

Enhanced Electro-actuation in Dielectric Elastomers: The Nonlinear Effect of Free Ions

Bin Zheng, Xingkun Man,* David Andelman,* and Masao Doi*

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ABSTRACT: Plasticized poly(vinyl chloride) (PVC) is a jelly-like soft dielectric material that attracted substantial interest recently as a new type of electro-active polymers. Under electric fields of several hundred volt/mm, PVC gels undergo large deformations. These gels can be used as artificial muscles and other soft robotic devices, with striking deformation behavior that is quite different from conventional dielectric elastomers. Here, we present a simple model for the electro-activity of PVC gels and show a nonlinear effect of free ions on its dielectric behaviors. It is found that their particular deformation behavior is due to an electro-wetting effect and to a change in their interfacial tension. In addition, we derive analytical expressions for the surface tension as well as for the apparent dielectric constant of the gel. The theory indicates that the size of the mobile free ions has a crucial role in determining the electro-induced deformation, opening up the way to novel and innovative designs of electro-active gel actuators.

Polyvinyl chloride (PVC) mixed with plasticizers form a jelly-like soft gel dielectrics and have attracted growing interest in recent years as soft actuators.^{1,2} Compared with conventional dielectric elastomers,^{3–10} plasticized PVC gels offer several advantages in terms of their fast response, low working voltage, larger sustained stress, and overall stability and durability.^{1,2,11} Such properties make these materials attractive for novel applications in soft robotics, artificial muscles, medical assistance devices,^{12,13} and in electro-optical devices.^{14,15}

PVC gels are found to have quite different properties than conventional dielectric elastomers.^{16–20} First, their apparent dielectric constant is 10^3 – 10^4 times larger than in conventional materials. Second, upon the application of an electric field, the PVC gel shows amoeba-like spreading on the anode (and not on the cathode), as is shown schematically in Figure 1. This behavior is quite unique and different from the gel contraction observed under similar conditions in most dielectric elastomers.^{5,10} Note that when the polarity of the applied voltage is reversed, the PVC gel deformation will also change its direction. This finding was explained by proposing that the electrostatic adhesive force generated by charges accumulates at the anode.²³ The injected charges from the cathode are important, as there will be no deformation if the cathode is blocked by coating it with an insulating film.¹⁹ Furthermore, a thin plasticizer-rich layer was found experimentally to coat the anode (but not the cathode).¹⁷ It has been conjectured^{19,20,23} that such characteristic behavior is due to the existence of small amounts of free ions in the PVC gel, but to the best of our knowledge, no available model exists that can predict such a material deformation for experimental conditions and eventually to facilitate design of actuators in device applications.

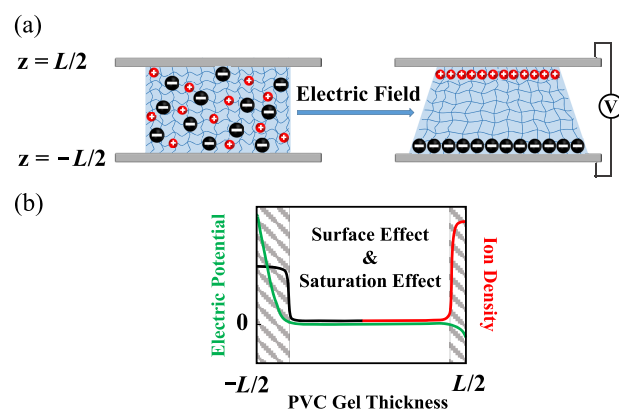


Figure 1. Schematic drawing of the asymmetric deformation of the PVC gel in an electric field and the electrostatic profiles as a function of the interplate distance. (a) The PVC gel is placed between two flat and parallel electrodes located at $z = \pm L/2$. The charge density on the anode and cathode is $q_{\pm} = \pm Q/S_{\pm}$, respectively, where S_{\pm} is the contact area and the total charge on each of the two electrodes is $\pm Q$ (in units of e). Before applying the electric field, the cations and anions inside the PVC gel are homogeneously distributed. (b) Schematic plots of the ion distribution and electrostatic potential inside the PVC gel. The electric field in the bulk of the gel film is nearly zero, and the large electric field exists only near the electrodes where a very dense layer of free ions is created.

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In this Letter, we propose an electro-wetting mechanism to explain the PVC gel electro-activity. The deformation of dielectric elastomers (DEAs) conventionally originates from the Coulomb attraction between the electrodes, that is, the Maxwell stress. However, the latter is a bulk effect, while our interfacial effect has a different origin. The voltage drop we consider is of the order of hundreds of volts,¹⁸ making it much larger than in regular electrolytes (few volts).^{24,25} Only at the electrode proximity, a very concentrated layer of free ions is established, where the ionic saturation effect is considered. We shall show that the ionic size asymmetry creates a large voltage drop at the anode side, yielding a negative contribution to the interfacial tension between the gel and the anode, hence, leading to an instability at the anode with amoeba-like deformation of the gel.

We consider a setup where a PVC gel is confined between two flat electrodes located at $z = \pm L/2$. The gel is an insulator with a dielectric constant ϵ_r . We further assume that the free charges that are embedded in the gel have an asymmetric size, $a_- > a_+$, where a_{\pm} is defined as the cubic root of the ion molecular volume and is related to the close-packing density, $1/a_{\pm}^3$. The two electrodes have a fixed and opposite total charge $\pm Q$ (in units of e), yielding a charge density $q_{\pm} = \pm Q/S_{\pm}$ at the anode and cathode, respectively, where S_{\pm} is the contact area of the gel with the corresponding electrode. As S_{\pm} can vary as the deformation occurs, so will q_{\pm} .

Next we derive separately the electrostatic free energy and elastic energy associated with the confined gel. As the voltages applied in the experiment are high (on the order of 10^3 V), we employ the saturation condition of the free-ion density in order to avoid any nonphysical accumulation of ions near the electrodes (see Figure 1b). The electrostatic free energy was proposed in refs 24 and 25 and is written as

$$\begin{aligned} \frac{F_{\text{elec}}}{k_{\text{B}}T} = & \int (f - f_{\text{bulk}}) d^3r = \int d^3r \left\{ -\frac{1}{8\pi l_{\text{B}}} (\nabla\Psi)^2 \right. \\ & + (c_+ - c_-)\Psi + c_+ \ln(c_+ a_+^3) + c_- \ln(c_- a_-^3) \\ & + a_+^{-3} (1 - c_+ a_+^3) \ln(1 - c_+ a_+^3) \\ & \left. + a_-^{-3} (1 - c_- a_-^3) \ln(1 - c_- a_-^3) - \tilde{\mu}_+ c_+ - \tilde{\mu}_- c_- \right\} \end{aligned} \quad (1)$$

where the first two terms account for the electrostatic energy, the following two terms account for ionic chemical potential, and the last four terms account for the entropy of mixing, where the hard-core nature of the ions is included through the last two terms. $l_{\text{B}} = e^2/(4\pi\epsilon_0\epsilon_r k_{\text{B}}T)$ is the Bjerrum length, $\Psi = e\psi/k_{\text{B}}T$ is the dimensionless electrostatic potential, $c_{\pm}(r)$ is the monovalent cation and anion number density, and $\tilde{\mu}_{\pm} = \mu_{\pm}/k_{\text{B}}T$ is the dimensionless chemical potential. Note that, for simplicity, we ignore the oscillatory behavior of free-ion number density near the electrodes. This effect stems from the repulsive interactions between the hard-core of ions when ionic liquids are confined between charged plates.^{21,22} Thus, the close-packing ion density is taken as a constant $1/a^3$. In our model, the ions interact with the polymer matrix, but the interaction does not appear explicitly in the present theory. The free energy form of eq 1 indicates the interaction between ions and polymer matrix is implicit; namely, the interaction is included in the dielectric constant ϵ and in the incompressible condition of the system (ions and polymer chains). More

details on eq 1 and the reason behind this simplified form of its entropy terms are given in the Supporting Information.

The variation of the free energy F_{elec} with respect to Ψ and c_{\pm} yields a modified Poisson–Boltzmann equation^{24,25} that includes the steric effects:

$$\Psi''(z) = -\frac{\kappa_{\text{D}}^2}{2} \left(\frac{e^{-\Psi(z)}}{1 - \phi_+^b + \phi_+^b e^{-\Psi(z)}} - \frac{e^{\Psi(z)}}{1 - \phi_-^b + \phi_-^b e^{\Psi(z)}} \right) \quad (2)$$

where the ionic volume fraction is $\phi_{\pm}^b \equiv c_{\text{b}} a_{\pm}^3$ and the Debye length is $\lambda_{\text{D}} = 1/\kappa_{\text{D}} = 1/\sqrt{8\pi l_{\text{B}} c_{\text{b}}}$, where c_{b} is the bulk free ion concentration. The boundary condition at $z = \pm L/2$ is $\Psi'(\pm L/2) = \pm 4\pi l_{\text{B}} q_{\pm}$. In accordance with the experimental conditions, we model the high voltage regime by choosing large values of the surface charge density, q_{\pm} . Moreover, the thickness L of the confined gel, which is of the order of millimeters, is much larger than the Debye length (on the order of nanometers), $L \gg \lambda_{\text{D}}$. Hence, the electrostatic potential is vanishingly small in the bulk of the gel, $\Psi(z) \approx 0$, and the free-ion densities there are $c_+(z) \approx c_-(z) \approx c_{\text{b}}$. This means that the two charged boundaries are decoupled, and one can focus on half of the system with a new boundary condition introduced at the symmetric midplane ($z = 0$) with $\Psi(0) = 0$, $\Psi'(0) = 0$, and $c_+(0) = c_-(0) \approx c_{\text{b}}$.

The excess free energy for $-L/2 \leq z \leq 0$ (per unit area) can be written as

$$\begin{aligned} g(q_+) = & \int_{-L/2}^0 dz [f(c_+(z), c_-(z), \Psi(z)) - f(c_{\text{b}}, c_{\text{b}}, 0)] \\ & + q_+ \Psi(-L/2) \end{aligned} \quad (3)$$

where f is the free-energy density in eq 1, and the last term of g accounts for the electrostatic energy of the electrode.

We assume that the surface tension is homogeneous at the electrodes and consider the spreading of the gel at the electrode by analyzing the changes in the contact area at each of the two electrodes. The total surface free energy is obtained by multiplying the contact area at the electrodes with the calculated free energy density. If an electrode has an area S_+ and charge Q , the surface free energy of the anode is given by $S_+[\gamma_0 + g(Q/S_+)]$, where γ_0 is the surface tension when $Q = 0$. The excess surface tension $\Delta\gamma_+$ of the anode is then given by

$$\Delta\gamma_+(q_+) = \frac{\partial[S_+g(Q/S_+)]}{\partial S_+} = g(q_+) - q_+ \frac{\partial g(q_+)}{\partial q_+} \quad (4)$$

Similarly, $g(q_-)$ and $\gamma_-(q_-)$ are the excess free energy and the surface tension between the PVC gel and the cathode, respectively, for the other half film $[0, L/2]$.

Finally, the mechanical deformation of the gel is given by the balance in the full free-energy between the excess free energy and the gel elastic energy,

$$G = S_+g(Q/S_+) + S_-g(-Q/S_-) + F_{\text{el}}(S_+, S_-) \quad (5)$$

where $F_{\text{el}}(S_+, S_-)$ is a phenomenological gel elastic energy given by

$$\frac{F_{\text{el}}}{k_{\text{B}}T} = \frac{k_{\text{b}}}{2} (S_+ - S_0)^2 + \frac{k_{\text{b}}}{2} (S_- - S_0)^2 + \frac{k_{\text{s}}}{2} (S_+ - S_-)^2 \quad (6)$$

where S_0 is the initial (no electric field) contact area of the gel at both electrodes. The two elastic constants appearing above are $k_b = K_b/k_B T$ and $k_s = K_s/k_B T$. In this phenomenological elastic energy, we have integrated the contribution from the profile of the whole (bulk) gel deformation, in addition to the surface contribution, yielding a deformation of a thin gel film under an electric field. This elastic deformation of the gel leads to a change in the contact area. As this gel deformation is very complex, we take instead a phenomenological approach. We use the change in the contact area to characterize the gel elastic deformation by introducing two phenomenological elastic constants, k_b and k_s , that account for the whole bulk deformation. The nonuniform distribution of ions may affect the elasticity of the matrix as the excluded volume of the ions is taken explicitly into account in our theory. However, in the present work, the ionic layer formed by the accumulation of free ions near the electrodes is very thin (about 80 Å), as compared with the thickness of the gel film (about 1 mm). Therefore, we assume that such a thin layer will not affect substantially the elasticity of the soft gel film. Then, the full free-energy can be expressed as a function of S_+ and S_- , and the gel deformation is determined by minimizing G with respect to S_+ and S_- .

Figure 2 shows the excess in surface tension $\Delta\gamma_+ = \gamma_+ - \gamma_0$ on the gel/anode interface as a function of anion size a_- after

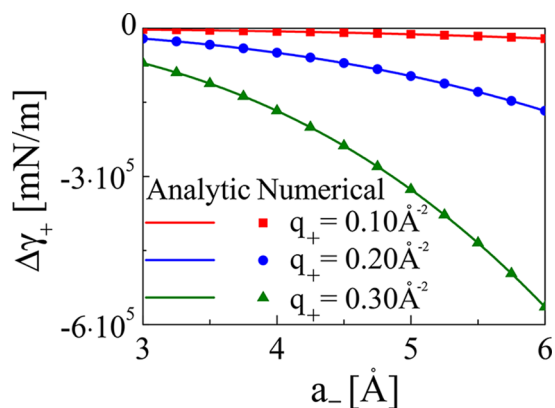


Figure 2. Excess surface tension $\Delta\gamma_+$ at the anode as a function of the ion size a_- . The bulk ion concentration is $c_b = 1$ mM and the cation size is 3 Å. The solid lines represent analytical calculations from eq 10, while the data points are obtained via numerical calculations from eqs 1–4.

numerically solving eqs 1–4. We can see that when an electric field is applied, the accumulation of anions introduces a negative contribution to the surface tension (instability) at the gel/anode interface. Hence, the gel tends to expand at the anode side, which is known as an electro-wetting effect. Moreover, the absolute value of the surface tension $\Delta\gamma_+$ is always an increasing function both of the ion size and charge density.

The derived theoretical framework allows us to deduce an analytical expression of $\Delta\gamma_+$ in the large q limit. Under a large electric field, an electric double-layer is formed near the anode (and similarly near the cathode), and most of the voltage drop occurs in this double layer, which is shown in Figure 1 in the Supporting Information. We then assume that in the localized electric double-layer at the anode, the anions are closely packed, $c_- = 1/a_-^3$, while cations are completely excluded, $c_+ = 0$. Considering charge neutrality, we can estimate the electric

double-layer thickness to be $L_e^+ = q_+ a_-^3$. Then, a simplified explicit expression of the electrostatic potential, $\Psi(z)$, is obtained:

$$\Psi(z) = \frac{2\pi l_B}{a_-^3} \left(z + \frac{L}{2} \right)^2 - 4\pi l_B q_+ \left(z + \frac{L}{2} \right) + 2\pi l_B q_+^2 a_-^3 \quad (7)$$

for $0 < |z + L/2| < L_e^+$ and $\Psi = 0$ for $-L/2 + L_e^+ < z < 0$. In addition, the free-energy F_{elec} can be calculated from eq 1.

$$\frac{F_{\text{elec}}}{k_B T} = -\frac{4\pi}{3} l_B (a_- q_+)^3 - q_+ \tilde{\mu} \quad (8)$$

and the surface free-energy becomes

$$\frac{g(q_+)}{k_B T} = \frac{2\pi}{3} l_B (a_- q_+)^3 \quad (9)$$

Finally, the excess surface tension on the anode side is

$$\frac{\Delta\gamma_+}{k_B T} = -\frac{4\pi}{3} l_B (a_- q_+)^3 \quad (10)$$

The above $\Delta\gamma_+$ expression is shown as solid lines in Figure 2 and is in excellent agreement with the numerical results (data points). A similar behavior close to the gel/cathode interface, $\Delta\gamma_- \sim (a_+ q_-)^3$, is obtained, by simply exchanging negative with the positive charges.

The electro-wetting induces an asymmetric deformation that can be further quantified by two parameters, the difference $\Delta S = S_+ - S_-$ and the mean $\bar{S} = (S_+ + S_-)/2$. As shown in Figure 3a, for anions and cations, of equal size, $\eta = 1$, the gel asymmetric deformation is zero, $\Delta S = 0$, and independent of q_0 , where $q_0 = Q/S_0$ is the initial surface charge density. In addition, Figure 3a shows that the deformation ΔS increases as η increases, indicating that the size ratio, η , plays a crucial role in controlling the gel asymmetric deformation. It is worth noticing that the η -induced gel deformation can be rather large, on the order of 10–20%. Moreover, the gel deformation can be further enhanced for $\eta \neq 1$ by increasing q_0 . Figure 3b indicates that the mean contact area \bar{S} is an increasing function of η .

For large q , an analytical expression of ΔS^* and \bar{S}^* can be obtained. Inserting the surface free energy g , eq 9, into the full free energy, G of eq 5 is written as

$$\frac{G}{k_B T} = \frac{2\pi l_B Q^3}{3} \left(\frac{a_-^3}{S_+^2} + \frac{a_+^3}{S_-^2} \right) + \frac{k_b}{2} (S_+ - S_0)^2 + \frac{k_b}{2} (S_- - S_0)^2 + \frac{k_s}{2} (S_+ - S_-)^2 \quad (11)$$

After rewriting G as a function of δS_{\pm} , where $\delta S_{\pm} = S_{\pm} - S_0$, and expanding G around $\delta S_{\pm} = 0$ to the second order, the gel deformation ΔS^* and \bar{S}^* can be obtained by minimizing G with respect to δS_{\pm} ,

$$\frac{\Delta S^*}{S_0} = \frac{1}{3} \frac{(\eta^3 - 1)}{\eta^3 (\xi q_0^3 + \kappa + 1) + (1 + 2\kappa)/(\xi q_0^3) + \kappa + 1} \quad (12)$$

$$\frac{\bar{S}^* - S_0}{S_0} = \frac{1}{6} \frac{\eta^3 (2\xi q_0^3 + 2\kappa + 1) + 2\kappa + 1}{\eta^3 (\xi q_0^3 + \kappa + 1) + (1 + 2\kappa)/(\xi q_0^3) + \kappa + 1} \quad (13)$$

where $\xi = 4\pi a_+^3 l_B / (S_0 k_b)$ and $\kappa = k_s / k_b$.

The analytic expressions obtained from eqs 12 and 13 are shown in Figure 3 as solid lines and agree well with the

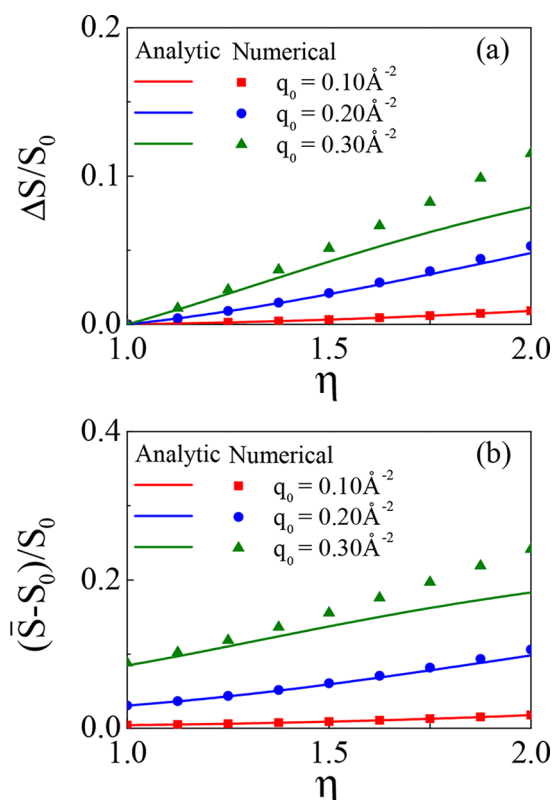


Figure 3. Asymmetric deformation of the PVC gel, $\Delta S/S_0$ and $(\bar{S} - S_0)/S_0$, as a function of the size ratio between anion and cation η . All solid lines are analytical results of eqs 12 and 13, while the dotted data points are taken from the numerical calculations. (a) The dependence of $\Delta S/S_0$ on η for three $q_0 = Q/S_0$ values; (b) The effect of η on $(\bar{S} - S_0)/S_0$ for three q_0 values. The other parameters are the bulk ions concentration $c_b = 1 \text{ mM}$, the elastic constant $k_b = k_s = 15 \text{ \AA}^{-2}$, $a_+ = 3 \text{ \AA}$, $S_0 = 100 \text{ \AA}^2$, $\epsilon_r = 10$, and $l_b = e^2/(4\pi\epsilon_0\epsilon_r k_B T) = 55 \text{ \AA}$, at room temperature, $T = 300 \text{ K}$.

numerical results (dotted points). Note that the scaling law $\Delta S \sim \eta^3$ emphasizes the significant role played by the ionic size asymmetry on the asymmetric gel deformation. Such simple dependence of ΔS^* on η , q_0 , and other experimental parameters found in eq 12 can be verified experimentally.

Our analysis of $\Delta\gamma$ and ΔS indicates that the intriguing gel deformation is caused by accumulation of cations/anions having nonequal sizes. The applied electric field introduces a negative contribution to the surface tension $\Delta\gamma$. In our model, $\Delta\gamma$ can compete with volume contributions like elasticity of the gel film because $\Delta\gamma$ is huge, on the order of 10^5 mN/m , and the PVC gel is soft due to the added plasticizers. Then, the absolute value of $\Delta\gamma$ increases with ion size, while the gel volume remains constant during the deformation. Hence, the gel tends to spread at the anode where the larger size anions accumulate. Finally, the finite amount of asymmetric deformation is determined by the competition between the surface tension and gel elastic energy.

In addition, a very large apparent dielectric constant was observed in the PVC gel's experiments.¹⁸ We note that such large values have been observed in the past for electrolytes²⁶ and serve as evidence for the existence of free ions inside the material. A more detailed consideration of the large values of the dielectric constant is presented in the Supporting Information, where a simple analytic model relates these large dielectric values to the existence of free ions within the

PVC gel. More precisely, its large value is determined by the ratio between the macroscopic film thickness (on the order of millimeters) and microscopic electric double-layer thickness (on the order of nanometers).

In summary, we developed a theoretical model unveiling the fundamental mechanism of the electro-activity of PVC gels. The combination of the electro-wetting effect and the ionic saturation is considered in this model. Furthermore, explicit analytic results related to the material deformation as seen in the experiments are presented. The theory shows that the size ratio between the mobile anions and cations plays a crucial role in determining the electro-induced deformation. An important conclusion is that increasing this ionic size ratio enhances the gel electro-actuation, which has yet to be verified in experiments. Finally, we hope that the physical concepts as explored in our model will provide insight into the design and applications of polymer actuation materials.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsmacrolett.1c00045>.

- (1) The detailed explanation of the free energy.
- (2) The ion distribution and electrostatic potential inside the PVC gel.
- (3) A simple analytic model describing the large observed values of the apparent dielectric constant (PDF)

■ AUTHOR INFORMATION

Corresponding Authors

Xingkun Man – Center of Soft Matter Physics and its Applications, and School of Physics, Beihang University, Beijing 100191, China; orcid.org/0000-0003-4266-1539; Email: manxk@buaa.edu.cn

David Andelman – Raymond and Beverly Sackler School of Physics and Astronomy, Tel Aviv University, 69978 Tel Aviv, Israel; orcid.org/0000-0003-3185-8475; Email: andelman@post.tau.ac.il

Masao Doi – Center of Soft Matter Physics and its Applications, and School of Physics, Beihang University, Beijing 100191, China; Email: masao.doi@buaa.edu.cn

Author

Bin Zheng – Raymond and Beverly Sackler School of Physics and Astronomy, Tel Aviv University, 69978 Tel Aviv, Israel

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsmacrolett.1c00045>

Notes

The authors declare no competing financial interest.

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