

Article

Enhanced Electromechanical Performance of Dielectric Elastomer by Co-Crosslinking of Silane-Functionalized TiO_2 with Polyacrylate

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Abstract

Dielectric elastomer actuators (DEAs) are attracting much attention as candidates for next-generation flexible actuation. Among various DE matrices, polyacrylate rubber (AR) is especially promising owing to their intrinsically high dielectric constant (ϵ_r) and good mechanical performance. In particular, its mechanical behavior is close to that of porcine bladder tissue, making it a potentially good material for soft biomedical actuators for artificial bladder constructs. To achieve high actuated strain, which requires high ϵ_r , high breakdown strength, and low elastic modulus, an AR DE composite filled with silane-functionalized TiO_2 was fabricated, exhibiting good electromechanical performance enabled by strengthened interfacial polarization. To improve compatibility between TiO_2 and AR matrix, TiO_2 was preferentially modified with a silane coupling agent (CA) that features a double bond as its functional group, which can be introduced on TiO_2 surface and participate in vulcanization with AR, thereby forming co-crosslinking bridges that strengthen interfacial bonding, improve filler dispersion, and increase interfacial polarizability within the matrix. As a result, at relatively low filler loadings, the composite exhibits a significantly increased ϵ_r , while maintaining low modulus, low dielectric loss and high elasticity. The 10 CA@ TiO_2 /AR composite exhibits a maximal actuated strain of 7.9% at 31.9 kV/mm without pre-stretch, which is 1.48 times that of pure AR and 1.32 times that of the 10 TiO_2 /AR composite.

Keywords: dielectric elastomer (DE); polyacrylate rubber; TiO_2 ; co-crosslinking; electromechanical performance



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1. Introduction

Flexible actuators capable of generating large, rapid, and energy-efficient deformation are pivotal for the next-generation of microrobots, wearable and biomedical devices, and adaptive optical systems [1,2]. Among electroactive polymer technologies, dielectric elastomer actuators (DEAs) have emerged as a leading option thanks to their lightweight, large actuated strain, high output force, and remarkable energy density [3,4]. These characteristics make them suitable for use in artificial bladder applications, offering potential therapeutic benefits for patients with bladder dysfunction by helping restore normal urinary storage and release capabilities. A DEA comprises a flexible dielectric elastomer (DE) film sandwiched between compliant electrodes (Figure 1a). When a high voltage is applied,

the resulting Maxwell stress compresses the film in thickness and drives in-plane expansion, directly converting electrical input into mechanical work. The actuated strain (S_a) of a DE can be evaluated using the widely accepted theory proposed by Perline et al. [5].

$$S_a = -\frac{P}{Y} = -\frac{\epsilon_0 \epsilon_r E^2}{Y} \quad (1)$$

where ϵ_0 is the vacuum permittivity, ϵ_r , and Y are the dielectric constant and elastic modulus of DE, respectively, and E is the applied electric field. According to Equation (1), an ideal DE with excellent electromechanical performance should possess a high ϵ_r and a low Y , which together yield high electromechanical sensitivity ($\beta = \epsilon_r/Y$), as well as a high electrical breakdown strength (E_b) [6,7]. Such an ideal DE can further be adapted for artificial bladder applications, where large, rapid, and reliable long-term actuation performance is essential.

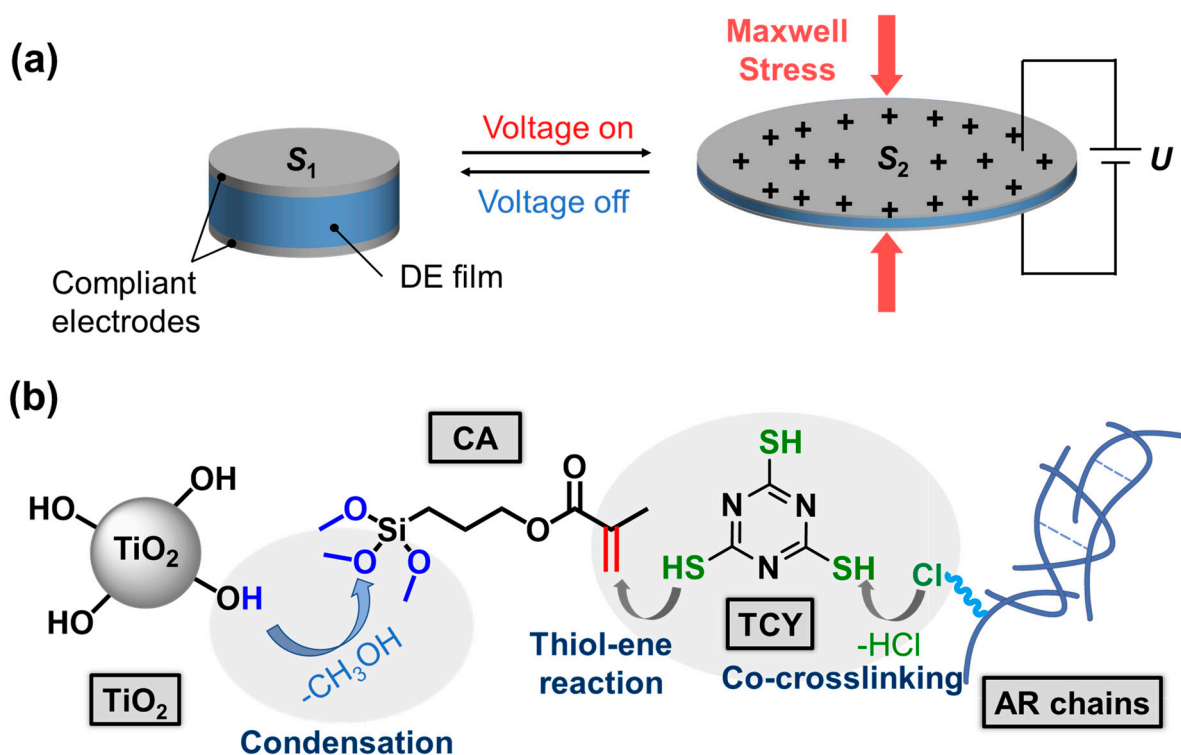


Figure 1. (a) The construction and electrical actuation mechanism of a typical DEA. (b) Schematic illustration of the fabrication method for CA@TiO₂/AR composites.

Thanks to increased chain mobility from plasticizers, the Y of DEs can be reduced by introducing additives such as polyethylene glycol [8], silicone oil [9], epoxidized soybean oil [10], and ionic liquid [11] into the polymer matrix. For example, Ruan et al. used tri-*n*-butyl phosphate to plasticize brominated butyl rubber (BIIR), which weakened intermolecular interactions among BIIR chains and disrupted the filler network, thereby lowering Y [12]. However, introducing plasticizers can cause viscoelastic issues such as creep, stress relaxation and increase hysteresis loss [13]. In addition, limited compatibility with many polymer matrices promotes migration of plasticizer molecules and shortens device service life.

Beyond mechanical softening strategies, increasing ϵ_r is a practical way to enhance the electromechanical response of DEs. This can be achieved through three main strategies. A representative choice is using polarized rubber matrix such as polyacrylate rubber (AR) [14,15], with abundant ester groups providing strong dipolar polarization or grafting dipole functional groups onto the non-polar rubber molecular chains. The second strategy

is to blend the DE matrix with conductive fillers such as carbon nanotubes [16], carbon nanospheres [17], and graphene [18]. Thanks to interfacial polarization and microcapacitor networks formed by sub-percolating conductive fillers, ϵ_r increases sharply as the filler content approaches the percolation threshold [19]. The drawback is that they also raise electrical conductivity, which markedly lowers E_b and increases dielectric loss (ϵ''), reducing energy conversion efficiency. The third strategy is to introduce high- ϵ_r ceramic fillers such as TiO_2 [20,21] or BaTiO_3 [22], a widely used and effective approach. However, their high surface energy promotes agglomeration, which degrades dispersion and easily forms defect sites, thereby harming electromechanical performance. Surface modification of the inorganic fillers can mitigate these issues by improving compatibility and dispersion [23]. For example, Yang et al. coated TiO_2 with poly(dopamine) to strengthen interfacial adhesion with the rubber matrix, yielding simultaneous improvements in mechanical integrity and dielectric properties in polymer composites [24]. A disadvantage is that the cost of dopamine remains a barrier to large-scale industrial use [25].

In this study, we aim to investigate the potential application of DEAs in artificial bladders. For this purpose, a commercial chlorine cure-site AR was selected owing to its intrinsically high ϵ_r and good mechanical performance with its mechanical behavior close to that of porcine bladder tissue (Figure S1). This AR consists mainly of poly(ethyl acrylate) and contains approximately 5 wt% chlorine cure-site monomer [26]. These chlorine sites allow curing with trithiocyanuric acid (TCY) via nucleophilic substitution to form stable thioether crosslinking and a robust network [27]. However, the ester-rich, polar matrix requires tailored interfaces between the filler and the matrix to achieve good dispersion and electromechanical performance. In this work, TiO_2 modified with a low-cost silane coupling agent (CA), γ -methacryloxypropyl trimethoxy silane, was employed to enhance the electromechanical performance of AR DEs (Figure 1b). Within AR composites, the purpose of using CA@TiO_2 was to form a siloxane-rich interphase on the filler surface and create co-crosslinking bridges to the AR network via reactions of its double bond with thiol-bearing crosslinkers. This interfacial architecture improves compatibility and stabilizes dispersion. As a result, the interfacial area increases and interfacial polarization strengthens, yielding a higher ϵ_r at the same filler content. Meanwhile, fewer defects and a more uniform local field raise E_b . At low content (10 wt%) of CA@TiO_2 , the ϵ_r increases, while maintaining low ϵ'' low dielectric loss and low hysteresis loss (high elasticity), the electromechanical sensitivity ($\beta = \epsilon_r/Y$) increases, delivering significantly enhanced actuated strain.

2. Experimental

2.1. Materials

Polyacrylate rubber (grade AR71) was supplied by Zeon Corporation (Tokyo, Japan). TiO_2 nanoparticles with an average particle size of 15–25 nm were purchased from Beijing Dekedaojin Technology Co., Ltd. (Beijing, China). Trithiocyanuric acid (TCY, 99%, Shanghai Jinghai Chemical Co., Ltd., Shanghai, China), 2-mercaptobenzothiazole (BZ, 99%, Kemiou Chemical Co., Ltd., Tianjin, China), γ -methacryloxypropyl trimethoxy silane (CA, 99%, Aladdin, Shanghai, China) and anhydrous ethanol ($\geq 99.7\%$, Beijing Chemical Works, Beijing, China) were used as received without further purification.

2.2. Preparation of CA@TiO_2 Filler

TiO_2 particles were first mixed with 5 wt% CA (dissolved in ethanol) using a high-speed mixer at a rotating speed of 30,000 r/min for 4 min, then placed in an oven and dried at 80 °C for 2 h. The pretreated TiO_2 was subsequently washed three times with ethanol and dried again at 80 °C for 8 h to obtain CA@TiO_2 filler.

2.3. Preparation of Composites

TiO₂/AR or CA@TiO₂/AR compounds containing TiO₂ or CA@TiO₂ (0, 10, 30, or 50 phr), TCY (0.25 phr, vulcanizing agent), BZ (0.5 phr, accelerator), and AR (100 phr) were prepared by physical mixing on a 6-inch two-roll mill at room temperature for 30 min, with detailed formulations provided in Table S1. The TiO₂/AR and CA@TiO₂/AR dielectric composite films were obtained by vulcanizing the corresponding compounds under a pressure of 15 MPa at 170 °C.

2.4. Characterization Methods

See the Supporting Information.

3. Results and Discussion

3.1. Structural Characterization of CA@TiO₂

The CA-functionalized TiO₂ filler is schematically illustrated in Figure 2a, while the corresponding FT-IR spectra of TiO₂ and CA@TiO₂ are presented in Figure 2b. Compared with the pristine TiO₂ filler, the CA@TiO₂ exhibits new characteristic peaks at 1162 cm^{−1} and 1197 cm^{−1}, corresponding to the Si-O-Si bond stretching vibration of CA, as well as peaks at 2981 cm^{−1} and 2865 cm^{−1}, which are attributed to -CH₂- stretching vibrations from CA. In addition, new absorption peaks appear at 912 cm^{−1}, 1085 cm^{−1}, 1640 cm^{−1}, and 1721 cm^{−1}, representing the stretching vibrations of Si-OH, Si-O-C, C=C, and C=O groups of CA, respectively [28]. These results confirm that CA was successfully grafted onto the TiO₂ surface. TGA was used to determine the grafting amount of CA on the TiO₂ surface. As shown in Figure 2c, when heated from 200 to 650 °C, pristine TiO₂ and CA@TiO₂ show weight losses of 1.5% and 3.8%, respectively. Based on Equation (S1), the grafting density on TiO₂ was calculated as 0.12 mmol/g.

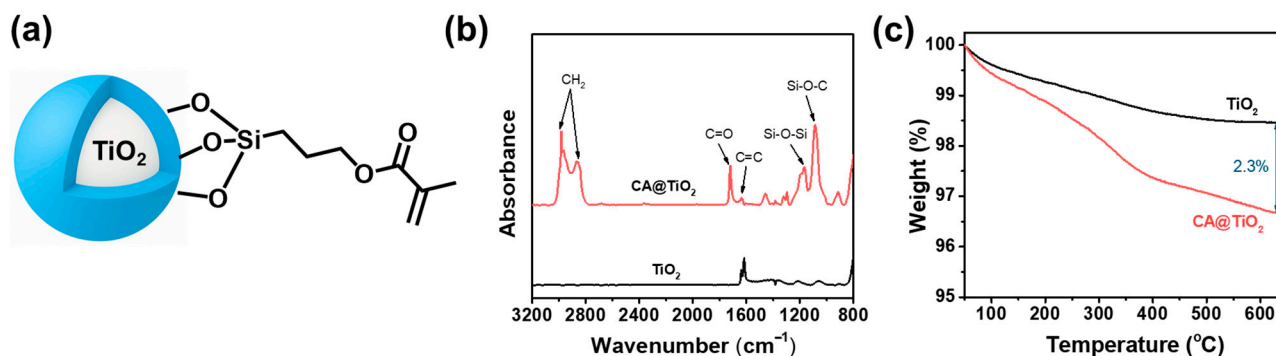


Figure 2. (a) Schematic representation of CA@TiO₂ filler. (b) FTIR spectra and (c) TGA thermograms of pristine TiO₂ and CA@TiO₂ filler.

3.2. Microstructure of AR Composites

The morphology of TiO₂/AR and CA@TiO₂/AR composites was examined by SEM, and the results are shown in Figure 3. The unmodified TiO₂ exhibits poor dispersion, with pronounced particle agglomeration due to the large surface energy mismatch between TiO₂ and the AR matrix. In contrast, CA@TiO₂ fillers are dispersed more uniformly in the AR matrix. This improvement arises from CA serving as a co-crosslinking bridge through interfacial covalent bonding, which enhances the compatibility between TiO₂ particles and the AR matrix.

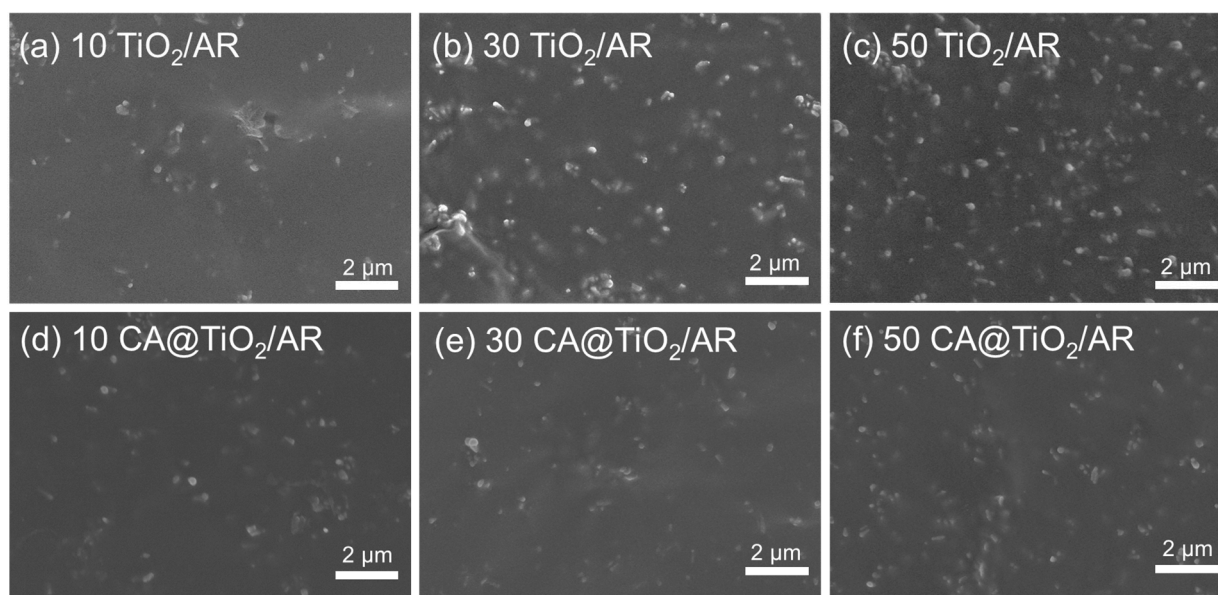


Figure 3. SEM micrographs of AR composites filled with (a) 10 phr TiO_2 , (b) 30 phr TiO_2 , (c) 50 phr TiO_2 , (d) 10 phr CA@TiO_2 (e) 30 phr CA@TiO_2 and (f) 50 phr CA@TiO_2 .

3.3. Mechanical Properties of AR Composites

The stress–strain curves of TiO_2/AR and $\text{CA@TiO}_2/\text{AR}$ composites are shown in Figures 4a and 4b, respectively, and the mechanical properties of all AR composites are summarized in Table S2. The tensile strength of the AR composites exceeds that of pure AR and increases with filler content, owing to the reinforcing effect of rigid fillers. Specifically, the filler particles can promote stress transfer from the flexible AR matrix to the rigid inorganic filler during stretching. At the same filler loading, the $\text{CA@TiO}_2/\text{AR}$ composites exhibit higher tensile strength and elongation at break than the TiO_2/AR composites, owing to the better filler dispersion and stronger filler–matrix interaction, which reduce stress concentration and improve load transfer efficiency.

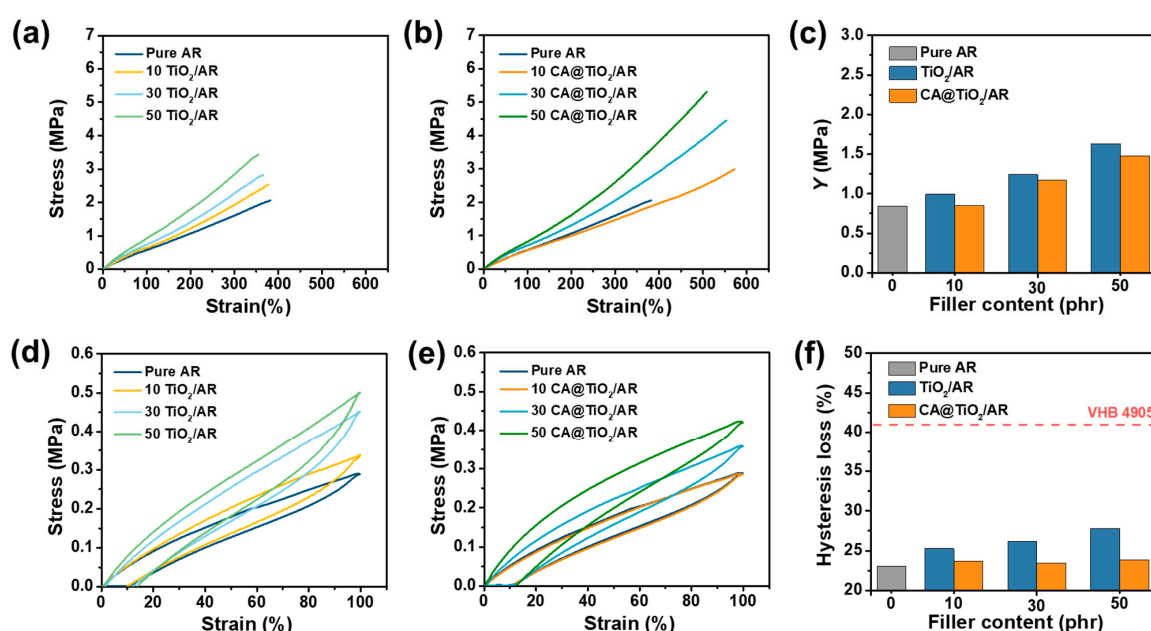


Figure 4. Stress–strain curves of (a) TiO_2/AR and (b) $\text{CA@TiO}_2/\text{AR}$ composites. (c) Y of all AR composites. Cyclic stress–strain curves of (d) TiO_2/AR and (e) $\text{CA@TiO}_2/\text{AR}$ composites. (f) Hysteresis loss of all AR composites.

The γ of the AR composites is shown in Figure 4c. At 10 phr TiO_2 or CA@TiO_2 , γ differs only slightly from pure AR, indicating that the filler network structure has not been formed yet under low filler fraction. At higher contents, γ increases significantly because of the formation of filler networks that stiffen the matrix. In addition, at the same filler content, the γ of $\text{CA@TiO}_2/\text{AR}$ composites is lower than that of TiO_2/AR composites, which is attributed to better dispersion and a more compliant siloxane interphase that reduces the compactness of the network structure.

In addition, low hysteresis loss (also known as viscoelastic hysteresis) is an important evaluation criterion for elasticity of DEs. The cyclic stress–strain curves of TiO_2/AR and $\text{CA@TiO}_2/\text{AR}$ composites are shown in Figure 4d and 4e, respectively, and the hysteresis loss of all AR composites are summarized in Figure 4f. Whether or not the fillers are surface-modified, the hysteresis loss of AR composites increases with TiO_2 content, which is attributed to increased interfacial friction, interfacial sliding, and the strengthening of filler network at higher loadings. At the same filler content, $\text{CA@TiO}_2/\text{AR}$ composites show a lower hysteresis loss than TiO_2/AR composites due to a robust interphase and the better dispersion of TiO_2 . These interfacial interactions provide stronger anchoring points and suppress interfacial slippage, facilitating chain recovery and reducing energy dissipation. The hysteresis loss of the 10 $\text{CA@TiO}_2/\text{AR}$ composite is comparable to that of pure AR (23%), indicating the high elasticity being maintained. Moreover, the hysteresis loss of all AR composites remains below 28%, well below that of VHB 4905 (41%), a widely used acrylic-based DE [29].

3.4. Dielectric Properties of AR Composites

The ϵ_r of TiO_2/AR and $\text{CA@TiO}_2/\text{AR}$ composites over 10^{-1} – 10^6 Hz is shown in Figure 5a and 5b, respectively. The ϵ_r of all composites decreases with increasing frequency, indicating strong frequency dependence arising from dipolar relaxation. At low frequencies, dipoles associated with the ester side groups in AR can follow the alternating field, yielding high polarization and therefore higher ϵ_r . As the frequency increases, dipole reorientation cannot keep pace, polarization diminishes, and ϵ_r falls. In addition, ϵ_r increases with TiO_2 content, whether or not the fillers are surface-modified, because of interfacial polarization between the dielectric filler and the rubber matrix.

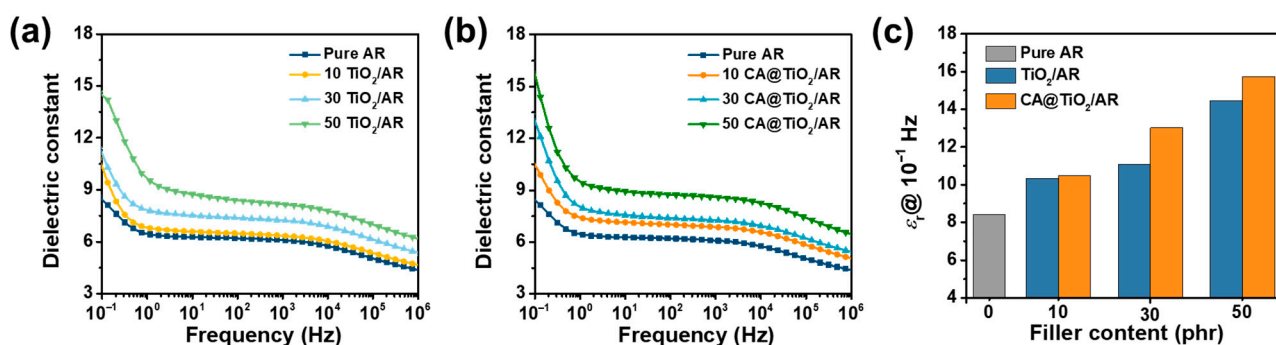


Figure 5. ϵ_r versus frequency of (a) TiO_2/AR and (b) $\text{CA@TiO}_2/\text{AR}$ composites. (c) ϵ_r @ 10^{-1} Hz of all AR composites.

More importantly, the ϵ_r @ 10^{-1} Hz of $\text{CA@TiO}_2/\text{AR}$ composites is higher than that of TiO_2/AR composites at the same filler content (Figure 5c). This enhancement mainly originates from the improved dispersion of CA@TiO_2 in the AR matrix, which increases the number of effective polymer–filler phase interfaces and thereby enhances the overall contribution of interfacial polarization. Notably, this trend is different from our previous reported SiO_2/SiR system, where the ϵ_r @ 10^{-1} Hz of $\text{CA@SiO}_2/\text{SiR}$ composites is lower than that of SiO_2/SiR composites at the same filler loading [30]. Although surface modification

in the CA@SiO₂/SiR system improves dispersion and enlarges the total interfacial area, the CA layer weakens the interfacial polarization at each individual SiO₂-SiR interface, leading to an overall decrease in ϵ_r . These opposite trends indicate a trade-off between filler dispersion and interfacial structure in determining the overall interfacial polarization contribution. As a result, the 50 CA@TiO₂/AR composite shows the highest ϵ_r @ 10⁻¹ Hz of 15.73, which is 1.87 times than that of neat AR (8.43).

Consistent with the frequency dependence of ϵ_r , the ϵ'' of all composites also exhibits an obvious frequency-dependent behavior (Figure 6). At low frequencies, the ϵ'' is relatively high, mainly because interfacial polarization and dipolar relaxation can fully respond to the slowly varying electric field, resulting in greater energy dissipation. With increasing frequency, the ϵ'' gradually decreases because the dipoles and interfacial charges can no longer follow the rapid alternation of the electric field in a timely manner. Therefore, the polarization process becomes progressively suppressed, leading to reduced ϵ'' at higher frequencies.

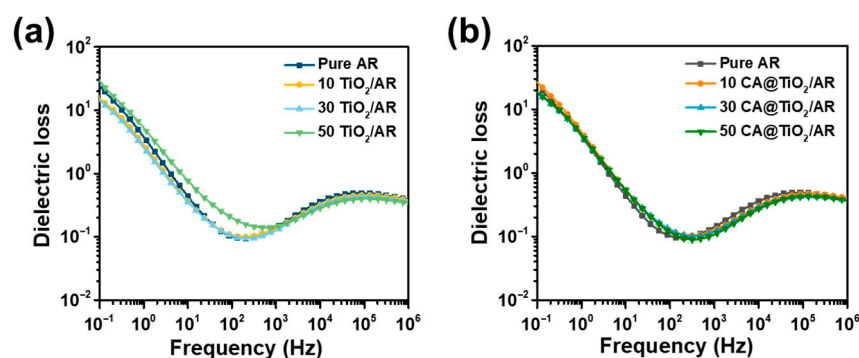


Figure 6. ϵ'' versus frequency at room temperature of (a) TiO₂/AR and (b) CA@TiO₂/AR composites.

To clarify the physical origin of the dielectric properties, the dielectric relaxation behavior of pure AR and CA@TiO₂/AR composites was investigated over a wide temperature range at three low frequencies (0.1, 1, and 10 Hz), which are relevant to the expected operating conditions of the artificial bladder (Figure 7) [31,32]. As shown in Figure 7a–c, the ϵ_r of all samples gradually increases with increasing temperature. In addition, the composites with higher CA@TiO₂ content exhibit higher ϵ_r , especially at low frequencies. This behavior can be attributed to the enhanced interfacial polarization and the increased mobility of polar groups at elevated temperature. Meanwhile, as shown in Figure 7d–f, all samples exhibit a broad temperature-dependent dielectric-loss relaxation feature. However, no obvious change in peak position or peak intensity with increasing filler content, suggesting that the introduction of CA@TiO₂ does not significantly alter the dipolar relaxation behavior of the AR matrix.

In addition, the E_b is a key parameter that limits the maximum actuated strain, as indicated by Equation (1). As shown in Figure 8a,b, E_b of all composites increases with TiO₂ content for both pristine and CA-modified fillers, consistent with previous reports [22,33]. This can be attributed to the introduction of rigid TiO₂ particles, which restrict the segmental mobility of AR chains and increase the tortuosity of charge transport pathways, thereby suppressing space charge migration and electrical treeing and improving the local electric field distribution [34]. Moreover, CA@TiO₂/AR composites show a higher electrical breakdown field than TiO₂/AR composites at the same filler content, which is attributed to improved filler dispersion and fewer defect sites, and the formation of an insulating layer on the filler surface (Figure 8c). Accordingly, the 50 CA@TiO₂/AR composite shows the highest E_b of 73.5 kV/mm, which is 1.11 times that of neat AR (66.4 kV/mm).

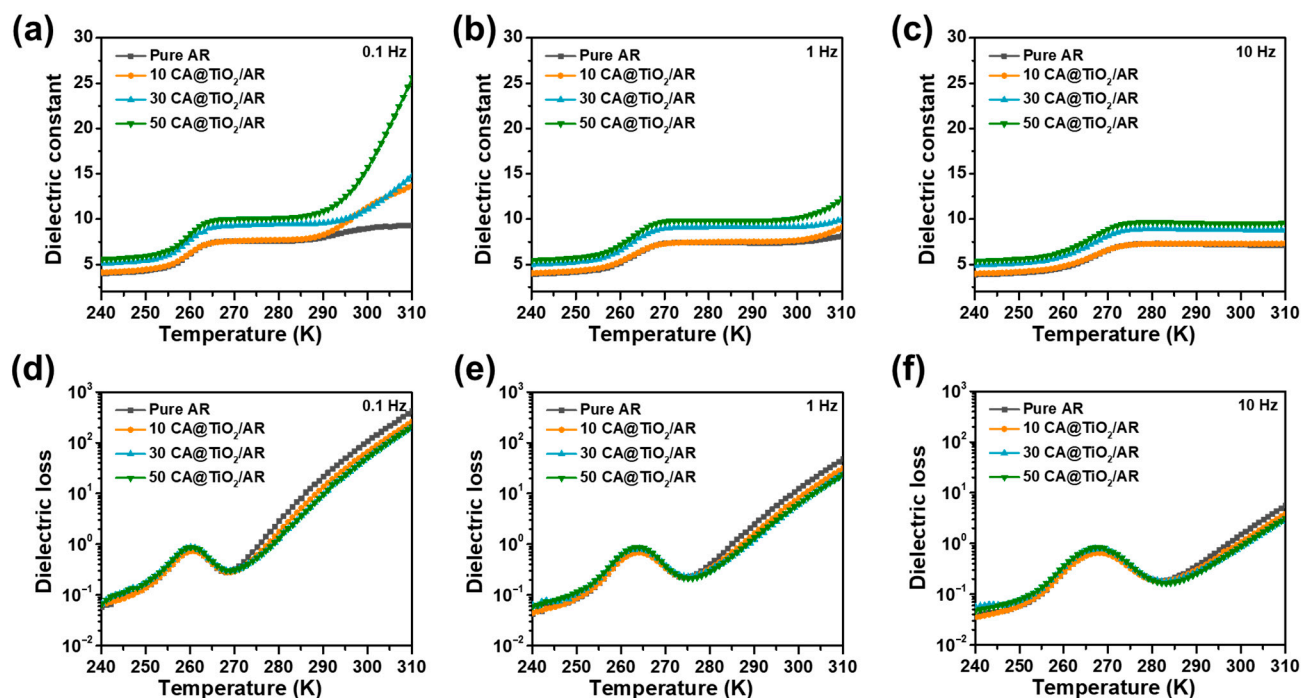


Figure 7. ϵ_r versus temperature of CA@TiO₂/AR composites at (a) 0.1 Hz, (b) 1 Hz, and (c) 10 Hz; ϵ'' versus temperature of CA@TiO₂/AR composites at (d) 0.1 Hz, (e) 1 Hz, and (f) 10 Hz.

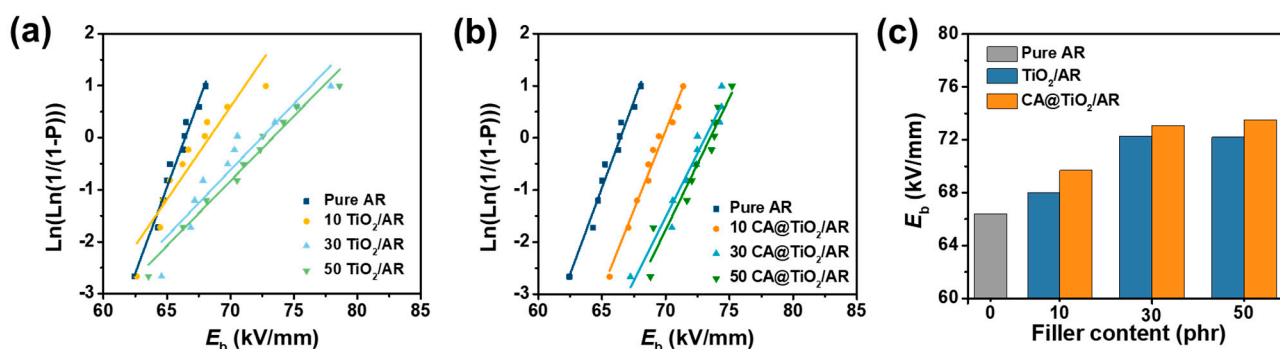


Figure 8. E_b in electric insulating oil calculated from Weibull distribution of (a) TiO₂/AR and (b) CA@TiO₂/AR composites, and (c) Comparison of E_b among all AR composites.

3.5. Actuation Performance of Composite

Actuated strain is central to evaluating actuation performance. We measured it with a circular in-plane actuator without any pre-stretch (Figure 9a). The actuated strain at different electric fields for TiO₂/AR and CA@TiO₂/AR composites is shown in Figure 9b and 9c, respectively, and the corresponding actuation performance is summarized in Table S3. For all samples, the actuated strain increases with electric field and follows a quadratic dependence on the electric field, as shown in Equation (1). At a given electric field (e.g., 20 kV/mm), CA@TiO₂/AR composites exhibit larger actuated strain than TiO₂/AR composites at the same filler content, with the 10 CA@TiO₂ composite showing the largest actuated strain (Figure 9d). These results arise from the differences in electromechanical sensitivity ($\beta = \epsilon_r / Y$), consistent with Equation (1). As shown in Figure 9e, CA@TiO₂/AR composites have higher β than TiO₂/AR composites at the same filler content, because they combine a higher ϵ_r with a lower Y . Meanwhile, β in CA@TiO₂/AR composites first increases and then decreases with filler content. At the 10 phr CA@TiO₂ loading, ϵ_r increases with only a slight change in Y , leading to the highest β . By comparison, at higher filler contents, ϵ_r still increases while Y rises more rapidly, leading to a drop in β . As a result,

owing to its highest β and a relatively high E_b , the 10 CA@TiO₂/AR composite reaches the maximal actuation strain of 7.9% at 31.9 kV/mm, which is 1.48 times that of pure AR and 1.32 times that of the 10 TiO₂/AR composite (Figure 9f).

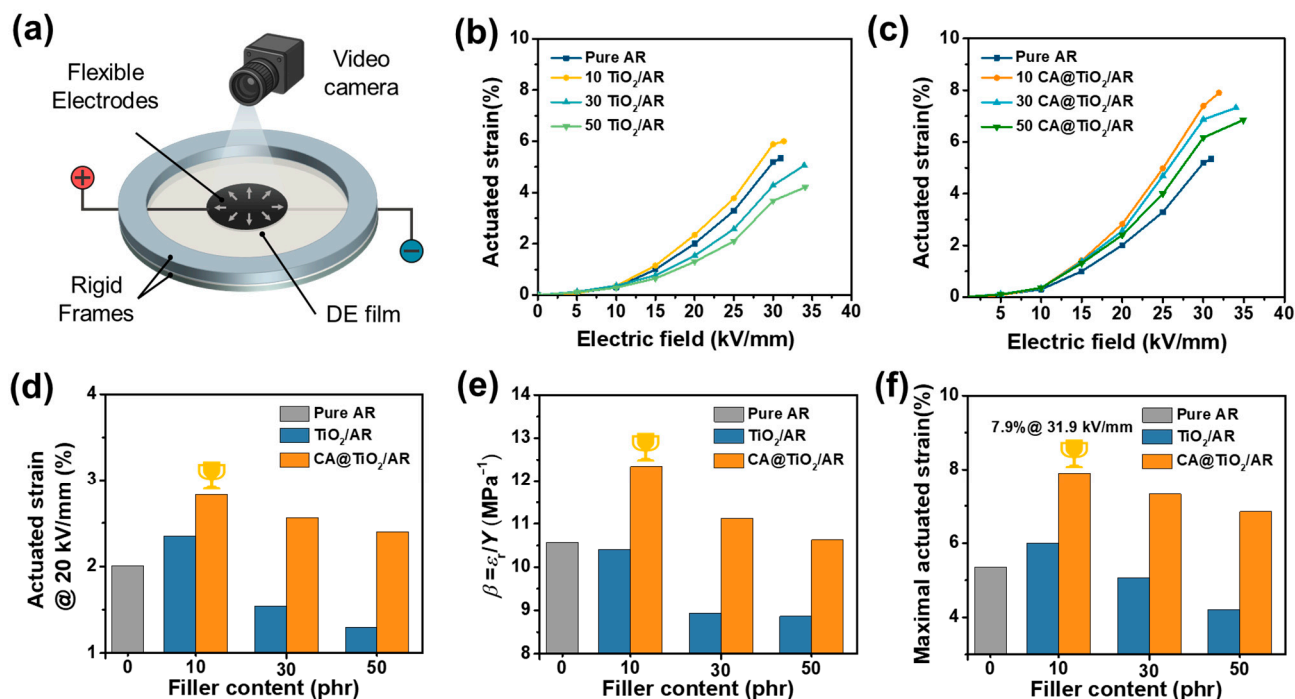


Figure 9. (a) Schematic of actuated strain measurement using a circular in-plane actuator. Actuated strain versus electrical field of (b) TiO₂/AR composites and (c) CA@TiO₂/AR composites. (d) Actuated strain @ 20 kV/mm, (e) electromechanical sensitivity, and (f) maximum actuated strain of all AR composites.

4. Conclusions

In summary, we demonstrated a simple and effective route to enhance the electromechanical performance of AR DEs by incorporating silane-functionalized TiO₂. Compared with pristine TiO₂, CA@TiO₂ forms a siloxane-rich interphase and covalent bridges to the AR network, which improves compatibility, stabilizes dispersion, and thus increases the number of effective polymer-filler phase interfaces and thereby enhances the overall contribution of interfacial polarization. Therefore, at the same filler content, CA@TiO₂/AR composites show lower Y , higher tensile strength, lower hysteresis loss (indicating good elasticity), higher ϵ_r , and higher E_b . Overall, the electromechanical sensitivity ($\beta = \epsilon_r/Y$) of AR composites reaches a maximal value at 10 phr CA@TiO₂ and declines at higher loadings as Y increases more rapidly than ϵ_r . As a result, the 10 CA@TiO₂/AR composite exhibits a maximal actuated strain of 7.9% at 31.9 kV/mm without pre-stretch, which is 1.48 times that of pure AR and 1.32 times that of the 10 TiO₂/AR composite. This low-cost surface modification strategy is broadly applicable for improving the electromechanical performance of DE composites and provides a practical route to high-performance, scalable soft actuators with potential use in artificial bladder applications.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/polym18070872/s1>, Figure S1: Stress-strain curves of porcine bladder in the longitudinal and transverse directions; Table S1: Experimental formulation of AR composites; Table S2: Mechanical and dielectric properties of AR composites; Table S3: Electrical actuation performance of AR composites.

Author Contributions: Conceptualization, L.P. and W.S.; Methodology, L.P. and W.S.; Software, L.P. and W.S.; Validation, L.P. and W.S.; Formal analysis, L.P. and W.S.; Investigation, L.P. and W.S.; Resources, L.P. and W.S.; Data curation, L.P. and W.S.; Writing—original draft, L.P. and W.S.; Writing—review and editing, L.P. and W.S.; Visualization, L.P. and W.S.; Supervision, Y.H., N.N. and J.W.; Project administration, Y.H., N.N. and J.W.; Funding acquisition, Y.H., N.N. and J.W. All authors have read and agreed to the published version of the manuscript.

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