Macromolecules **2001,** *34,* 7179−7189

Electromechanical Response of Electrorheological Fluids and Poly(dimethylsiloxane) Networks

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Received May 10, 2001; Revised Manuscript Received July 12, 2001

ABSTRACT: A novel type of electromechanical actuator that utilizes the speed of electrorheological fluids (ERFs) coupled with the elastic response of polymeric gels has been investigated. ERF's were made using polyaniline (PANI) particles suspended in a trimethyl-terminated poly(dimethylsiloxane) (3MPDMS) fluid. These ERF's were then cured into matrices of both ordered and randomly cross-linked PDMS networks (XPDMS). Electric fields were applied to these systems using flexible electrodes. Both electrode displacement and compression modulus of the composite gels were measured as a function of cross-link density of the gels, viscosity, and electrical properties of the 3MPDMS fluid, conductivity and dielectric constant of both the pure PANI and the gel matrix, weight percent PANI in the ERF, volume percent ERF in the final gel composite, and applied electric field strength. The dc electric field required to move the electrodes toward each other by a fixed distance depended more on the type and amount of cross-linking in the XPDMS matrix, the viscosity of the ERF phase, and whether the ERF particles were prealigned in the XPDMS network than it did on the compression modulus of the systems. In addition, the results indicated that the motion generated by this actuator also depends on the amount of force exerted on the electrodes by the polarized ERF particles in the gels.

Introduction

The exchange of electrical energy for mechanical energy has long been of both theoretical and practical interest. Electromechanical energy conversion has made many modern conveniences possible, such as automobiles, refrigerators, and, more recently, robotics. Perhaps the ultimate electromechanical actuator is mammalian striated skeletal muscle, which has a response time of 15−120 ms, requires only a 0.1V potential change, and yields a 30% change in sarcomere length.[1] Nature has thus made a very efficient and fast electromechanical actuator, which we have yet to duplicate.

Electrorheological fluids (ERFs), which are generally composed of 10−100 μm diameter nondeformable dielectric particles, such as glass beads, starch, or poly(ethylene oxide), suspended in an insulating fluid, can convert electrical input into a mechanical change very quickly, within a few milliseconds after the application of an electric field. These ERFs change from viscous fluids to viscoelastic solids when a sufficiently large electric field is applied; this is often referred to as the ER effect.[2] Upon application of the electric field, the ERF particles become polarized and align in the field direction, forming so-called pearl chains and fibrils that bridge the gap between the electrodes; these aligned particles stiffen the ERF and cause the observed change in the rheological behavior of the material. The nature of the polarization of the particles is rather complicated and has been addressed in several reviews.[2−5] When the electric field is removed, the particles tend to remain aligned unless a force, such as a shear force, is applied to the system.

We have previously[6] shown that we could make an electromechanical actuator that could apply a force repeatedly by using a commercially available ERF. We could obtain a repeatable electrode displacement of 100 μm when this ERF was placed in a poly(dimethylsiloxane) gel (XPDMS) between two flexible electrodes, 5 mm apart, and a 6.2 kV/cm dc electric field was applied. It was possible to attach 1 g flags to the electrodes and to wave these flags visibly at frequencies up to 15 Hz. In the present work, we have synthesized our own ERFs in the form of polyaniline (PANI) particles that were then combined with various trimethyl-terminated poly(dimethylsiloxane) (3MPDMS) fluids. The viscosities of the 3MPDMS fluids were different and the conductivities and dielectric constants of the PANI particles could be controlled and varied. In addition, both “model” ordered networks and a randomly cross-linked network were investigated to determine what effect the matrix order had on the electromechanical response. The balance between the mechanical strength of the XPDMS gel, which increases with increasing cross-link density, and the compressibility of the gel (related to its ability to change its dimensions), which decreases with increasing cross-link density, was also investigated. For the purpose of making an electromechanical actuator, both the electrode displacement and the force generated after application of an electric field were of interest. Instead of measuring the force generated under various conditions, it was simpler and seemed more useful to measure the compression modulus of the various XPDMS/ERF composite gels, both in the absence and in the presence of an electric field. The compression modulus, Y_c, is obtained using a very small uniaxial compression of a slab of material constrained between two plates; for our purposes, an electric field could also be applied between the two plates used for compressing the sample.

$$Y_c = \frac{F/A}{\Delta L/L_0} \qquad (1)$$

where F is the uniaxial force applied to a sample over its original area A, L_0 is the original width of the sample

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10.1021/ma0108158 CCC: $20.00

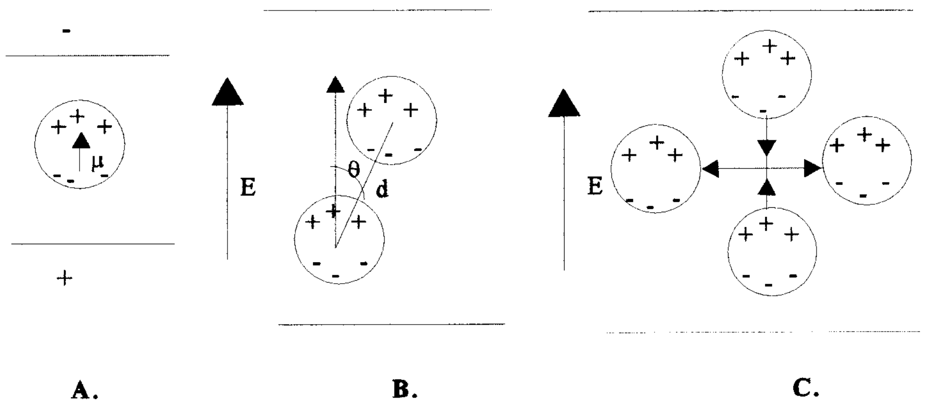

Figure 1. Polarization and interactions between particles when $\epsilon_p > \epsilon_f$: (A) induced "normal" dipole, μ, in a single spherical particle; (B) an angle θ between the line joining the centers, c, and the field direction between two particles, d; (C) an attractive interaction between two particles at $\theta = 0$ and a repulsive interaction between two particles at $\theta = \pi/2$.

between the plates, and ΔL is the change in sample width after the force has been applied.

The ER Effect

Because we are using electrorheological fluids, a simplified description of the ER effect is included here. It is generally accepted that the electric field polarizes the particles and that the interaction of these induced dipoles provides the forces leading to pearl chain formation followed by fibrillation and the change in rheology. For simplicity, the point dipole approximation is often used to discuss the ER effect even though particles in pearl chains or fibrils should really be treated as finite particles. In addition, even though it has been known for some time that conductivity (the leaky dielectric model) is important in the explanation of the ER effect in dc or low-frequency ac electric fields, the older dielectric model is simpler to understand and can be invoked to explain the ER effect in high-frequency ac fields. In the dielectric model, it is assumed that an ERF is composed of spherical particles of dielectric constant ϵ_p, suspended in a fluid of dielectric constant ϵ_f. As shown in Figure 1, when exposed to an external electric field, a mismatch in the permittivity between a particle and the medium causes polarization of the particle leading to charge separation on the particle surfaces.[3] The particle then acquires an induced dipole moment, μ:

$$\mu = 4\pi\epsilon_0\epsilon_f\beta a^3 E \qquad (2)$$

where ϵ_0 is the permittivity of free space, a is the radius of each (spherical) particle, E is the applied electric field, and β is the effective polarizability of each particle:

$$\beta = \frac{\epsilon_p - \epsilon_f}{\epsilon_p + 2\epsilon_f} \qquad (3)$$

As can be seen in eq 3, if there is no dielectric mismatch, $\epsilon_p = \epsilon_f$, and the particle and medium polarize to the same extent, there is no effective dipole moment of the particles.[3] Since purely dielectric ERFs must have $\epsilon_p > \epsilon_f$ (see below), $\beta > 0$; this indicates that the induced positive charge on the particle surface is facing the negative electrode *and is attracted by that electrode* as shown in Figure 1A. This polarization is called a "normal dipole". In the point–dipole limit, when the spheres are far apart, the force on a sphere of radius a by an identical sphere arranged as in Figure 1B is[7]

$$F(d,\theta) = \frac{3}{16}\pi\epsilon_0\epsilon_p a^2\beta^2\left(\frac{a}{d}\right)^4[(3\cos^2\theta - 1)\epsilon_d + \sin(2\theta)\epsilon_\theta]E^2 \qquad (4)$$

where $F(d,\theta)$ is the electrostatic force between the two spheres, d is the center-to-center separation between them (Figure 1B), θ is the angle between the line of centers and the applied field, E, and e_d and e_θ are unit vectors along and perpendicular to the direction specified by the line between the centers of the two spheres, respectively. Equation 4 clearly shows that the force between the spheres is positive and along the d direction (the spheres attract each other) when $\theta = 0$, and negative along the d direction (the spheres repel each other) when $\theta = \pi/2$. Thus, the force is attractive between the poles and repulsive between the equators of the spherical particles and the interaction energy between the two particles is minimized when the particles are aligned parallel and maximized when the two particles are aligned perpendicular to the applied field (Figure 1C). As a result, the particles tend to align in the direction of the electric field, forming pearl chains. The chain structure is what gives rise to the yield stress and increase in viscosity that makes up the ER effect.

Pearl chains can also form when $\epsilon_p < \epsilon_f$, in which case the effective polarizability $\beta < 0$. In this case the induced positive charge on the particle surface is facing the positive electrode; this is called a "reverse dipole". These pearl chains will be repelled by the electrodes, so they cannot span the whole space between the electrodes and there can be no large ER effect.

This discussion so far has been limited to the interaction of two spherical particles in a uniform electric field in the point dipole approximation. ERFs are actually concentrated suspensions, not necessarily of spherical particles, and in this situation, the particles do not act like point dipoles and are subjected to local fields arising from the dipole field generated by neighboring particles as well as the applied field. Furthermore, the conductivity of both the fluid and the suspended particles must be considered in low-frequency ac and in dc electric fields (the leaky dielectric theory). In this case, the polarizability of a suspended particle in an electric field depends on the conductivity mismatch between the particles and the fluid as well as the dielectric mismatch. Khusid and Acrivos[8] have determined that two

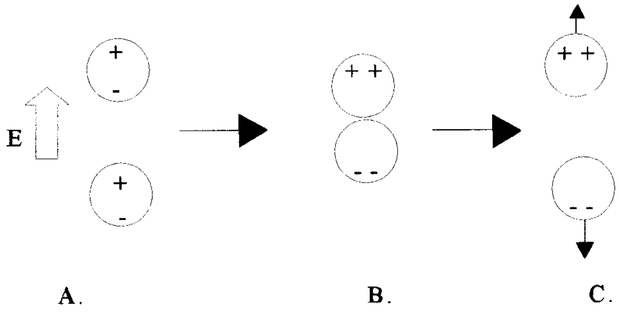

Figure 2. Possible interaction between two conductive particles in a dc electric field: (A) two particles with "normal" induced dipoles at $\theta = 0$; (B) charge reorganization after the particles touch; (C) expected particles repulsion after charge reorganization.

such particles attract each other so that the ER effect can occur only under one of the following two sets of conditions:

$$\frac{\sigma_p}{\sigma_f} > 1 \quad \text{and} \quad 6\frac{\epsilon_p}{\epsilon_f} > 2 + 5\frac{\sigma_p}{\sigma_f} - \left(\frac{\sigma_p}{\sigma_f}\right)^2 \tag{5a}$$

$$\frac{\sigma_p}{\sigma_f} < 1 \quad \text{and} \quad 6\frac{\epsilon_p}{\epsilon_f} < 2 + 5\frac{\sigma_p}{\sigma_f} - \left(\frac{\sigma_p}{\sigma_f}\right)^2 \tag{5b}$$

when the time after application of the dc field is much greater than the characteristic time of dielectric relaxation, t_p

$$t_p = \frac{\epsilon_0(\epsilon_p + 2\epsilon_f)}{\sigma_p + 2\sigma_f} \tag{6}$$

and where σ_p and σ_f are the conductivities of the particle and fluid, respectively, and ϵ_0 is the permittivity of vacuum. Therefore, if the time is sufficient, there is a redistribution of charge at the interface between the particle and fluid which leads to these conditions (parts a and b of eq 5) for the ER effect. When the time, $t \ll t_p$, the polarization follows eq 3.

The change in particle polarization is not the only complication that arises from finite values of particle and fluid conductivity. As shown in Figure 2A, a pair of conductive particles in an electric field that are attracted to one another by their induced dipole moments may make electrical contact when they touch, and a redistribution of charge results (Figure 2B). These redistributed charges should then give rise to a force in the applied field which pulls the particles apart and toward the electrodes (Figure 2C). The possibility of such a disruptive force in a dc electric field has been discussed by Jones,[5] but nevertheless, weakly conducting ERF's do form pearl chains and fibrils in dc electric fields. A film of fluid probably remains between the particles, and there is current flowing along the particle chains.[9] The small gap between particles, and the charge transfer which occurs between particle and fluid at the high electric fields used, make the conductivity of the fluid phase extremely important. This is although the dielectric fluids used in ERF's have extremely low conductivities, on the order of 10^{-12} S/cm. A variety of physical phenomena can give rise to these low but finite conductivities in a dielectric liquid,[10] such as an ionization of the fluid, electronic carrier injection, or, in the case of ERF's, ionic impurities from the ERF particles.

Experimental Section

Materials. The poly(dimethylsiloxane) (PDMS) precursors of the cross-linked samples used in this work are described in Table 1; one of these was a partly fluorinated copolymer. The PDMS fluids that were used to swell the cross-linked samples and to prepare various ERF's are described in Table 2. Tetraethyl orthosilicate (TEOS) was obtained from Sigma Chemical Co., St. Louis, MO, Scientific Polymer Products, Inc., Ontario, NY, and Gelest, Inc., Tullytown, PA. 95% Bis(2-ethylhexanoate)tin (2EHSn) was obtained from Lancaster, Inc., Windham, NH, and Gelest, Inc. Platinum−divinyltetramethyldisiloxane complex, 2.1−2.4% Pt concentration (PtDVT), was obtained from Gelest, Inc. A commercial sample of a two part cure PDMS, RTV 6136 (parts A and B) was obtained from the General Electric Silicone Products Division, Waterford, NY. Other chemicals were ACS grade aniline, ammonium persulfate, and concentrated HCl from Fisher Scientific, Fair Lawn, NJ, and HPLC grade benzene from Aldrich Chemical Co., Milwaukee, WI.

Synthesis and Doping of Polyaniline Particles and Preparation of ERF's. Polyaniline (PANI) was synthesized using the oxidative polymerization method of MacDiarmid et al.,[11] with a slight variation. Two grams of aniline was added slowly to 250 mL of 1 M HCl, cooled to 0 °C, in a three-neck flask with constant stirring. Ten milliliters of a solution of ammonium persulfate in 1 M HCl was prechilled to 0 °C and then added to the mixture slowly, over a 20 min period. The reaction was allowed to proceed for 1 h before another 10 mL of the ammonium persulfate solution was slowly added, over 20 min. The reaction was then allowed to continue for 2 h. The reaction temperature was kept at −5 °C for the duration of the reaction using a 2-propanol−ice bath. The resulting PANI crystals were collected by vacuum filtration. Different doping levels of the PANI were obtained by a proton exchange reaction in different pH solutions as previously shown by other workers.[11,12] The PANI was finely ground using a mortar and pestle and placed in 200 mL of water, and the pH of the solution was measured using a Corning 430 pH meter, Corning Inc., Corning, NY. The pH levels were monitored and adjusted by adding dilute NaOH to obtain pH's in the range 2−9.5. Doping at any pH was considered complete when the pH remained constant for 24 h. The time needed for completion of doping depended on the sample size of the PANI, ranging from 4 to 8 weeks. After doping, the PANI particles were stored in a vacuum oven with desiccant at 45 °C, 20 mmHg, for at least 24 h before grinding with a mortar and pestle and mixing with 3M-PDMS to make ERF's containing 5, 7.5, 10, and 20 wt % PANI. Most of the ERF's contained 10 wt % PANI. In some cases, HOF−PDMS was mixed with the PANI particles.

Synthesis of Cross-Linked Poly(dimethylsiloxane)s (XPDMS). Condensation Cure (C-XPDMS). The method of Mark and Sullivan[13] was modified only slightly. HO-PDMS was combined with TEOS in a 4:1 mole ratio, using 0.3−0.5 wt % of 2EHSn as catalyst. The gel was then placed under dynamic vacuum and allowed to cure for 24−96 h depending on the molecular weight of the HO-PDMS used. This C-XPDMS synthesis provided a gel with an ordered cross-linked system. C-XPDMS/ERF gel composites were prepared as above with the addition of an ERF and were then cured under a 20 mmHg dynamic vacuum. Cure times ranged from 1 to 5 days, depending on the C-XPDMS/ERF ratio used; the higher the ratio, the less time the system took to cure. The PANI in the ERF had to be extremely dry, that is, stored under vacuum, in order for the system to cure. Also, the particle content had to be less than 5 wt % in the gel or the mixture thickened and never formed a fully cross-linked gel system. Therefore, only 6/4 and 5/5 w/w C-XPDMS/ERF systems were investigated further.

Vinyl Addition Cure (V-XPDMS and F-XPDMS). The method of Quan[14] was used, but with a different platinum catalyst. V-PDMS was combined with HMS-013 in a 5:1 weight ratio, using PtDVT as the catalyst. This type of cross-linking provided a randomly cross-linked gel. F-PDMS was combined with HMS-013 in a 6:4 weight ratio, also using PtDVT as the

Table 1. Characteristics of PDMS Precursors of the Cross-Linked Samples, Where All Number Average Molecular Weights, M_n, Were Determined by the Suppliers

terminating group or comonomer in precursor	abbreviation	$M_n \times 10^{-4}$
hydride terminated	H-PDMS	2.8[a]
silanol terminated	HO-PDMS	2.6[a]
		3.6[a,b]
		4.35[a]
		9.0[b]
vinyl terminated	V-PDMS	2.8[a]
0.5−1.0 mol % methylhydrosiloxane comonomer	HMS-013	5.5[a]
vinyl terminated; 35−45 mol % trifluoropropylmethylsiloxane comonomer	F-PDMS	2.5−3.5[a]

[a] Gelest, Inc., Tulleytown, PA. [b] Scientific Polymer Products, Inc., Ontario, NY.

Table 2. Viscosities and Molecular Weights of PDMS Fluids Used to Swell the Samples and to Prepare ERF's, Where All Number Average Molecular Weights, M_n, Were Determined by the Supplier, Gelest, Inc., Tullytown, PA

fluid	abbreviation	viscosity (cst)	M_n
trimethyl-terminated PDMS	3M-PDMS	2	410
		20	2000
		200	9430
		500	17 250
		5000	49 350
silanol-terminated trifluoropropylmethyl siloxane	HOF-PDMS	40	550−800

catalyst. To make vinyl addition cure gel composites, the V-XPDMS and F-XPDMS gel components were mixed with an ERF and the mixtures were allowed to cure at atmospheric pressure. The weight ratios of the XPDMS mixture to ERF were 1/9, 3/7, 4/6, 5/5, 6/4, 7/3, and 9/1. Samples were also prepared containing 5 wt % PANI in V-XPDMS gel in the absence of a fluid phase.

Flexible Electrodes. Gold was sputtered onto "Xerox Gold Series" transparencies using a Dentoncoat II Desktop Sputterer, Denton Vacuum Inc., Moorestown, NJ, with a gold source, at 50 mTorr, 40 mA, for 60 s. The electrodes were 100 μm thick, measured using a micrometer.

Curing of XPDMS/ERF Gels between Flexible Electrodes. Two flexible electrodes, 5 cm long *x* 5 mm deep, were arranged 5 mm apart, surrounded by a cured RTV matrix. The XPDMS/ERF system to be tested was prepared and then poured between the flexible electrodes and allowed to cure. In some cases, the cure proceeded under an 8 kV/cm electric field.

XPDMS/ERF Gels for Compression Modulus Testing. The XPDMS/ERF system to be tested was poured into a circular mold, diameter 2.8 cm and depth 3 mm. The gel was cured and the mold was then removed. In some cases, the XPDMS/ERF system was cured under an 8 kV/cm electric field.

Physical Measurements. Determination of M_c. The average molecular weight between cross-links of the XPDMS gels, M_c, was determined from swelling in HPLC grade benzene[15] at room temperature. The version of the Flory−Rehner equation that gave the best values for C-XPDMS samples to Hamurcu and Baysal[15] was used in this work for all samples.

$$M_c = \frac{-\rho_2 V_1\left(v_{2m}^{1/3} - \frac{1}{2}v_{2m}\right)}{\ln(1 - v_{2m}) + v_{2m} + \chi v_{2m}^{2}} \tag{7}$$

where

$$V_s = \frac{w_0}{\rho_2} + \frac{(w_s - w_0)}{\rho_1} \text{ and } v_{2m} = \frac{w_0}{V_s \rho_2}$$

and where V_1 is the molar volume of benzene at room temperature, 89 cm^3 mol^{-1}, ρ_1 is its density, 0.87 g cm^{-3}, ρ_2 is the density of PDMS, 0.98 g cm^{-3}, χ is the Flory interaction parameter between PDMS and benzene, and w_0 and w_s are the masses of the dry and swollen sample, respectively. The concentration dependent interaction parameter between PDMS and benzene was used in the same way as used by Hamarcu and Baysal.[15]

Conductivity and Dielectric Constants: PANI. PANI samples, 0.07 g each, were compressed into 1.2 cm diameter cylindrical pellets, 0.5 mm thick, under a force of 12 tons. The top and bottom surfaces of the pellets were then sputter coated with gold. The capacitance and resistance of each sample were measured on a Hewlett-Packard Co., Cupertino, CA, 4192A impedance analyzer, from 1 kHz to 1 MHz, with an applied voltage of 0.3 V. The dielectric constant, ϵ_p, was calculated from the capacitance and the cell geometry.

Gels and ERF's. The dielectric constants of the gels and the ERF's were calculated from capacitance readings measured on a Solartron 1260 Impedance/Gain Phase analyzer, Solartron, Allentown, PA, at room temperature, 10 kHz, and 1 V. Dc conductivities were determined using a Keithley Instruments, Inc., Solon, OH, Electrometer, model 6517.

Compression Modulus. The compression moduli of the XPDMS/ERF gels were calculated from force measurements taken on an Instron Corp., Canton, MA, 4204 Universal Testing Instrument, with computer software control. A displacement rate of 0.5 mm/min was used with the applied force approximately 0.25 kN. The sample was placed between two copper plates that could be attached to a Trek 610C dc power supply/amplifier, Trek, Inc., Medina, NY, via alligator clips; ceramic insulators were used between the copper plates and the Instron. Dc electric fields from 0 to 22 kV/cm could be applied to the samples during compression measurements.

Electrode Displacement Measurements: The Trek 610C high voltage power supply/amplifier was used to provide both dc and ac voltages. Ac voltage signals from a Stanford Research Systems, Sunnyvale, CA, model DS335, 3.1 MHz synthesized function generator were amplified using the Trek 610C. The gel/electrode systems were examined on an optical microscope stage using a Leitz Laborlux 12 Pol microscope, Leica, St. Gall, Switzerland. A Hamamatsu CCD camera (Hamamatsu Corp., Bridgewater, NJ) was attached to the microscope and connected to a Sony VCR and a Sony UP 3000 color video printer interfaced with a Pentium PC. Single electrode displacements were analyzed using Optimas Image Analysis Software (Optimas Corp., Bothell, WA). With this apparatus, it was possible to observe the fast displacement of our flexible electrodes, on the order of 100 ms, but it was not possible to obtain the exact speed of the displacement.

Results and Discussion

M_c of the XDPMS Gels. The number-average molecular weight of the chains between cross-links, M_c, of the RTV, V-XPDMS, and C-XPDMS gels is shown in Table 3. As expected, the M_c's of the V-XPDMS gels depend on the proportion of initiator used, while the C-XPDMS gels have M_c values within ∼10% of the M_n values of the HO-PDMS precursors. It should be noted that M_c data could only be obtained for the pure RTV and XPDMS gels. It was therefore necessary to assume that the M_c of each gel was unchanged when it was

Table 3. Molecular Weight between Cross-Links in the XPDMS Gels Prepared Using Various Weight Ratios of PDMS Precursor to Initiator

XPDMS	PDMS:initiator weight ratio	M_n of PDMS precursor ($\times 10^{-4}$)	M_c ($\times 10^{-4}$)
RTV	1:1[a]		1.6
V-XPDMS	5:1	2.8	5.6
	3:2	2.8	0.47
	4:3	2.8	0.39
C-XPDMS	4:1	3.6	3.2
		4.35	4.0
		9.0	8.7

[a] Weight ratio A:B.

Table 4. (a) Dielectric Constants, ϵ, and (b) Conductivitites, σ, of the PANI Samples at Different Frequencies[a]

(a) Dielectric Constants

	ϵ			
sample	1 kHz	10 kHz	100 kHz	1 MHz
P2	2.00×10^5	1.56×10^5	1.04×10^5	4.27×10^3
P5	1.66×10^5	1.20×10^5	2.20×10^4	627
P6	7.28×10^4	5.91×10^3	2.98×10^3	1.35×10^3
P7	70	32.7	16.6	11.0
P7.5	15.9	10.5	8.07	7.02
P8	7.66	6.29	5.64	5.27
P9	7.36	6.39	5.98	5.81
P9.5	8.32	6.92	6.25	6.07
X7.5	145	15.0	5.58	2.6
X8		4.01	3.37	3.28
X9	16.9	10.9	8.51	7.67

(b) Conductivities

	σ (S/cm)				-log σ 100 kHz
sample	1 kHz	10 kHz	100 kHz	1 MHz	
P2	4.04×10^{-4}	1.98×10^{-3}	1.34×10^{-2}	1.32×10^{-2}	1.87
P5	1.96×10^{-4}	1.56×10^{-3}	3.03×10^{-3}	3.72×10^{-3}	2.52
P6	8.12×10^{-5}	1.27×10^{-4}	1.45×10^{-4}	1.79×10^{-4}	3.84
P7	1.85×10^{-7}	3.84×10^{-7}	2.08×10^{-6}	2.27×10^{-5}	5.68
P7.5		2.16×10^{-7}	2.95×10^{-6}	4.27×10^{-5}	5.53
P8		4.68×10^{-7}	5.27×10^{-6}	7.95×10^{-5}	5.28
P9		2.92×10^{-6}	8.77×10^{-6}	9.53×10^{-5}	5.06
P9.5		5.89×10^{-7}	7.27×10^{-6}	1.06×10^{-4}	5.14
X7.5	5.21×10^{-6}	5.94×10^{-6}	7.44×10^{-6}	1.36×10^{-5}	5.13
X8		7.74×10^{-10}	1.50×10^{-8}	8.10×10^{-8}	7.82
X9	9.46×10^{-9}	2.21×10^{-8}	6.92×10^{-8}	2.90×10^{-7}	7.16

[a]Designations P and X denote different samples equilibrated at different times. The number after the P or X is the approximate pH at which the sample was equilibrated.

prepared in the presence of 3MPDMS fluid, PANI particles, or an ERF.

Electrical Properties. The 10 kHz dielectric constant of the F-XPDMS gel and the HOF-PDMS fluid was 7.2; all nonfluorinated XPDMS gels and 3MPDMS fluids had dielectric constants equal to 2.4. The 10 kHz conductivity of the F-XPDMS gels was 1.0×10^{-8} S/cm; all nonfluorinated XPDMS gels and the 20 cst 3MPDMS fluid had a conductivity equal to 1.0×10^{-12} S/cm. It was assumed that the other 3MPDMS fluids had the same conductivity as the 20 cst fluid. The dielectric constants and conductivities of the PANI samples are shown in parts a and b of Table 4. Conductivities generally increased with increasing frequencies as noted by many others in many solids and liquids. The conductivity increase was, however, not proportional to a fractional power of the frequency, though such a proportionality has been found at frequencies up to 100 kHz in crystalline silicon,[16] pressed powdered poly(acenequinone),[17] and up to 1 MHz in films of poly(*N*-methylpyrrole)[18] (only at temperatures $\leq$ 112 K). Dielectric constants, as expected, decreased with increasing frequency, some showing a major dispersion in the frequency range investigated. This is probably due to interfacial, or Maxwell−Wagner polarization. We may note that PANI samples equilibrated at similar pH's had quite different dielectric constants and conductivities, possibly due to varying water content; the samples were dried but not kept away from the atmosphere. Various workers[5,9,19,20] have obtained conductivities and dielectric constants of similar samples; some of the conductivities have orders of magnitude similar to those shown in Table 4, but dielectric constants are quite different. Again, different water contents in the different samples are probably responsible for the differences. In this work, we use our own measured values of conductivity and dielectric constant, either at 10 or at 100 kHz. Our conclusions do not depend on the frequency at which the quantities were measured.

Compression Modulus. The compression moduli of all the gel systems that were cured in the absence of an electric field are shown in Table 5. In the one case in which the percent PANI in the ERF was varied from 0 to 20 wt % (PANI P8 with 20 cst 3MPDMS), the compression modulus, in the absence of an electric field, increased by a factor of 2 when the wt % PANI was increased from 0 to 20. This was expected because the PANI particles should act as filler particles. Table 5 shows an even larger increase in compression modulus, by a factor greater than 10, when PANI particles were added to the F-XPDMS system.

Table 5 also shows that the application of an electric field had no effect on the compression modulus of any of the gels that contained no ERF, except for the one containing F-XPDMS. The increase in compression modulus of the F-XPDMS-containing gel in the electric field cannot be explained, as far as we know, by the differences in electrical properties between F-XPDMS and the other XPDMS gels, that is, by the much higher dc conductivity, 10^{-8} vs 10^{-12} S/cm, and dielectric constant, 7.2 vs 2.4 at 10 kHz, of the F-XPDMS-containing gel. However, the F-XPDMS is a copolymer, and we do not know the r_1/r_2 ratio for monomer addition. The copolymer may be random or blocky, and in either case, the electric field may cause aggregation of the repeat groups with the higher dielectric constant, thus forming additional physical cross-links that strengthen the gel and increase its modulus.

It is not surprising that all the gels that contained an ERF had an increased compression modulus in a 3.3 kV/cm field; it is interesting to note, however, that only two of the four gels examined in a 6.3 kV/cm field showed a further increase in the compression modulus. This indicates a possible saturation effect. Shiga et al.,[21] who studied the compression and shear moduli of silicone gel filled with poly(methacrylic acid cobalt(II) salt) particles at 46 wt % filler content and 13 wt % absorbed water in electric fields up to 50 kV/cm, found that both moduli increased with the square of the applied field intensity, with no saturation effect. A theoretical prediction for electrorheological fluids by Whittle[22] predicts that the shear modulus should increase with the square of the applied field. We have found no such result in the present work.

Table 5 also shows that the compression modulus of the 50/50 F-XPDMS/ERF gel increased by only a factor of 1.1 in a 3.3 kV/cm dc electric field, much less than that of the equivalent gel that contained no ERF.

Table 5. Compression Moduli, in the Presence and Absence of a Dc Electric Field, of the Gel Systems Prepared in the Absence of an Electric Field

gel system		PDMS viscosity	PANI used in ERF		compression modulus (kPa) for applied electric field (kV/cm) of		
ingredients	wt/wt	(cSt)	type	wt %	0	3.3	6.3
RTV/3MPDMS	60/40	200	none		2.4	2.4	
RTV/Bayer ERF	60/40				3.7	5.1	
V-XPDMS/3MPDMS	50/50	20	none		1.4	1.4	
		200	none		2.1	2.1	
V-XPDMS/ERF	50/50	20	P6	10	2.2	2.9	
		20	P7	10	2.0	4.0	3.8
		200	P7	10	1.8	3.6	
		20	P8	5	1.9	2.8	
				10	2.1	3.0	3.4
				20	2.7	3.8	
		200	P8	10	2.0	2.9	
		20	P9	10	2.1	2.3	2.6
		20	P9.5	10	2.9	4.6	4.6
F-XPDMS/HOF-PDMS	50/50	40			1.3	16.5	
F-XPDMS/ERF	50/50	40	P9	10	11.3	13.2	

Furthermore, the 50/50 F-XPDMS/ERF gel had a compression modulus that was *lower* than that of the equivalent gel that contained no ERF in the same electric field. Both effects may be linked with the possibility that the PANI particles suppressed the aggregation of the repeat groups with the higher dielectric constant in the electric field that was suggested for the gel that contained no ERF.

Figure 3 shows a comparison of the compression modulus of different 50/50 V-XPDMS/ERF composite gels vs the logarithm of the 100 MHz conductivity of the PANI particles contained in the ERF (see Table 4b); a figure using the 10 kHz conductivities looks similar. Some of the gels were cured in an electric field in order to permanently align the PANI particles as much as possible; this alignment could be observed under the microscope. Thus, these particles are referred to as prealigned below. Figure 3 shows that the compression modulus in a 3.3 kV/cm dc electric field could be increased by an order of magnitude by prealigning the PANI particles in the ERF during curing. One may also note in Figure 3 that the compression modulus in the applied dc electric field of both types of composite gels seems to have a small peak near the PANI particles with conductivity around 5.27×10^{-6} S/cm (PANI P8) whether the particles were prealigned or not. PANI particles that were not prealigned are referred to as "random".

The reason for the large increase in compression modulus when the PANI particles in the ERF are prealigned into columns is probably that the PANI particles that are close together and prealigned in the electric field direction attract each other more strongly than randomly placed particles in an electric field applied after curing of the gel. Also, columns of particles should be able to resist compression better than randomly placed particles.

Flexible Electrode Displacement Measurements in Applied Electric Fields

In our previous work using commercially available silicone gels and ERF's,[6] we showed that the displacement of the flexible electrodes surrounding the composite gels (1) increased with an increase in the applied dc electric field, (2) depended on the silicone/ERF ratio, and (3) decreased with increasing frequency of the applied electric field at constant applied (rms) voltage. In addition, we showed that the pure silicone gel and a sample of the silicone gel that had been swollen with silicone fluid had electrode displacements that were only one-sixth to one-fifth as large as that of the silicone gel/ERF composite under the same conditions. In the present work, we have investigated the effect of additional variables that could be controlled by preparation of our own gels and ERF's.

Necessity for Fluid in ERF. First, we investigated whether the liquid component of the ERF was a necessary component of the composite gel, or whether the ERF particles alone (without the liquid in which they were usually suspended) would also provide an electromechanical response. To this end, 5 wt % of many of the PANI samples from Table 4 were cured into a V-XPDMS matrix and electrode displacements ranging from 2 to 20 μm, not very reproducible, were observed in an 8 kV/cm dc field. Under the same conditions, the electrodes surrounding the pure V-XPDMS gel, with no PANI particles present, moved reproducibly about 10 μm. Therefore, the PANI particles alone had little effect on the electrode displacement, thus demonstrating the importance of the fluid phase of the ERF to the electromechanical response. ERF's containing 10 wt % PANI were used for most other measurements; this is in the range used with other ERF's by other investigators.

Effect of Dielectric Constant and Conductivity of ERF Particles. The discussion of "The ER Effect" above shows that the conductivity and dielectric constants of the particles are of great importance when considering the force of attraction between particles in an electric field. The different PANI samples whose dielectric constants and conductivities are shown in Table 4, parts a and b, were prepared to examine whether this was true in the present work. Figure 4, for example, shows the single-electrode displacement, in an 8 kV/cm electric field, of 40/60, 50/50, and 60/40 by volume *V*-XPDMS/ERF composite gels containing 200 cst 3MPDMS vs the 100 kHz conductivity of the PANI particles contained in the ERF. This figure shows a large effect of PANI particle conductivity on single-electrode displacement only in the case of the 40/60 XPDMS/ERF. A smaller effect was observed for the 50/50 XPDMS/ERF and essentially no effect for the 60/40 XPDMS/ERF.

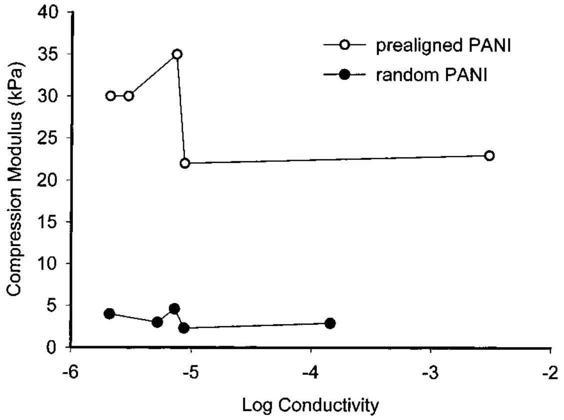

Figure 3. Compression modulus of 50/50 V-XPDMS/ERF composite gels, some containing random and others containing prealigned PANI particles, vs the logarithm of the 100 kHz conductivity of the PANI particles, in a 3.3 kV/cm dc electric field. The 20 cst 3MPDMS fluid and 10 wt % PANI were used for the ERF.

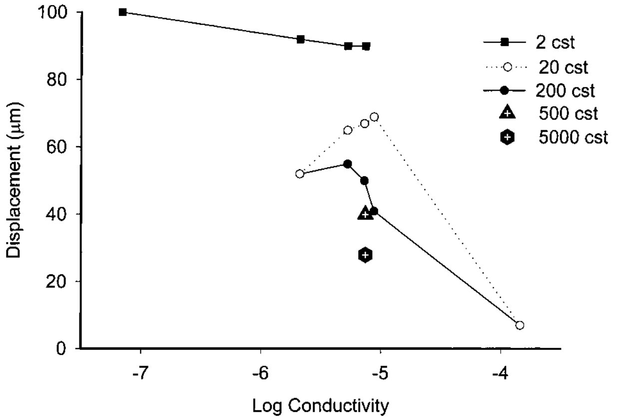

Figure 5. Single-electrode displacement vs 100 kHz conductivity of the PANI particles in 50/50 V-XPDMS/ERF composite gels in an 8 kV/cm dc electric field as a function of the viscosity of the 3MPDMS fluid used in the ERF. 10 wt % PANI was used in each ERF.

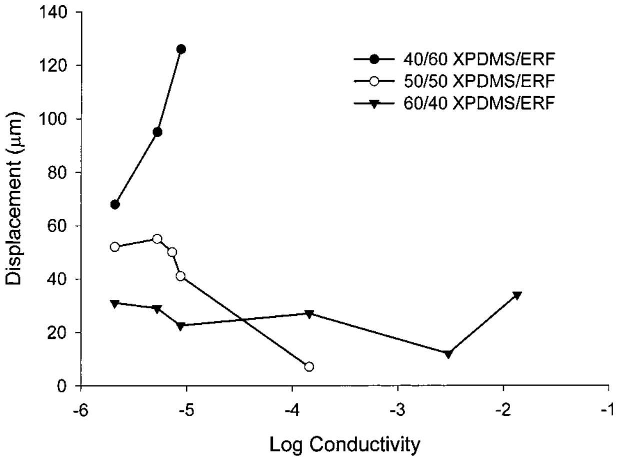

Figure 4. Single-electrode displacement vs 100 kHz conductivity of the PANI particles as a function of the percent ERF in the composite gel in an 8 kV/cm dc electric field. The 200 cst 3MPDMS fluid and 10 wt % PANI particles were used in each ERF.

Effect of Percent ERF. Figure 4 also shows the effect of both percent ERF in the composite gel and conductivity at 100 kHz of the PANI on electrode displacement. A graph using the 10 kHz conductivities looks very similar. The measured displacement of the electrodes increased when the volume percent of ERF was increased in the composite gel no matter which PANI sample was used when the PANI particle conductivity was small. This increase is reasonable for several reasons: (1) a larger percent of ERF in the composite gel means that the gel contained more 3MPDMS fluid and was thus mechanically weaker, and (2) the composite gels that contained the larger percent of ER fluid also contained more PANI particles per unit volume. It is the PANI particles that become polarized and exert attractive forces on each other and on the electrodes (see "Mechanism of the Electromechanical Response" below).

Although Figure 4 seems to indicate that electrode displacement increases almost monotonically with increasing ERF content of the composite gels, our previous work[6] showed that there was a particular ratio of silicone gel to ERF, 60/40 by volume in that system, at which maximum electrode displacement occurred. If there is a volume ratio of the V-XPDMS/ERF composite used in the present work at which a maximum electrode displacement could occur, it looks as if it would have to be 40/60 or lower. However, systems with volume ratios lower than 40/60 were too fluid to use between our electrodes.

Effect of Viscosity of ERF Fluid Component. Figure 5 shows the effect of the viscosity of the 3MPDMS fluid in the ERF and the 100 kHz conductivity of the PANI particles in the ERF on the single-electrode displacement of 50/50 V-XPDMS/ERF composite gels in an 8 kV/cm electric field. The single-electrode displacement was highest in the case of the 2 cst fluid for all PANI samples examined in the various ERF's. When the same PANI sample was used in different 3MPDMS fluids, the single-electrode displacement decreased as the 3MPDMS viscosity increased. Data obtained for prealigned PANI particles in the 200 cst fluid were within experimental error of those for random PANI in the same fluid in most cases. Thus, prealigning the PANI particles had almost no effect on electrode displacement even though the effect on compression modulus was quite large (Figure 3).

Inability to Use F-XPDMS/ERF. The F-XPDMS/ERF composite was originally chosen for study because the P9 particles have a lower dielectric constant than the partly fluorinated matrix and should, therefore, according to the dielectric theory be unable to exhibit the ER effect. (An attempt to produce fibrils in the pure ERF solution in an electric field was, as expected, unsuccessful.) It turns out, however, that, according to the leaky dielectric theory, specifically eq 5a, this system should exhibit the ER effect perfectly well. There appears to be a disagreement between theory and experiment. At any rate, use of the F-XPDMS/ERF composite between flexible electrodes led only to a slow displacement, taking about 10 s, of the electrodes over about 30 μm; the same phenomenon was observed when the F-XPDMS/HOF-PDMS mixture, without PANI particles, was used. The PANI particles thus had no effect on the electrode displacement in this system in which, as discussed above, the presence of the PANI particles led to a lowered compression modulus, 13.2 instead of 16.5 kPa, in a 3.3 kV/cm field electric field. Thus, it looks as if this system, in which the particles cause a decrease in compression modulus in an electric field, cannot

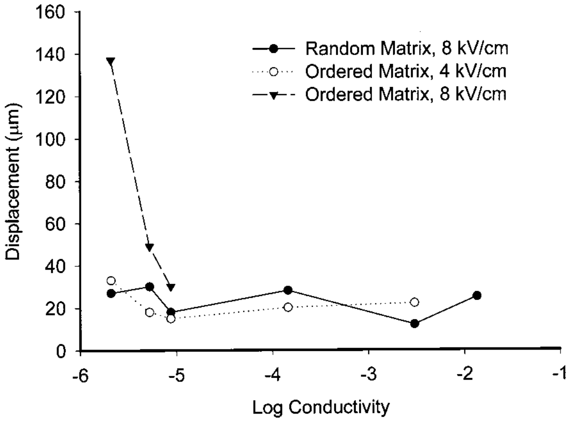

Figure 6. Single-electrode displacement vs 100 kHz conductivity of the PANI particles in the ERF as a function of the gel matrix type using 60/40 XPDMS/ERF composite gels. The 20 cst 3MPDMS fluid and 10 wt % PANI were used in the ERF.

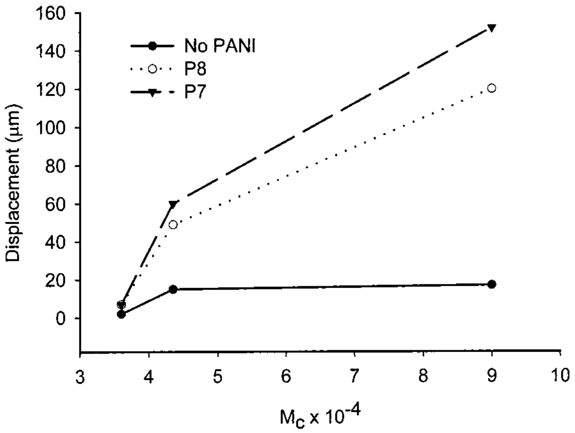

Figure 7. Single-electrode displacement vs the molecular weight between cross-links, M_c, using 60/40 C-XPDMS/ERF composite gels with two different PANI samples in an 8 kV/cm dc electric field. A control gel containing no PANI particles is also shown. The 20 cst 3MPDMS fluid and 10 wt % PANI were used in the ERF.

provide rapid movement of the electrodes in an electric field.

Necessity for Chemical Cross-Links in the Gel. It was of interest to find out whether a chemically cross-linked network was really necessary for this work. For this purpose, a high molecular weight, but not chemically cross-linked, PDMS was prepared using the vinyl addition reaction between a vinyl-terminated PDMS and a hydride-terminated PDMS. This polymer was viscoelastic, appearing gellike to short-term probing, but exhibiting long-term flow characteristics. The entanglements of the long chain acted as physical cross-links, as usual in such systems; the entanglement molecular weight of PDMS is around 9600 at 298 K.[23] When this high molecular weight PDMS system was placed between two flexible electrodes and an 8 kV/cm electric field was applied, the electrode displacement was small and slow; it took 8 s for each electrode to move 6 μm. When an ERF was added to the high molecular weight PDMS system and the same electric field applied, the displacement increased slightly to 12 μm, but the time response remained a slow 8 s. None of these nonchemically cross-linked networks returned to its original dimensions after the electric field was turned off. Chemical, not physical, cross-linking of the network was necessary to obtain a fast electromechanical response.

Effect of Type and Amount of Cross-Linking in the Gel. It was also of interest to find out whether random or ordered networks would give larger electrode displacements. Figure 6 compares the single-electrode displacement using a randomly cross-linked 60/40 V-XPDMS/ERF composite gel in an 8 kV/cm dc field with that using an ordered cross-linked 60/40 C-XPDMS/ERF composite gel in both a 4 and 8 kV/cm dc field vs the conductivity of the PANI particles in the ERF. The randomly cross-linked network did not have a noticeable electrode displacement in a 4 kV/cm dc electric field and therefore these data are not shown in figure. The C-XPDMS gel with an M_c of 4.0×10^4 was used because it had a M_c most similar to that of the V-XPDMS gel, 5.6×10^4. It can be seen in Figure 6 that the “model” network system, where the cross-links are at ordered, consistent intervals (C-XPDMS) exhibited the same electrode displacement at half the applied voltage needed for a randomly cross-linked network (V-XPDMS) with a similar molecular weight between cross-links. PANI particles with conductivities above 1.45×10^{-4} S/cm could not be used in the ERF’s in 8 kV/cm electric fields because the gels shorted out.

Figure 7 shows the single-electrode displacement vs molecular weight between cross-links for a 60/40 C-XPDMS/3MPDMS composite gel and for two different 60/40 C-XPDMS/ERF composite gels, one containing 10 wt % PANI P7 particles and the other containing 10 wt % PANI P8 particles in the ERF, in an 8 kV/cm dc electric field. This Figure shows that the electrode displacement increased as the molecular weight between cross-links increased in all the composite gels, but the PANI particles were necessary to obtain the larger electrode displacements. This again demonstrates the importance of the complete ERF for an appreciable electromechanical response in these composite gels. When the molecular weight between cross-links is increased in a gel, swelling of the gel increases in the same solvent. A more swollen cross-linked polymer should have a lower compression modulus than a less swollen polymer so that more compression will take place under similar circumstances. We may also note that the electrode displacement increased as the conductivity decreased from sample P8 to sample P7 (at all frequencies investigated), and the dielectric constant of the PANI particles increased (at all frequencies investigated).

PANI Particle Movement in a V-XPDMS/ERF Composite Gel in a DC Electric Field. It was of interest to find out whether the PANI particles in the composite gels moved in the electric field and whether such movement was connected to the magnitude of the electrode displacements. To find out how much the PANI particles moved, a number of optical micrographs were obtained. Figure 8A shows an optical micrograph of a 50/50 V-XPDMS/ERF composite gel with a number of PANI particles outlined. Figure 8B shows the locations of these particles after a 10 kV/cm dc electric field was applied; the circles show the original locations of these particles. Although the particles moved mostly in the field direction, a small amount of sideways motion was also observed. When the field was turned off, the PANI particles returned close to their original positions (Figure 8C). Table 6 shows the average component of the PANI particle movement in the field direction and the average single-electrode displacement measured in three different composite gels. Table 6 shows that there is only a slight correlation between the average particle

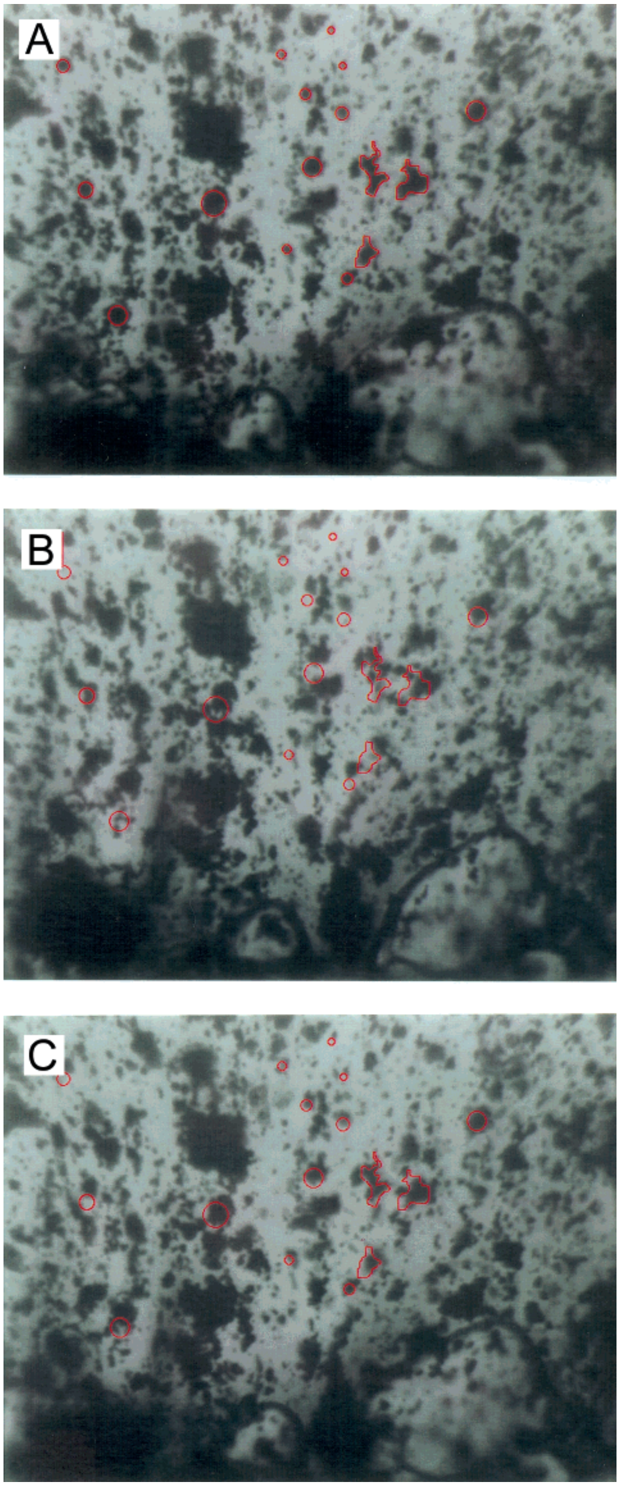

Figure 8. Optical micrographs of a 50/50 V-XPDMS/ERF composite gel prepared using the 200 cst 3MPDMS fluid and 10 wt % PANI P6 in the ERF: (A) number of representative PANI particles outlined in the system before application of an electric field; (B) same portion of the gel after application of a 10 kV/cm dc electric field, where the original position of each particle (as seen in part A) has been outlined; (C) same portion of the gel after the field was turned off, where the original position of each particle (as seen in part A) has been outlined and it can be seen that the particles have returned to positions close to their original positions.

Table 6. Average Component of PANI Particle Movement in the Field Direction and Average Electrode Displacement Observed in Various V-XPDMS/ERF Systems in a 10 KV/cm Dc Field[a]

composite gel	av particle motion (μm)	av single electrode displ (μm)
50/50 V-XPDMS/ERF (PANI P6)	25	58
50/50 V-XPDMS/ERF (prealigned PANI P8)	10	45
60/40 V-XPDMS/ERF (PANI P6)	22	35

[a] The 20 cst 3MPDMS fluid containing 10 wt % PANI was used in each ERF.

motion in the field direction and the single-electrode displacement between the different gels. The average PANI particle motion in the 50/50 gel containing PANI P6 was about 14% more than that observed in the 60/40 gel, but the single-electrode displacement in the cell containing the 50/50 gel was as much as 65% greater than that in the cell containing the 60/40 gel. Furthermore, in the gel containing the prealigned P8 particles, the particle motion was much smaller than in the other gels, while the electrode displacement was close to the average of that observed with the other two gels. It is reasonable to say that the prealigned particles were already close to their lowest energy positions in an electric field so that they did not have to move much.

A Mechanism for the Electromechanical Response. The discussion above indicates that the compression modulus of all the gels that allowed full electrode displacement in about 100 ms increased in an applied dc electric field after the addition of PANI particles. In the one case, the F-XPDMS/HOF-PDMS, in which the compression modulus did not increase when PANI particles were added, the electrode displacement took as long as 10 s. Therefore, any mechanism proposed for fast electrode displacement should take note of this effect.

Actually, any discussion of a mechanism for the rapid electromechanical response must involve the force between the electrodes in an electric field. The force per unit area of electrode, F/A, between two infinite parallel plate electrodes with a simple dielectric between them, ignoring end effects, is[24]

$$\frac{F}{A} = \frac{\epsilon_0 \epsilon E^2}{2} \tag{8}$$

where ϵ is the dielectric constant of the material between the electrodes. Equation 8 shows that, in this approximation, the force between the electrodes increases with increasing dielectric constant of the medium between them. Since we have not found a similar equation in the leaky dielectric theory, we use eq 8 as a first approximation. To estimate the dielectric constants of our composite gels to use in eq 8, we use the logarithmic mixing rule[25]

$$\log \epsilon = \varphi_1 \log \epsilon_1 + \varphi_2 \log \epsilon_2 \tag{9}$$

where ϕ_1 and ϕ_2 are the volume fractions of the two components of the mixture, respectively, and ϵ_1 and ϵ_2 are their respective dielectric constants. According to Von Hippel,[25] eq 8 was confirmed by Báchner for mixed dielectrics containing rutile (TiO_2), the other component being zirconium oxide, polystyrene, or cellulose plastic. Equation 9 will probably give too low a value for ϵ in the case of the samples containing prealigned PANI; Conrad and Chen[9] found that an electrorheological fluid containing aligned zeolite particles had a ~50% higher dielectric constant than one in which the particles were randomly dispersed.

Table 7. Estimated and Observed Electrode Displacements Using 50/50 XPDMS/ERF Composites in an 8 KV/cm Dc Electric Field, Where Each ERF Contained 10 wt % PANI

PANI	fluid (cst)	modulus (kPa) used for calcn of electrode displacement	$\epsilon_{\text{composite}}$ calcd from eq 8	electrode displacement calcd (μm)	electrode displacement obsd (μm)
P6	20	2.9	3.3	8.7	7
P7	20	3.9	2.6	5.0	52
P7	200	3.6	2.6	5.4	52
P8	20	3.4	2.5	5.2	65
P8	200	2.9	2.5	6.1	55
P9	20	2.6	2.5	6.9	69
P9.5	20	4.6	2.5	3.9	67

Let us note that the force of attraction between the electrodes exists even when the electrodes cannot move in response to this force, either when the electrodes are fixed in space or when their motion is mechanically imposed, as it was in the compression modulus determination in the Instron. The measured compression modulus in the electric fields used was therefore not affected by the force of attraction between the “electrodes” although the displacements of the flexible electrodes *were* so affected.

We can now estimate the force between the electrodes in an electric field, first using eq 9 to calculate the dielectric constant of our composite gels. We use the measured value of 2.4 for the dielectric constant of the XPDMS gel and the PDMS fluid, considered as a single component in the composite, and the values of the dielectric constants of PANI P6, P7, P8, P9, and P9.5 at 10 kHz from Table 4. Since there was 10 wt % PANI in all the samples shown in Table 7, the volume fraction of the PANI in all these 50/50 XPDMS/ERF composites was about 0.05. Table 7 shows the values obtained. Equating the force per unit area in eqs 1 and 8, we can then estimate the expected electrode displacements in various applied electric fields using the measured compression moduli and the estimated dielectric constants of the composites used. If we assume that the moduli measured at lower electric fields in Table 5 remain relatively constant up to 8 kV/cm, we obtain predicted compression distances (these are twice the predicted single-electrode displacements) of the composite gel samples shown in Table 7 together with the observed values for all composite gels for which both compression modulus and single-electrode displacements were available.

Table 7 shows that the observed single-electrode displacements were much greater, as much as an order of magnitude greater, than those that could be predicted from the compression moduli in the case of ERF’s made from the lower dielectric constant, less conductive PANI samples. Only when PANI P6, which had a dielectric constant and conductivity several orders of magnitude greater than that of the other PANI’s in Table 7, was used in the ERF was the observed single-electrode displacement somewhat less than that calculated. These results indicate that there are possibly two factors at work: (a) the dielectric constant of the PANI particles contributes more to the force between electrodes than eq 9, inserted into eq 7, would indicate, and (b) a relatively high conductivity of the PANI particles probably decreases the force between the electrodes below that calculated using the pure dielectric theory (eq 7). When PANI P6 was used, factor b predominated, while factor a predominated when the other PANI’s in Table 7 were used. It would be useful to have new theoretical treatments or simulations to find out what the force between the electrodes should be using the leaky dielectric theory, in general, and for a medium containing discrete leaky dielectric particles in a leaky dielectric medium, in particular. Suffice it to say at this time that most of our XPDMS/ERF composites resulted in much greater single-electrode displacement than could be justified from consideration of their measured compression modulus and the pure dielectric theory. At least one composite resulted in a much smaller single-electrode displacement.

Equation 8 shows that the force between electrodes in an electric field increases as the dielectric constant of the material between the electrodes increases. The polarization of the material increases the force. It seems reasonable to state that the polarization of the electrorheological particles, in the absence of relatively high conductivity of the particles, is great enough to attract the electrodes more than allowed for in our simple calculations which did not consider the particles separately.

An Improved Actuator. A small electromechanical actuator was previously created using the 60/40RTV/ERF gel.[6] In the present work, an improved actuator was built using a 50/50 XPDMS/ERF composite gel with prealigned PANI P8 using the 20 cst 3MPDMS fluid and 10 wt % PANI. This actuator was able to lift a 20% heavier flag, weighing 1.2 g, than the earlier flag-waving actuator made from 60/40RTV/Bayer ERF. The response time of both actuators was less than 100 ms.

Conclusions

1. Fast electromechanical response of PDMS/PANI composite gel systems, occurring in ~100 ms, required chemical cross-links in the gel, an added fluid, and PANI particles; the compression modulus of these gels in an electric field was decreased when the PANI particles were absent. A much slower response was obtained using a composite gel whose compression modulus in an electric field increased when the PANI particles were absent.
2. The compression modulus of composite gels containing prealigned PANI particles was an order of magnitude greater than that of composite gels containing random PANI particles, holding all other variables constant.
3. Ordered polymer networks, when used for the composite gels, allowed for greater electrode displacements than random networks, holding all other variables constant.
4. The movement of the PANI particles in the composite gel when the electric field is turned on and off is not well correlated with the electrode displacement.

Acknowledgment. The authors would like to acknowledge the Office of Naval Research for support under Grant N00014-95-0136. Thanks also to Toh-Ming Lu, Department of Physics, and Don Van Steele, Department of Materials Engineering, for the use of their equipment.

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MA0108158