg actuators exhibited a low mechanical loss and a high actuation stability due to the significant mitigation of viscoelastic effects. As shown in the **Fig. R1b**, the VHB 4910 and pristine PVCg actuators exhibited apparent shifts in displacement over 1000 actuation cycles, while the CEC/PVCg and PDMS actuators did not show visible shift. The relative displacement shifts (RDS) were calculated over 1000 cycles to quantify their actuation stability. Our CEC/PVCg actuators showed the high actuation stability with the low RDS value (7.78 % of RDS), which was 87 % and 94 % reductions as compared to PVCg (59.40 % of RDS) and VHB 4910 actuators (136.09 % of RDS).

For sensing applications, we evaluated the sensitivity and stability of output signals of PDMS, VHB 4910, PVCg, and CEC/PVCg sensors under the same conditions to make a direct comparison. As shown in **Fig. R2** and detailed description in our *responses to the reviewer's COMMENT #1*, our CEC/PVCg sensors showed the highest sensitivity among all four types of sensors and very stable signal outputs over 1440 cycles of flection.

Altogether, it is fair to make a conclusion that the CEC/PVCg dielectric elastomers presented in this study demonstrated the superior performances in both actuation and sensing over currently existing dielectric elastomers, such as PDMS and VHB acrylic based materials. We have now provided the data of direct comparisons in actuation and sensing performance among different materials in **Fig. 5** and **Fig. 6** in the revised manuscript. We also provided the relevant discussions in the revised manuscript at Page 11, Paragraph 3, Page 12, Paragraph 2, Page 14, Paragraph 2, and Page 14, Paragraph 3, which has been shown in *our response to the reviewer's COMMENT #1*.

COMMENT #4: *In the introduction, the authors pointed out that the VHB material, widely used in literature, has a clear drawback in viscous property. However, the synthesized new material improves its viscous property compared to VHB but significantly sacrifices its ability of large actuation, which, to the reviewer's opinion, is not satisfactory for dielectric elastomer applications.*

Response: The large actuation strains of VHB materials as reported from previous literature are largely relying on the use of very high driving electric fields and large pre-strains as applied in the evaluation. For instance, the commercial VHB elastomer generated area strain of 215 % with very high electrical field of 239 V/μm and large pre-strain of 540 % [R13] while producing much smaller area strain of 34 % with electrical field of 70 V/μm and pre-strain of 400 % [R14]. However, the generation of large actuation under low driving electric fields and small pre-strains have been highly desirable for DEA applications [R18]. To make a fair comparison between VHB 4910 and our CEC/PVCg, we have evaluated their actuation performances under the same settings. As shown in **Fig. R1a**, our CEC/PVCg actuators showed significantly larger actuation strain of 12.22 %

than the VHB 4910 actuators with area strain of 0 % under a low driving electric field of 9.09 V/μm and a small pre-strain of 25 %. The VHB 4910 actuators produced an area strain of 3.45 % only by further increasing the driving electric field up to 22.5 V/μm. Moreover, our CEC/PVCg actuators exhibited a lower mechanical loss and a higher actuation stability than VHB 4910 actuators due to the significant mitigation of their viscoelastic effects. As shown in the **Fig. R1b**, the relative displacement shifts (RDS) were analyzed from the recorded displacement profiles over 1000 cycles to quantify the actuation stability. VHB 4910 actuators (136.09 % shifts) displayed 18 times higher relative shifts than our CEC/PVCg actuators (7.78 % shifts). In addition, our CEC/PVCg sensors also showed higher sensitivity and produced much more stable sensing signals than VHB 4910 sensors (**Fig. R2**). Therefore, we believe these new data fully support our conclusion that the CEC/PVCg dielectric elastomer in this study exhibited superior performances in both actuation and sensing over commercially available VHB acrylic elastomers.

We have now provided the data of direct comparisons in actuation and sensing performance among different materials in **Fig. 5** and **Fig. 6** in the revised manuscript. We also provided the relevant discussions in the revised manuscript at Page 11, Paragraph 3, Page 12, Paragraph 2, Page 14, Paragraph 2, and Page 14, Paragraph 3, which has been shown in *our response to the reviewer's COMMENT #1*.

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Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
I have carefully read the response from the authors according to the suggestion of reviewer 2. all of 6 comments is explained or added some new contents to make a good understanding for potential readers. Though the dielectric material in this manuscript has also several disadvantages, the content in this revision is very rich and gives a new insight to learn the dielectric elastomer actuator. In this case, I suggest this revision with high quality can be considered to accept.

Reviewer #3:
Remarks to the Author:
The authors have conducted detailed experiments to compare the new PVC material with the existing materials. The results are encouraging. The paper can be recommended subject to the following minor revisions.

In the revised manuscript, the authros compared the actuation strain of PVC with VHB 4910 at the same electric field, which was called a "fair comparison", and stated the actuation performance is better, which I still do not fully agree. The reason is that as an dielectric actuation material, the energy conversion density is proportional to permittivity*electric field sqaured. The key material parameter is the electrical breakdown strength. As the authors cited, many literatures have made efforts in improving the permittivity and lowering modulus but not in breakdown strength.That's why VHB has such an outstanding high energy conversion density in actuation as well as in energy harvesting. A detailed review can be seen in Lu et al. Mechanics of dielectric elastomer structures A review, 2020. I would suggest that the authors frankly admit that the new material is better than VHB in low-voltage actuation and low hysteresis but not in a comprehensive manner. This advantage could be helpful in many applications that do not pursue large deformation or high energy density.

Responses to Reviewer #3:

COMMENT: *A detailed review can be seen in Lu et al. Mechanics of dielectric elastomer structures A review, 2020. I would suggest that the authors frankly admit that the new material is better than VHB in low-voltage actuation and low hysteresis but not in a comprehensive manner. This advantage could be helpful in many applications that do not pursue large deformation or high energy density.*

Response: We thanks for and agreed with the reviewer's comments on the comprehensive comparison between our dielectric elastomer and commercial VHB4910. Following the reviewer's suggestion, we have revised our statements throughout the manuscript by stating that "*Our CEC/PVCg actuators demonstrate superior actuation performances over the existing DE actuators under low electrical fields*". We have added more discussion on the limitation of our material compared to existing VHB4910 and PDMS in the revised manuscript at Page 4, Paragraph 3, Page 11, Paragraph 3, Page 17, Paragraph 2, and Page 17, Paragraph 3 as shown below:

"In this study, we reported a valuable strategy to produce a PVCg-based dielectric elastomer with unprecedented properties, i.e. high permittivity, low viscoelasticity, and high flexibility, and further demonstrated its superior performances in both actuation and sensing applications, especially under low driving electrical fields."

*"Therefore, our CEC/PVCg actuators generated significantly larger actuation strains, i.e. > 5 times, than commonly used PDMS and VHB 4910 actuators within < 22.5 V/μm electrical field, which was largely attributed to the augmentation of the electromechanical coupling sensitivity k of CEC/PVCg (**Fig. 3e**)."*

"As a result, the CEC/PVCg actuators demonstrate superior actuation performance over the existing DE actuators, such as PDMS and VHB 4910, under low driving electrical fields."

"One limitation of current devices is the intrinsic low breakdown strength, i.e. 21.79 V/μm, of the traditional plasticized PVCg, as compared to the existing PDMS and VHB 4910. The use of higher driving electrical fields can offer high energy conversion density and energy harvesting with VHB 4910-based actuators [51].