

## CHAPTER IV

### EFFECT OF ELASTOMER MATRIX TYPE ON ELECTROMECHANICAL RESPONSE OF CONDUCTIVE POLYPYRROLE/ELASTOMER BLENDS

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#### Abstract

Electrorheological properties of pristine elastomers and polypyrrole/elastomer blends were investigated to identify the most suitable elastomer to be used for electroactive actuator applications. Seven types of elastomer: poly acrylate copolymers (AR70, AR71, AR72, and SAR), poly(dimethyl siloxane) (PDMS), poly(styrene butadiene) (SBR), and poly(styrene isoprene styrene) (SIS) were chosen as the candidate dielectric elastomers. Experiments were carried under the oscillatory shear mode and with applied electric field strength varying from 0 to 2 kV/mm. For pure elastomers, the dynamic modulus,  $G'$  response varied by 7 order of magnitude depending on the type of elastomer and electric field strength; the storage modulus ( $G'$ ) increased monotonically with increasing electric field. The storage modulus sensitivity,  $\Delta G'/G'_0$ , increased with electric field strength and attained maximum values of 97.15 % for SBR, 148.82 % for SAR, 232.64% for AR70, 12.78 %, for PDMS, 69.47% for AR71, 54.38% for AR72, and 10.00 % for SIS at the electric field strength of 2 kV/mm. There appear to be correlations between the elastomer storage modulus sensitivity and  $G'_0$  and/or electrical conductivity. For the undoped polypyrrole/elastomer blends with the particle concentrations of 1, 2, 3, 4 and 5 vol%, the storage modulus response increased linearly with concentration in the absence of electric field but nonlinearly with electric field on, possibly due to the obstruction of the matrix dipole interaction at small concentration and the non uniform polypyrrole particle dispersion in the matrix.

**Keywords:** Electroactive polymer, Poly(pyrrole), Elastomer, Dynamic moduli  
Electrorheological properties

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## Introduction

Dielectric elastomers are employed as actuator materials since they can yield large deformations through conversion of the electrical energy directly into the mechanical work. They can be considered to belong to a group of so called electroactive polymers (EAPs) <sup>[1]</sup>. Electroactive polymers (EAPs) offer promising and novel characters: lightweight, high energy density and high flexibility; they are material candidates for muscle-like actuators. Examples of elastomers are poly(acrylate) and styrenics; they are widely used in many industrial applications such as computer parts, footwear, and inner automobile parts. Differences in chemical structures, mechanical properties, and electrical conductivity amongst various elastomers lead to different electromechanical responses under electric field.

Recently, incorporation of a conductive polymer into a dielectric elastomer forming a blend has been of interest. Conductive polymers can offer a variety of benefits to the host elastomer: variable conductivity, improved thermal stability, and mechanical properties. Example are: the PET/Ppy composite for sound shielding<sup>[2]</sup>; carbon fiber was used to reinforce polydimethyl siloxane (PDMS)/polypyrrole (Ppy) blend to fabricate a flexible fiber wire<sup>[3]</sup>; poly(p-phenylenes)-silicone elastomer blend properties were investigated for actuator applications.<sup>[4]</sup>

In our work, we are interested in various elastomers and polypyrrole/elastomer blends as candidates for artificial muscles or actuators. The mechanical, electrical, and electromechanical properties were investigated in terms of electric fields strength, elastomer type, and conductive polypyrrole particle concentration.

## Experimental Part

### Materials

Pyrrole monomer (Sigma) was stored at 8–10 °C and dried with calcium hydride (CaH<sub>2</sub>, Fluka) for 24 h prior to use. Ammonium persulfate (APS, BDH Laboratory Supplies) was used as the oxidant without further purification. Dodecylbenzene sulfonic acid (DBSA, Sigma) was used as received as the dopant. M-Cresol (Merck) was used as the solvent in dissolving synthesized polypyrrole. polydimethylsiloxane (PDMS, Aldrich), poly(styrene isoprene styrene), (SIS, Shell), poly(styrene butadiene), (SBR, Dow Chemicals), poly acrylate copolymers, (AR71 and AR72, Zeon), and poly acrylate copolymer, (AR70, and SAR, Dow Chemicals) were used as the elastomers.

### Preparation and Doping of Poly(pyrrole) (Ppy)

Poly(pyrrole) was chemically synthesized following the method of Lee et al.<sup>[5]</sup>. Pyrrole monomer was dried and mixed with CaH<sub>2</sub> at the ratio of 100 g of CaH<sub>2</sub> per liter of pyrrole, and the reaction was allowed to proceed for 24 h before use. 0.3 mole of dried pyrrole monomer and 0.3 mole of DBSA were dissolved in 500 ml of distilled water. The mixture was stirred vigorously for 15 min at 0 °C. 0.06 mol of APS in 100 ml of distilled water was slowly added to the mixture solution at a rate of 5 ml/min. Reaction was allowed to proceed for 40 h and then terminated by pouring in 20 ml of methanol. The resultant polypyrrole powder was filtered and washed sequentially twice with 50 ml of distilled water, 50 ml of methanol, and 50 ml of acetone. The washing procedure described was repeated again followed by filtering and drying in a vacuum oven at 25 °C for 12 h. Synthesized poly(pyrrole) was grinded, sieved, and stored in a desiccator. For synthesizing undoped polypyrrole, the procedure was the same as that of the doped polypyrroles except the DBSA was not added into the mixture

### Preparation of Elastomers and Ppy/Elastomer Blends

All of elastomer specimens were formed by solution casting except PDMS which was used in the liquid form. AR70, SBR, SAR specimens were formed by water evaporation, but SIS, AR71, and AR72 specimens were formed by toluene

evaporation at 50 % vol/vol. The solutions were cast on to a mold (diameter 25 mm) and bubbles in solutions were removed under a vacuum atmosphere at 25°C for 12 hours.

The poly(pyrrole) powder was sieved with a mesh size of 50  $\mu\text{m}$  and dried at room temperature for 12 hours prior to use. The blends were prepared by mechanical blending of undoped synthesized poly(pyrrole) at various particle concentrations (1, 2, 3, 4 and 5%vol.). A specific amount of particle was then added and the mixture was mechanically stirred for 3 hours to disperse the particles. The mixture was cast on the mold (diameter 25 mm) and specimens were dried in oven at 40°C for 12 hours.

### Characterization and Testing

Fourier transform infrared spectrometer (Bruker, Equinox 55/FRA 1065) was operated in the absorption mode with 32 scans and a resolution of  $\pm 4 \text{ cm}^{-1}$ , covering a wavenumber range of 4000-400  $\text{cm}^{-1}$  using a deuterated triglycine sulfate detector. Optical grade KBr (Carlo Erba Reagent) was used as the background material. UV-Visible spectra (Perkin-Elmer, Lambda 10) of undoped and doped polypyrrole were measured in the absorbance mode in the wavelength range of 800-200 nm, scan speed of 240 mm/min, a slit width of 2.0 nm, and using a deuterium lamp as the light source. Ppy solution was prepared by grinding synthesized Ppy into a fine powder and 0.05 g of Ppy was dissolved in 100 ml. of m-cresol. A particle size analyzer (Malvern Instruments Ltd. Masterizer X Version 2.15) was used to determine the particle size of poly(pyrrole). The 45 mm lens was used in this experiment. A scanning electron microscope (JOEL, model JSM-5410OW) was used to examine the morphological structure and PPy dispersion. The samples in pellet form was cut into small pieces and adhered on a brass-stub by using an adhesive tape. Then, they were coated with thin layer of gold by using a ion-sputtering device. The magnifications used were 7,500, 1,500 and 350 times. A thermal gravimetric analyzer (DT-TGA 1790) was used to measure the degradation temperature with the temperature scan from 60 to 1000°C, and a heating rate of 10°C/min. The samples were weighed in the range of 1-5 mg, loaded into a platinum

pan, and heated under N<sub>2</sub> flow. An X-ray diffraction microscope (Rigaku, DMAX 2200) was used to determine the degree of crystallinity and the crystal size of polypyrrole particles.

A two-point meter was used for measuring electrical conductivity of polypyrrole in the sheet form. The Keithley, Model 8009 probe was used for measuring electrical conductivity of the elastomers and the elastomers blends. The probe was in contact with a surface of each film sample and it was also connected to a source meter (Keithley, Model 6517A) which supplied a constant voltage source and the output current. The applied voltage was plotted versus the output current to determine the linear Ohmic regime of each sample. The applied voltage and the current change in the linear Ohmic regime were converted to the electrical conductivity of the polypyrrole sheets using equation (1), and of the elastomers and elastomer blends using equation (2) as follow:

$$\sigma = \frac{1}{\rho} = \frac{1}{R_s \times t} = \frac{I}{K \times V \times t} \quad (1)$$

where  $\sigma$  is the specific conductivity (S/cm),  $\rho$  is the specific resistivity ( $\Omega \cdot \text{cm}$ ),  $R_s$  is the sheet resistivity ( $\Omega$ ),  $I$  is the resultant current (A),  $K$  is the geometric correction factor,  $V$  is the applied voltage (voltage drop, V), and  $t$  is the pellet thickness (cm).

$$\sigma = \frac{1}{\rho} = \frac{t \times I}{22.9 \times V} \quad (2)$$

where  $\sigma$  is the specific conductivity (S/cm),  $\rho$  is the specific resistivity ( $\Omega \cdot \text{cm}$ ),  $I$  is the resultant current (A),  $V$  is the applied voltage (voltage drop) (V), and  $t$  is the sheet thickness (cm).

A melt rheometer (Rheometric Scientific, ARES) was used to measure electrorheological properties. It was fitted with a custom-built copper parallel plate fixture (diameter of 25 mm). A DC voltage was applied with a DC power supply (Instek, GFG 8216A), which can deliver electric field strength to 2 kV/mm. A digital multimeter (Tektronix, CDM250) was used to monitor voltage input. In these experiments, the oscillatory shear strain was applied and the dynamic moduli ( $G'$  and  $G''$ ) were measured as functions of frequency and electric field strength. Strain

sweep tests were first carried out to determine the suitable strain to measure  $G'$  and  $G''$  in the linear viscoelastic regime. The appropriate strain was determined to be 700% for pure PDMS fluid, and 1% for other elastomers. Then frequency sweep tests were carried out to measure  $G'$  and  $G''$  of each sample as functions of frequency. The deformation frequency was varied from 0.1 to 100 rad/s. Prior to each measurement, the sample was presheared at a low frequency (0.039811 rad/s), and then the electric field was applied for 10 minutes to ensure the formation of equilibrium polarization before each measurement was taken. Each measurement was carried out at the temperature of 27°C and repeated at least two or three times.

## Results and Discussion

### Characterization of Poly(pyrrole)

The FT-IR spectrum of the undoped Ppy was recorded to identify characteristic absorption peaks<sup>[6,7]</sup>. The characteristic peaks of undoped Ppy were found at 3130  $\text{cm}^{-1}$ , 3000-2800  $\text{cm}^{-1}$ , 1588  $\text{cm}^{-1}$ , 1541  $\text{cm}^{-1}$ , 1140  $\text{cm}^{-1}$ , 1138  $\text{cm}^{-1}$  and 769  $\text{cm}^{-1}$ . These peaks can be assigned to the N-H stretching in pyrrole ring; the C-H stretching of aliphatic; the asymmetric C=C/C-C stretching vibration, symmetric C=C/C-C stretching vibration; and the C-H in plane bending and out of plane vibration of ring.<sup>[6-7]</sup> The FT-IR spectrum of the doped Ppy was recorded to identify characteristic absorption peaks<sup>[7]</sup>. The characteristic peaks of doped Ppy were found at 1176  $\text{cm}^{-1}$  and 617  $\text{cm}^{-1}$ . This peaks can be assigned to the S=O stretching vibration of DBSA dopant, and the characteristic vibration of DBSA dopant<sup>[7]</sup>.

The UV-visible absorption spectra of undoped poly(pyrrole) in *m*-cresol solution showed a reflectance peak at 340 nm corresponding to the  $\pi$ - $\pi^*$  transition of the neutral pyrrole trimer, and a peak at 440 nm corresponding to the  $\pi$ - $\pi^*$  transition of the polypyrrole in an oxidized form<sup>[7]</sup>. After doping, the polypyrrole was oxidized leading to the higher intensity peak at 440 nm<sup>[7]</sup>.

The TGA thermogram of undoped Ppy showed a degradation step at 400°C corresponding to the backbone degradation<sup>[8]</sup>. The doped Ppy showed two degradation steps at 500°C and 695°C, corresponding to the bipolarlon state and the

polarlon state of poly(pyrrole) obtained through the doping process; the bipolarlon state contains the double bond between pyrrole rings causing it to have higher thermal stability. For the elastomers, the degradation step appeared in only one transition range of 383 to 478 °C; PDMS has the highest thermal stability due to the Si-O bond in its backbone which is stronger than the C-C bonds of other elastomers.

The mean particle diameter of Ppy was determined to be approximately 25 µm for undoped polypyrrole and 37 µm for doped polypyrrole with standard deviations of 0.35 and 0.07 µm, respectively. The particle microstructure was observed using a scanning electron microscope (SEM). Figure 1 shows that the various shapes of the doped Ppy particles are larger than those of undoped polypyrrole particles.

X-ray diffraction patterns were taken between  $2\theta = 5-35^\circ$ . The characteristic peaks of Ppy were found at  $2\theta$  equal to about 20.32 and 26.26, with corresponding d-spacings of 4.37 and 3.39 Å, respectively. The intensity of high angle peak at  $26.06^\circ$  can be referred to the interplanar distance of pyrrole – pyrrole and polypyrrole-counter ion. The peak at  $20.32^\circ$  can be referred to the polypyrrole-counter ion or inter-counter ion interaction scattering<sup>[9]</sup>.

The specific conductivity values of undoped and doped Ppy were measured by the custom-built two point probe (Keithley, Model 6517A). The elastomer and blend electrical conductivity values were measured by the resistivity testing fixture (Keithley, Model 8009). The specific conductivity of undoped Ppy was 0.52 S/cm with standard deviation of 4.37E-02 S/cm. The specific conductivity of doped Ppy was 4.07 S/cm with a standard deviation of 3.16E-01 S/cm. The specific conductivity of polymer blends increases with increasing particle concentration varying from 1.28E-12 S/cm to 5.33E-12 S/cm.

#### Electrorheological Properties of Pure Elastomers

#### Effect of Storage Modulus of Elastomers

The effect of electric field strength on the rheological properties of the various pure elastomers was investigated in the range of electric field strength between 0-2 kV/mm. Figure 2 shows the storage modulus response ( $\Delta G'$ ) of various elastomers as functions of electric field, at frequency 1 rad/s, strain 1%, and at 27 °C (for PDMS, the strain used was 700%). The storage modulus  $G'(\omega = 1 \text{ rad/s})$  of the elastomers generally increases with electric field (0.5-2kV/mm). Amongst the various elastomers tested, the variation in  $\Delta G'$  covers 6 orders of magnitude between  $10^{-2}$  to  $10^4$  Pa. SAR has the highest  $\Delta G'$ , whereas PDMS possesses the lowest  $\Delta G'$ . We may note that the storage modulus in the absence of electric field ( $G'_0$ ) or the stiffness of an elastomer naturally depends on the chemical structure of the elastomer.  $G'_0$  values vary from 19,666 Pa to 50,606 Pa for PDMS, SBR, AR72, AR70, AR71, SAR, and SIS respectively; these values are tabulated in Table 1. Figure 3 shows the storage modulus sensitivity values ( $\Delta G'/G'_0$ ) at electric field strengths of 1 and 2 kV/mm vs.  $G'_0$  of the elastomers tested. It can be seen that ( $\Delta G'/G'_0$ ) first increases and then decreases monotonically with  $G'_0$ .

As an electric field is applied, electrical dipole moments are generated and the electrostatic interaction between the polymer chains and particles are induced leading to an intermolecular interaction acting like electrical crosslinks, thus the elastomer becomes stiffer or the storage modulus increases. The storage modulus sensitivity ( $\Delta G'/G'_0$ ) of AR70 and AR71 elastomers are highest. These elastomers contain a balance between the solid-like and the fluid-like behaviors; they have enough flexibility within the matrix in the absence of electric field such that its structure rigidity can be further induced with electric field. On the other hand, the lower storage modulus sensitivity of PDMS is a consequence of the fact that the matrix has a more fluid-like structure than other systems; movement and relaxation of polymer chains are still freely allowed in the presence of electric field and consequently they are affected with lesser intermolecular interactions. For the elastomers with higher storage modulus or  $G'_0$ , such as SAR, and SIS, they contain relatively more solid-like structures. The polymer segments and chains are initially



quite rigid and fixed in the absence of electric field; they simply cannot respond any further to the intermolecular interaction under applied electric field<sup>[10]</sup>.

#### Effect of Electrical Conductivity of Elastomers

The effect of electrical conductivity of elastomers on the rheological properties of pure elastomers with electric field strengths of 1 and 2 kV/mm were investigated in the range of electrical conductivity of elastomers between  $10^{-16}$  to  $10^{-6}$  S/cm. The elastomers are SIS, SBR, SAR, AR70, AR72, AR71, and PDMS respectively. Figure 4 shows the storage modulus sensitivity ( $\Delta G'/G'_0$ ) vs. electrical conductivity of elastomers. The storage modulus sensitivity increases with increasing elastomer electrical conductivity up to  $9.63 \times 10^{-13}$  S/cm. At higher electrical conductivity, the storage modulus sensitivity decreases monotonically. The highest storage modulus sensitivity belongs to AR70 with a value of 1.07 and 2.32 at electric field strengths of 1 and 2 kV/mm, respectively. The chemical structure of acrylate copolymer contains the carboxylate side group which can be more susceptible towards induced dipole moment relative to the styrenics elastomer, hence a stronger interaction and higher storage modulus sensitivity.

#### Effect of Particle Concentration

The effect of electric field strength on the rheological properties of pure AR70 and polymer blends at various concentrations was investigated in the range of electric field strength between 0-2 kV/mm. Figure 5 shows  $G'$  vs. frequency of AR70:Ppy\_un\_4 blend at various electric field strengths. The blend contains 4% v/v of undoped polypyrrole particles. Generally,  $G'$  increases monotonically with electric field at all frequencies investigated. Figure 6 shows the storage modulus without electric field  $G'_0$  as a function of Ppy particle concentration.  $G'_0$  increases linearly with Ppy concentration from 0 to 5 % v/v. At electric field strength of 2kV/mm,  $G'_{2kV}$  decreases with increasing concentration from 0 to 2 %v/v, and then increases linearly from 2 to 5 %v/v. Our results suggest that polypyrrole particles can

act as fillers which can support stress resulting in increase in the storage modulus in the absence of electric field. In the presence of electric field, the initial decrease of  $G'_{2kV}$  at low Ppy concentrations may stem from the obstruction of dipole moment interaction within the matrix; at this low Ppy concentration the particle-particle dipole interaction is not sufficiently strong. Other factor may include the morphology effect created by the sedimentation of polypyrrole particles during the solvent casting process. At higher Ppy concentrations, the effect of particle-particle dipole interaction becomes dominant and thus we find a linear increase in  $G'_{2kV}$ . The linear dependence of storage modulus on the particle concentration has been predicted by the following equation<sup>[4]</sup>:

$$G' = 3\varepsilon_0 \varepsilon_c \beta^2 E_0^2 \phi \quad (3)$$

where  $G'$  is the storage modulus (Pa),  $\varepsilon_m$  is the matrix electric permittivity,  $\beta_c$  ( $(\sigma_p - \sigma_c) / (\sigma_p + \sigma_c)$ ) is the electrical conductivity mismatch parameter,  $E_0$  is the apparent electric field strength,  $\phi$  is the volume fraction of particles, and  $\sigma_p$ ,  $\sigma_c$  are the electrical conductivities of the particle and the polymer matrix, respectively.

Concentration effect has been reported by others researchers. Krause *et al.*<sup>[11]</sup> found the compression modulus of polyaniline and PDMS blends was lower than that of pure PDMS as PDMS aggregation was suppressed by the polyaniline particles. Puvanattvattana *et al.*<sup>[10]</sup> found that the storage and loss moduli of polythiophene/polyisoprene blends increased with increasing Pth concentration. Shiga *et al.*<sup>[4]</sup> reported a similar effect for poly(p-phenylene)/silicone elastomer. They found that the enhancement of both the storage and loss shear moduli was negligible below 10.8 vol%, but increased dramatically above this threshold concentration.

#### Time Dependence of the Electrorheological Response

Finally, we investigated the temporal characteristic of pure AR70 and a polymer blend at particle concentration of 4 vol% (AR70:Ppy\_un\_4), at electric field strengths 1 and 2 kV/mm. The temporal characteristic of each sample

was recorded in the linear viscoelastic regime at a strain of 1 %, and frequency of 1 rad/s. Figure 7 shows the response in  $G'$  of the pure AR70 system to electric field strengths of 1 and 2 kV/mm during a time sweep test, in which an electric field was turned on and off alternately at every 500 s. At the electric field of 1 kV/mm,  $G'$  immediately increased and rapidly reached a steady-state value. Then, with the electric field off,  $G'$  decreased and recovered to nearly its original value; thus pure AR70 is a reversible system at 1 kV/mm. At the electric field of 2 kV/mm, the response of  $G'$  can be divided into two regimes: the initial regime in which  $G'$  rapidly overshooted to a large value on the first cycle followed by an irreversible decay with electric field off; and the steady state regime in which  $G'$  subsequently exhibited a reversible cyclic response. The time required for  $G'$  to reach the steady-state value on applying the field is called the induction time,  $\tau_{ind}$ . As shown in Table 3, the  $\tau_{ind}$  is independent of electric field strength; they are 424 and 433 sec at electric field strengths equal to 1 and 2 kV/mm, respectively. The time required for  $G'$  to decay towards its steady-state value when the electric field is turned off is called the recovery time,  $\tau_{rec}$ . It is essentially independent of the electric field strength, as shown in Table 2. The recovery times are 436 and 466 sec at electric field strength equal to 1 and 2 kV/mm, respectively. The independence of  $\tau_{rec}$  on the electric field strength suggests that the strains induced were small such that the relaxations were nearly the same.

Figure 8 shows the temporal response of AR70 and Ppy 4 vol% blend (AR70:4\_un) at electric field strengths of 1 and 2 kV/mm, respectively. The AR70:4\_un is clearly a reversible system at both of electric field strengths. After few initial cycles of electric field on and off, the response of  $G'$  reached a steady state regime. In this regime, the  $G'$  response at 2kV/m is greater than that of 1kV/mm. As electric field was turned off, the interactions between poly(pyrrole) particles and between particles and the matrix are quite reversible. Essentially the dipole bonding between adjacent poly(pyrrole) particles and the residual dipole moments inducing permanent interparticle interactions were not imminent. Both  $\tau_{ind}$  and  $\tau_{rec}$  slightly decreases with increasing field strength. As shown in Table 3,  $\tau_{ind}$  are 337 and 309

sec and  $\tau_{rec}$  are 207 and 138 sec at electric field strengths equal to 1 and 2 kV/mm, respectively.

### Conclusions

In this study, the effect of matrix type in the absence electric field ( $G'_0$ ), the effect of particle concentration with and without electric field on the dynamic moduli,  $G'$  and  $G''$ , under the oscillatory shear mode at electric field strength varying from 0 to 2 kV/mm were investigated. For pure elastomers, the storage modulus ( $G'$ ) monotonically increased with increasing electric field strength. We associate this finding with the increase in the number of electrical strands created by electric field. The AR70 elastomer appeared to have the largest storage modulus sensitivity under electric field.

For the effect of poly(pyrrole) particle concentration on the polymer blends with the concentrations of 0, 1, 2, 3, 4, and 5,  $G'_0$  increased linearly with particle concentration as polypyrrole particles within the matrix acted as effective fillers. In the presence of electric field, the particle-particle dipole interaction operated and dominated at relatively high concentrations. At low concentration, the presence of Ppy particles appeared to obstruct the number of electrical strands that can be generated.

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**Table 1** Rheological properties of various matrices

Materials	$G'_0$ (Pa)	$G'_{2kV}$ (Pa)	$\Delta G'_{2kV}$ (Pa)	$\Delta G'_{2kV}/G'_0$	$\sigma_{dc}$ (S/cm.)
AR 70 <sup>a)</sup>	19,666	65,418	45751	2.3264	9.63E-13
AR71 <sup>b)</sup>	31,240	89,761	58520	1.8732	5.16E-12
AR72 <sup>b)</sup>	12,333	17,025	4692.6	0.3805	1.03E-12
PDMS, 700%	0.2391	0.2696	0.0306	0.1278	2.35E-06
SAR <sup>a)</sup>	38,127	94,869	56742	1.4882	7.09E-15
SBR <sup>a)</sup>	21,557	36,429	14872	0.6899	5.55E-15
SIS <sup>b)</sup>	50,606	55,668	5061.7	0.1000	6.51E-17

All of materials were tested at frequency =1 rad/s, strain 1% and, temperature = 27 °C (except PDMS 700% was tested at strain 700%)

a) films were formed by water solution casting

b) films were formed by toluene solution casting

$G'_0$  is storage modulus without electric field

$\Delta G'_{2kV}$  is the the storage modulus responses between  $G'_0$  and  $G'_{2kV}$

$\Delta G'_{2kV}/G'_0$  is sensitivity of the storage modulus at 2 kV

**Table 2** Rheological properties of Ppy/elastomer blends of various particle concentrations

Materials	$G'_0$ (Pa)	$G'_{2kV}$ (Pa)	$\Delta G'_{2kV}$ (Pa)	$\Delta G'_{2kV}/G'_0$	$\sigma_{dc}$ (S/cm.)
AR 70	19,666	65,418	45,751	2.3264	9.63E-13
AR70:Ppy_un_1	29,540	54,764	25,224	0.8539	1.28E-12
AR70:Ppy_un_2	36,526	40,763	4,237.3	0.1160	1.30E-12
AR70:Ppy_un_3	45,020	55,635	10,615	0.2358	4.03E-12
AR70:Ppy_un_4	44,658	64,032	19,374	0.4338	4.94E-12
AR70:Ppy_un_5	59,303	82,940	23,637	0.3986	5.33E-12

Specimens were tested at frequency = 1 rad/s, strain 1% and, temperature = 27 °C  
 Films were formed by water solution casting.

$G'_0$  is storage and loss modulus without electric field

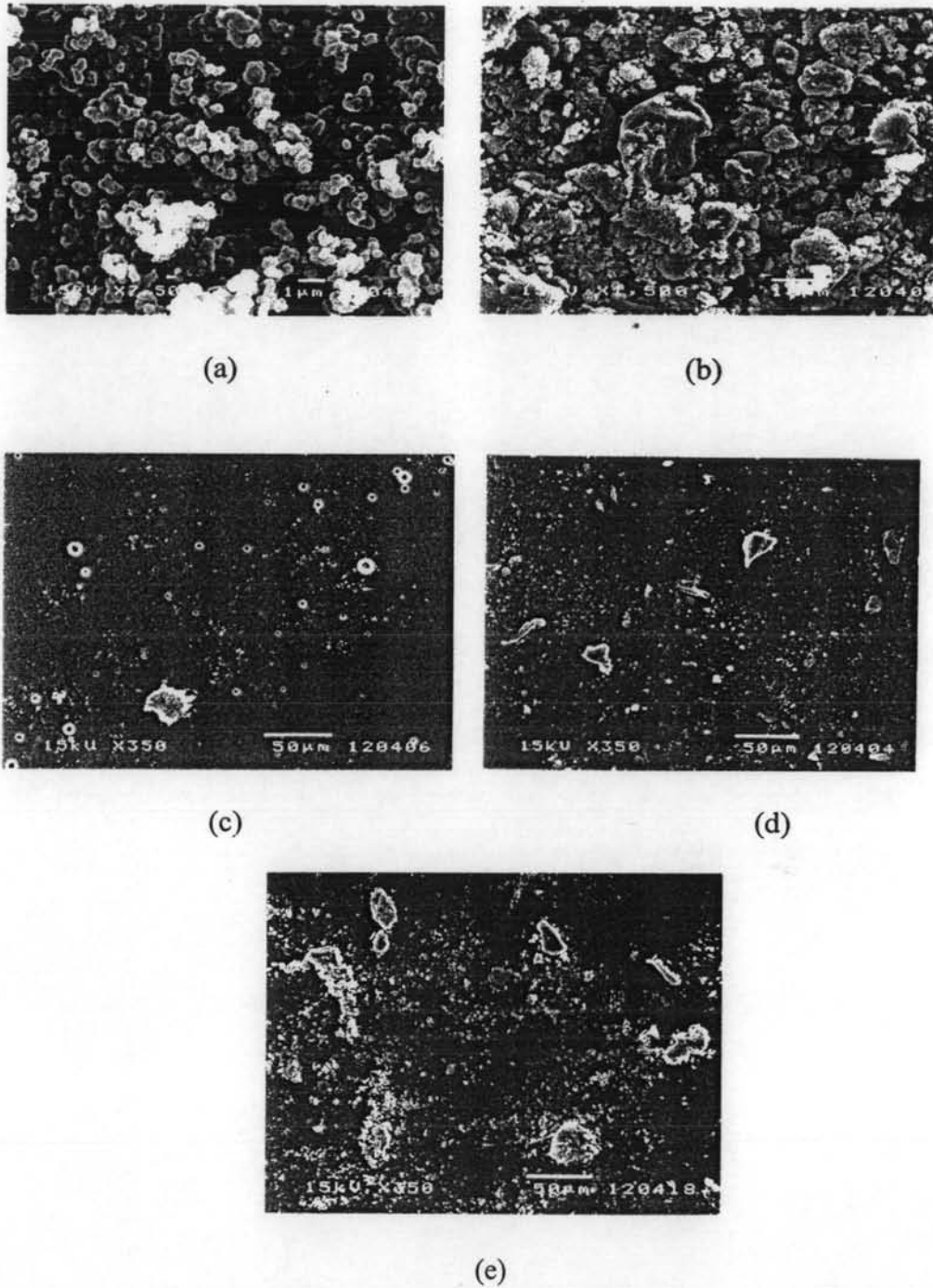
$\Delta G'_{2kV}$  is the the storage modulus responses between  $G'_0$  and  $G'_{2kV}$

$\Delta G'_{2kV}/G'_0$  is sensitivity of the storage modulus at 2 kV

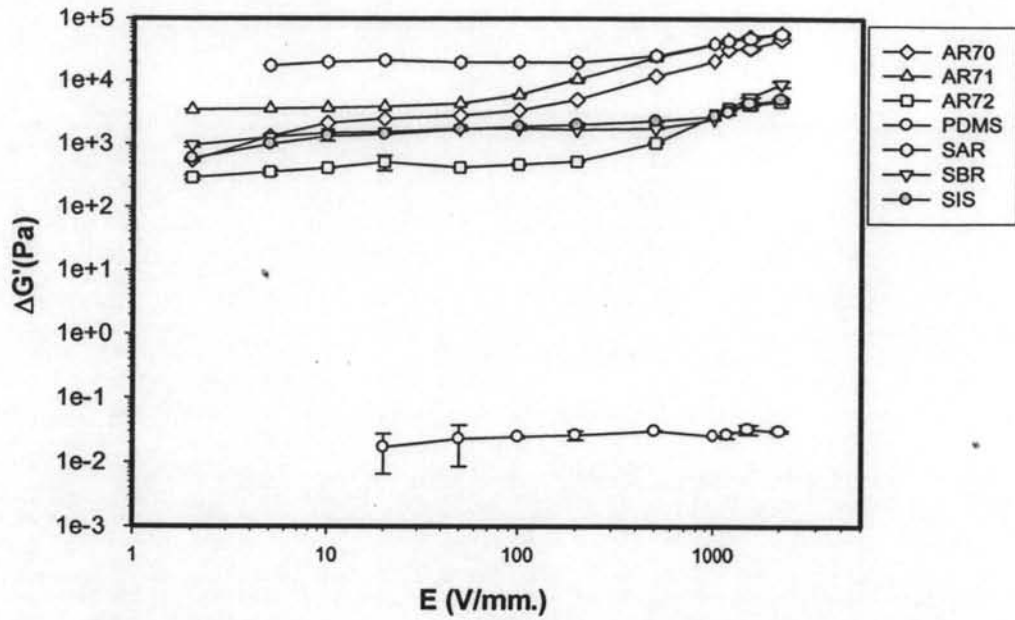
**Table 3** Induction time and recovery time of pure polyacrylate (AR70) and polypyrrole/ polyacrylate (AR70) elastomer blends

Samples	Electric field	induction time	recovery time	$\Delta G'_{ind}$	$\Delta G'_{rec}$
	(kV/mm)	( $\tau_{ind}$ ) (s)	( $\tau_{rec}$ ) (s)	(Pa.s)	(Pa.s)
Pure AR70	1	424	436	485	613
Pure AR70	2	433	466	2,995	2,896
AR70:Ppy_un_4	1	337	207	9,048	3,657
AR70:Ppy_un_4	2	309	138	50,022	47,301

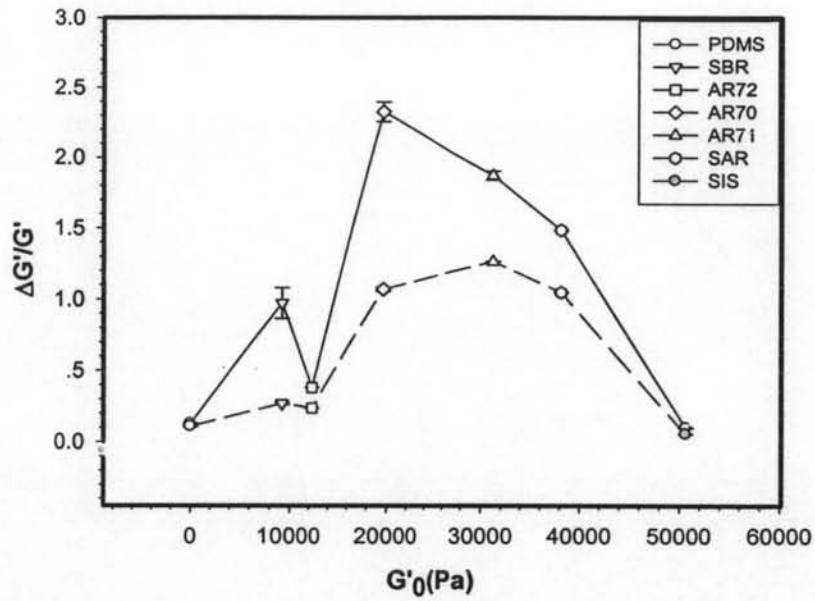




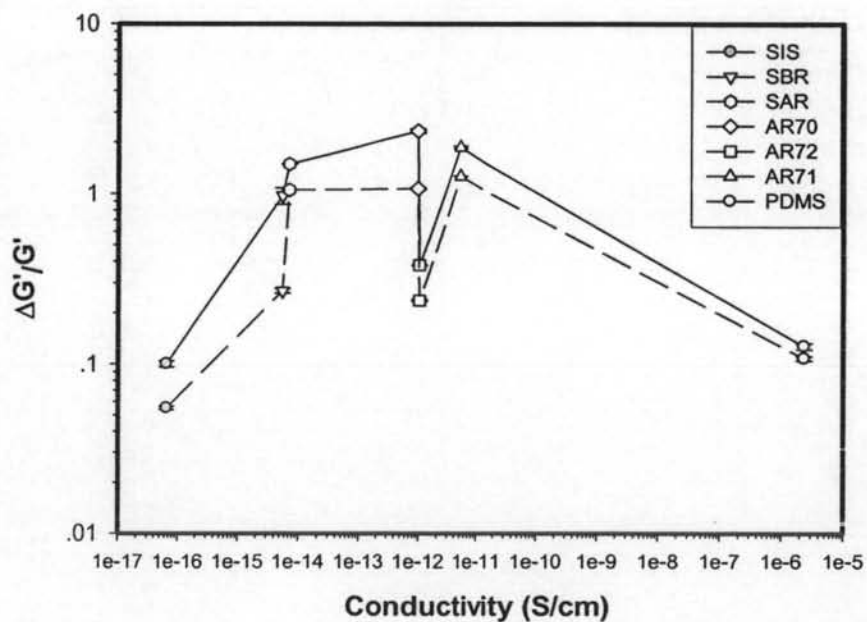
**Figure 1** The morphology of polypyrrole and polypyrrole/polyacrylate blends: a) synthesized undoped polypyrrole at magnification of 7,500; b) synthesized doped polypyrrole at magnification of 1,500; c) AR70:Ppy\_un\_1; d) AR70:Ppy\_un\_3; and e) AR70:Ppy\_un\_5 at magnification of 350.



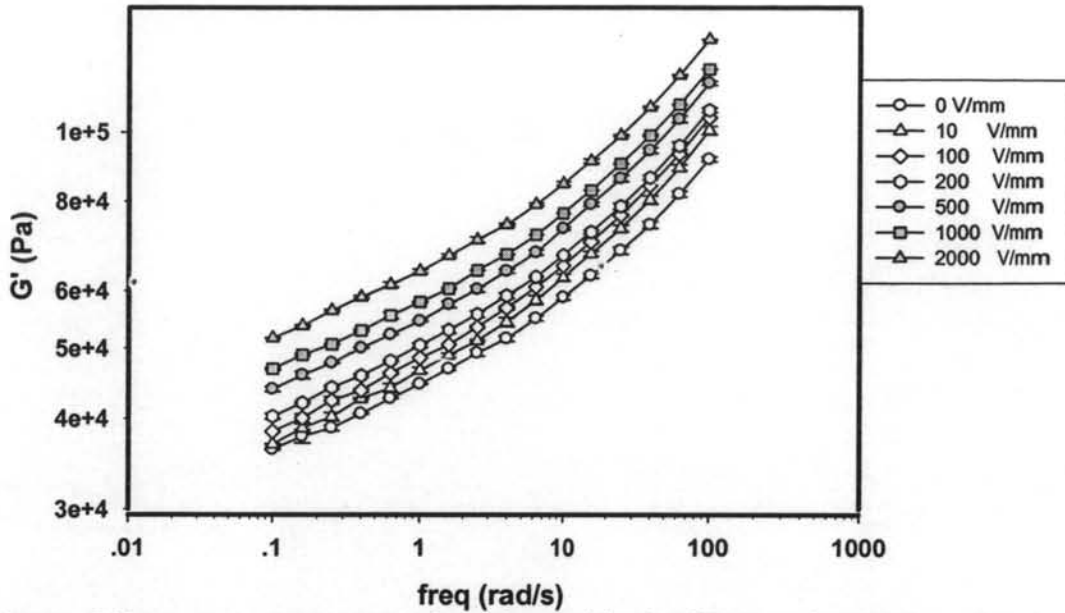
**Figure 2** Effect of matrices on the storage modulus response ( $\Delta G'$ ) vs. electric fields strength: 0 to 2 kV/mm at frequency 1 rad/s, strain 1% (except PDMS strain = 700%), and at 27 °C.



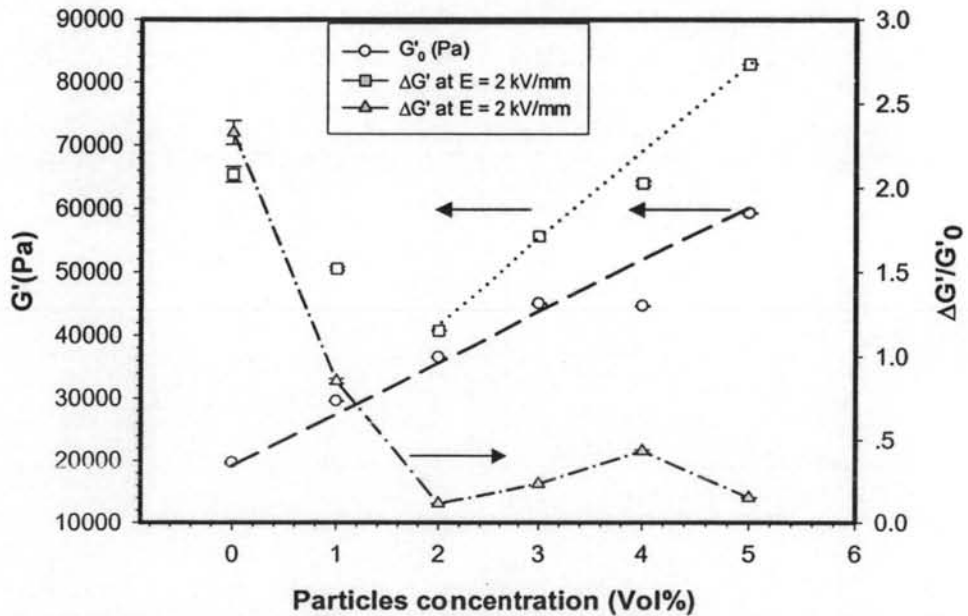
**Figure 3** Sensitivity vs.  $G'_0$ , strain 1.0% (except PDMS was tested at 700%), frequency 1.0 rad/s, and 27 °C at: (a) solid line, 2 kV; (b) dashed line, 1 kV.



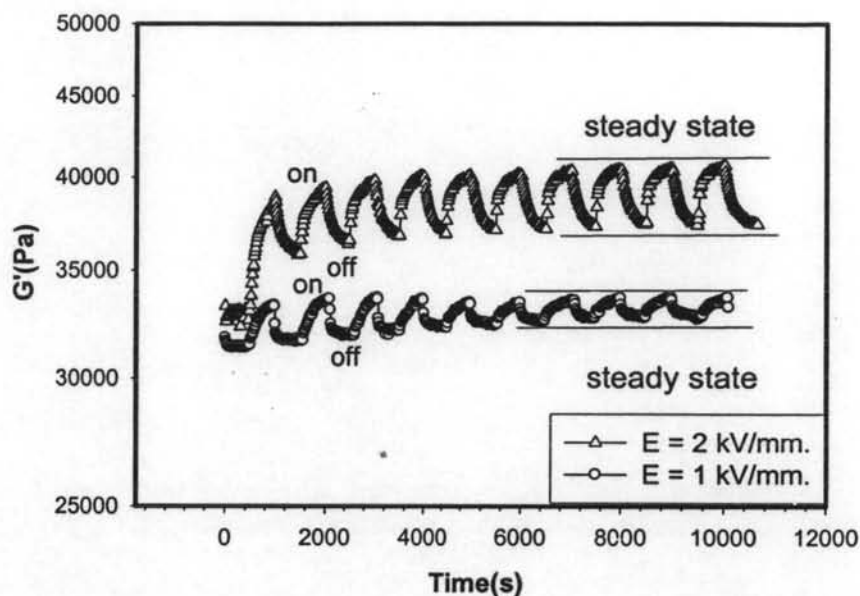
**Figure 4** Sensitivity vs. conductivity, strain 1.0% (except PDMS was tested at 700%), frequency 1.0 rad/s, and 27°C at: (a) solid line, 2 kV; (b) dashed line, 1 kV.



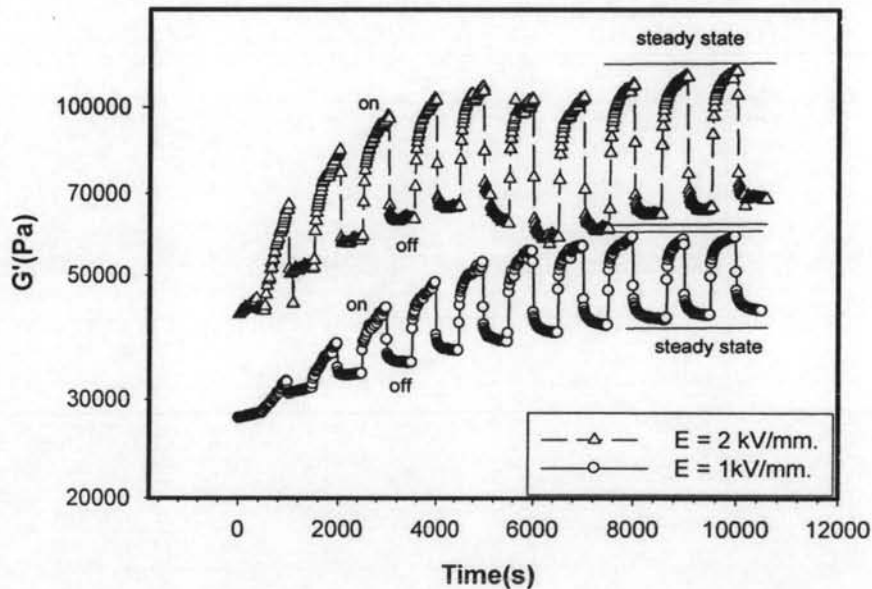
**Figure 5** Frequency sweep test of a polymer blend of Ppy and AR70 at 4% v/v of Ppy (AR70:Ppy\_un\_4), strain 1%, 27 °C, gap 0.295 mm at various electric field strengths.



**Figure 6** Sensitivity of the polymer blend of Ppy and AR70 ( $G'$  and  $\Delta G'_{2kV}/G'_0$ ), vs. Ppy concentration, at 27°C, frequency 1 rad/s.



**Figure 7** Temporal response of storage modulus ( $G'$ ) of AR70 matrix at electric field strengths of 1 and 2 KV/mm, frequency 1.0 rad/s, strain 1%, and at 27 °C.



**Figure 8** Temporal response of storage modulus ( $G'$ ) of polymer blend between AR70 and undoped Ppy at 4% v/v (AR70:Ppy\_un\_4), at electric field strength of 1 and 2 KV/mm, frequency 1.0 rad/s, strain 1.0%, and at 27 °C.