

CHAPTER V

ELECTRICAL PROPERTIES AND ELECTROMECHANICAL RESPONSES OF ACRYLIC ELASTOMERS AND STYRENE COPOLYMERS: EFFECT OF TEMPERATURE

Abstract

The electrical properties and electromechanical responses of acrylic elastomers and styrene copolymers were investigated towards electroactive applications such as artificial muscle and/or MEMS devices. The effect of temperature, between 300-370 K, on electrical conductivity, dielectric constant, storage and loss moduli (G' and G''), storage modulus responses ($\Delta G'_{2kV mm}$) and the storage modulus sensitivities ($\Delta G'_{2kV mm}/G'_0$) of acrylic elastomers and styrene copolymers were investigated under applied electric field strengths varying from 0 to 2 kV/mm. The acrylic elastomers (AR70, AR71, AR72) possess linearly positive storage modulus responses or sensitivities with increasing temperature and dielectric constant. On the other hand, the styrene copolymers (SAR, SBS, SIS) attain the maximum storage modulus responses or sensitivities at the glass transition temperature of the hard segments.

Keywords: Elastomer, Electromechanical Properties, Dielectric Constant

44

1. Introduction

Dielectric materials have the ability to store a charge; they are widely used in applications such as capacitors and insulator devices [1]. The dielectric constant generally varies with frequency, temperature, and pressure [1-2], due to the distinct polarization mechanisms. A dielectric polymer may be used as an electroactive polymer (EAP), which can respond physically under an applied electric field. One type of EAP is a dielectric elastomer, which can transform electric energy directly into mechanical responses such as strains [3]. Dielectric elastomers have many advantages; they are light weight, have a high degree of mechanical response, and have a fast responding time [4]. Examples of dielectric elastomers are acrylic elastomers [5], silicone elastomers [6], and polyurethane [7].

As an electric field is applied, the repulsion force between like charges on the same electrode generates tensile stresses in the length and width directions. The unlike charges on the opposite electrode generates the attractive force in the thickness direction [8-9]. This is known as the Maxwell stress effect [8]. Pelrine *et al.* (1998) and (2000) [4, 8] studied the behavior of various types of dielectric elastomers, silicone elastomers, polyurethane, fluoroelastomer, and natural rubber, and determined the % strain changes. The variable used to indicate the degree of electromechanical response of a dielectric elastomer is the effective pressure:

$$p = \varepsilon \varepsilon_0 E^2 \quad , \tag{1}$$

where *p* is the effective pressure, ε is the dielectric permittivity of the material, ε_0 is the dielectric constant of free space (8.85 pF/m), and E is the electric field strength (V/m) [4]. From the Maxwell stress equation (1), elastomers which have higher dielectric constants are expected to generate the same pressure at a lower applied voltage [8]. Most polymers used in various actuator devices have dielectric constants between 2 and 10 [10]. Since the dielectric constant depends on both frequency and temperature [1, 10], it appears that we may vary the electromechanical responses by merely changing frequency and operating temperature.

In our work, we investigate the electromechanical responses of various acrylic elastomers, acrylic-styrene copolymers, and tri-block copolymers: Styrene-acrylic

copolymer (SAR), Styrene-isoprene-styrene triblock copolymer (SIS), and Styrenebutadiene elastomer (SBR). We determine the elastomer dielectric constants at various frequencies and temperatures. In particular, the storage modulus response $(\Delta G'_{2kV/mm})$ defined as $G'_{2kV/mm} - G'_{0}$ are shown to be well correlated with the elastomer dielectric constants through varying temperature.

2. Experimental

2.1 Materials

Nipol AR71 ($T_g = 258$ K) and AR72HF (AR72) ($T_g = 245$ K) are acrylic elastomers produced by Nippon Zeon Polymix Advance Co., Ltd. Acrylic elastomer AR7018 (AR70), Styrene-Butadiene rubber latex (UCAR DL849, $T_g = 312$ K), and Styrene-Acrylic-copolymer latex (UCAR DA27, $T_g = 297$ K) are produced and were provided by Dow Chemical Co., Ltd. Styrene-isoprene-styrene triblock copolymer (Kraton D1112P) was obtained from Shell in Thailand Co., Ltd. The chemical structures of all elastomers are shown in Figures 1(a)-(d).

2.2 Preparation of Specimens

All elastomer specimens were fabricated through solution casting. The AR70, SAR, and SBR specimens were formed by water evaporation; SIS D1112P, AR71, and AR72 specimens were dissolved in toluene at 30 % vol/vol. The solutions were cast onto a mold (diameter 25 mm, thickness 1 mm) and the solvent was eliminated under a vacuum atmosphere at 300 K for 72 hours.

2.3 Characterization and Testing

2.3.1 The specific conductivity was measured by a parallel plate fixture attached to a rheometer (Rheometric Scientific, ARES) and connected to a source meter (Keithley, Model 6517A) which supplied a constant voltage and displayed the resultant current. The applied voltage and the resultant current in the linear Ohmic regime were used to calculate the electrical conductivity of the polymers using eq. (2) as follows:

$$\sigma = \frac{1}{\rho} = \frac{l}{AR} = \frac{lI}{AV} \qquad ,$$

where σ is specific conductivity (S/cm), ρ is the specific resistivity (Ω .cm), I is the measured resultant current (A), V is the applied voltage (V), R is the resistivity (Ω), l is the thickness of specimen (cm), and A is the surface area of specimen. The specific electrical conductivity of the polymers was measured at various temperatures between 300 and 370 K.

2.3.2 The dielectric constant values were measured by an LCR meter (HP, model 4284A) connected to the rheometer (Rheometric Scientific, ARES) with a 25 mm parallel plate fixture. The thickness of the specimens is typically 1 mm and the diameter is about 25 mm. The top and bottom sides of the specimens were coated with silver adhesive to improve the electrical contact between the specimens and the electrodes. The measurements were carried at temperatures between 300 and 370 K. AC voltage applied was varied between 1 and 10 V, depending on materials. The dielectric constant at a frequency of 20 Hz will be referred to as the dielectric constant of the materials.

2.3.3 The electromechanical properties were measured by a rheometer (Rheometric Scientific, ARES), fitted with a custom-built copper parallel plate fixture (diameter of 25 mm). DC voltage was applied with a DC power supply (Instek, GFG 8216A), which can deliver an electric field up to 4 kV. A digital multimeter (Tektronix, CDM 250) was used to monitor the 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 strains to measure the G' and G''in the linear viscoelastic regime. The appropriate strain was determined to be 1.0 % for all elastomers studied. The temporal G' response experiments of AR70 and SAR were carried out at 2 kV/mm at T = 300 K. Then frequency sweep tests were carried out to measure G' and G" of each sample as functions of frequency and temperature. The deformation frequency was varied from 0.1 to 100 rad/s. In each measurement, each elastomer was presheared at a low frequency, and then the electric field was applied for 20 minutes to ensure the steady state condition before the G' and G" measurements. The effect of temperature on the dynamic moduli, electrical conductivity, and dielectric constant was studied at various temperatures between 300 and 370 K.

3. Results and Discussion

3.1 The specific conductivity of materials

The specific conductivity values of AR70, AR71, AR72, SAR, SIS D1112P, and SBR at T = 300 K are 1.17×10^{-12} , 4.39×10^{-12} , 2.87×10^{-12} , 1.27×10^{-10} , 2.05×10^{-18} , and 1.47×10^{-15} S/cm, respectively. The specific electrical conductivities of all elastomers at various temperatures are shown in Figure 2. The dependence of the specific electrical conductivity on temperature can be expressed by the Arrhenius equation [11]:

$$\sigma_{dc} = \sigma_0 \exp\left(-\frac{\Delta E}{k_B T}\right) \quad , \tag{3}$$

where σ_{dc} is the specific electrical conductivity, σ_0 is the pre-factor, k_B is the Boltzmann's constant, *T* is the absolute temperature, and ΔE is the activation energy for conductivity. From eq. (1), we calculated the activation energy for conductivity (ΔE) and the pre-factor (σ_0) using the slopes and the y-intercept of Figure 2. The values are tabulated in Table 1. The data show that the activation energy for SAR is the highest (3.03×10^{-1} eV) and that of AR72 is the lowest (8.48×10^{-3} eV). The specific electrical conductivities of AR70, AR71, and SAR increase with increasing absolute temperature according to Eq. (3), a typical behavior for dielectric polymers as thermal energy promotes electrons to jump over the band gap. In the case of AR72, SIS, and SBR, the specific electrical conductivities increase only slightly with increasing absolute temperature.

3.2 The dielectric constants of materials

The dielectric constants of AR70, AR71, AR72, SAR, SIS D1112P, and SBR at T = 300 K, and at frequency = 20 Hz, are 6.21, 6.33, 4.14, 3.95, 2.74, and 2.87, respectively. The dielectric constant vs. frequency of the elastomers at T = 300 K are shown in Figure 3(a). From the Figure 3(a), the dielectric constants of all acrylic elastomers and SAR can be seen to decrease slightly with increasing frequency when f > 40 Hz. For SIS D1112P and SBR, the dielectric constants are frequency independent. From the Maxwell-Wagner model [1], the dependence of the dielectric constant on frequency can be expressed as:

$$\varepsilon' = \varepsilon_{\infty} + \frac{(\varepsilon_s - \varepsilon_{\infty})}{1 + \omega^2 \tau^2} \quad , \tag{4}$$

where ε' is the dielectric constant, ε_{∞} is dielectric constant at infinitely high frequency, ε_s is the dielectric constant at low frequency, ω is frequency, and τ is the relaxation time. From eq. (4), the dielectric constant is expected to be lower at a higher frequency. This is a direct result of the decrease in the interfacial polarization at high frequency [10, 11]. The interfacial polarization is the local accumulation of like charges from drifting through the materials [1]. Interfacial polarization occurs at the interface or boundary between two materials having a difference in dielectricity or conductivity. As the frequency is increased, the time available for the charge carriers to drift is reduced, and consequently the dielectric constant decreases [1]. Polar polymers have strong dipole moments and high dielectric values. Polar polymers such as acrylic elastomers are expected to have higher dielectric values than those of the styrene copolymers since the acrylic elastomers have a polar functional group (the acrylate group) in their structure [1]. Cannon et al. (2002) [12] studied the dielectric constant of a methyl methacrylate and 2-ethlyhexyl acrylate copolymer latex as a function of frequency. The dielectric constant of the acrylic copolymer after 47 days aging (CoME-NP20) at f = 20 Hz, is about 6.14 at room temperature. From our data tabulated in Figure 3(a), AR70 and AR71 have dielectric constants of 6.21 and 6.33, respectively; thus they are nearly the same as that of CoME-NP20 at room temperature. Among the styrene copolymers, SAR has the highest dielectric constant, 3.95, due to the polar group (acrylate group) on the acrylic segment. SBR has a higher dielectric value (2.87) than SIS D1112P (2.74) since the SBR structure is more flexible than SIS D1112P. The styrene copolymers (SBR and SIS) possess frequency independent dielectric constants at all frequencies, since they are non polar elastomers. In the case of non polar polymer, the interfacial polarization is expected to be very weak [1, 11]. Bishai et al. (2002) [13] studied the dielectric spectroscopy of a polymer blend between styrene-butadiene rubber and a polyester short-fiber. The dielectric constant of the polymer blend was independent of frequency below 10^5 Hz and it slightly decreased at a frequency above 10^6 Hz at T = 298 K. The dielectric constant at the lowest frequency (20 Hz) is about 2.8. Choi et al. (2002) [14] studied the dielectric spectroscopy of a natural rubber cispolyisoprene. The cis-polyisoprene has a dielectric constant of about 2.6 at f = 100 Hz and T = 298 K. The dielectric spectroscopy of cis-polyisoprene slightly decreased with increasing frequency. Von Hippel *et al.* (1946) [15] studied the dielectric spectroscopy of a polystyrene. The polystyrene possessed a dielectric constant, independent of frequency, of about 2.55 at f = 100 Hz and T = 298 K. From our data, the SIS D1112P triblock copolymer has a dielectric constant of 2.74 at f = 20 Hz, T = 300K. Thus, our data on the dielectric constants of SBR and SIS are comparable to those in previously reported work. The dielectric constant at frequency = 20 Hz vs. temperature of the elastomers are shown in Figure 3(b). The dielectric constants of the elastomers increase linearly with increasing temperature. The increase in temperature or the thermal energy available leads to the increase in the mobility of molecules. The motion of molecules in turn induces the dipole moments, leading to the increases in the dielectric constants [1, 16].

The dielectric constants vs. frequency of AR70 at various temperatures are shown in Figure 3(c). At the lowest temperature of 300 K, the dielectric constant is nearly independent of frequency. With increasing temperature, the dielectric constant becomes more frequency-dependent. At the low frequency, the dielectric constants of AR70 at various temperatures are clearly different. This implies that AR70 has a significant interfacial polarization dependence at low frequency and the dependence becomes weak at high frequency.

3.3 The cyclic temporal responses of G' of the materials

The cyclic temporal responses of G' of the acrylic elastomers and the styrene copolymers were studied using AR70 and SAR as the samples at E = 2 kV/mm, $\omega = 1$ rad/s, T = 300 K, and %strain = 1.0. The data are shown in Figure 4(a) and 4(b) and tabulated in Table 2. With electric field on, the storage moduli of AR70 and SAR increase monotonically towards maximum values. This is due to the interactions between charges within the elastomers [8]. As the electric field is removed, the interactions between charges almost vanish completely and the storage modulus decreases. With electric field turned on and off periodically during the total experiment period of 10^4 sec, AR70 and SAR become electromechanically reversible systems. AR70 has an induction time (the time in which the storage modulus increases and attains a steady state value) of 405 s and a recovery time (the

time in which the storage modulus decreases and reaches a steady state value) of 239 s in the steady state. The SAR has an induction time of 440 s and a recovery time of 271 s in the steady state. Thus, both AR70 and SAR have comparable induction and recovery times. The small differences may stem from the differences in elastic properties.

3.4 The effect of temperature on the electromechanical properties

The effect of temperature on the electromechanical properties of the elastomers was investigated at electric field strengths of 0 and 2 kV/mm within the temperature range of 300-370 K. Figure 5(a) shows G'_o and G'_{2kVmm} vs. temperature of the acrylic elastomers (AR70, 71, and 72) at $\omega = 1$ rad/s. We can see that the storage modulus increases linearly with temperature, in conformation with the classical network theory [17]:

$$G' = \nu k_B T , \qquad (5)$$

where k_B is Boltzmann's constant, T is the absolute temperature (K), and v is number of strands per unit volume (1/cm³). One strand corresponds to two crosslinks or junction points [17]. The number of strands per unit volume is equal to two times the number of crosslinks per unit volume. As temperature increases, the entropy of the elastomer also increases as strands are vibrating more rigouroulsy, and this generates a stronger retractive force on the network [17]. This leads to an increase in the storage modulus. The storage modulus (G') at 2 kV/mm is higher than that without electric field at any temperatures investigated. This is due to the interactions between induced dipole moments generated, in addition to the Maxwell stress effect (eq. 1) or the force between unlike charges on the opposite electrode created by electric field strength. We may note that the slope of G'o is lower than that of $G'_{2kV/mm}$. Equation (5) implies that v of the material under electric field is effectively larger than that without an electric field. At E = 2 kV/mm, the apparent number of strands per unit volume of AR70 is the highest (2.55 x 10^{20} stands/cm³) whereas AR72 has the lowest value $(3.29 \times 10^{19} \text{ strands/cm}^3)$. At E = 2 kV/mm, the number of crosslinks per unit volume of AR70 and AR72 are 5.10×10^{20} $crosslinks/cm^3$ and 6.58 x 10¹⁹ crosslinks/cm³, respectively, as tabulated in Table 3. Figure 5(b) shows the corresponding $\Delta G'_{2kV/mm}$ and $\Delta G'_{2kV/mm}/G'_o$ vs. temperature

at $\omega = 1$ rad/s. The storage modulus sensitivities and storage modulus responses of AR70 are highest and those of AR72 are the lowest among the acrylic elastomers studied.

Ma et al. (2004) [18] found that the strain responses under electric field of an acrylic elastomer (VHB 4910) sandwiched between silver electrodes came from two effects: the electrostriction and the Maxwell stress effect. The acrylic elastomer generated the highest strain responses under electric field relative to those of other electroactive materials.

Figure 6(a) shows G'_o and G'_{2kV/mm} vs. temperature of SAR. In this case we can see that the storage modulus decreases almost linearly with temperature with or without electric field. The temperature is increased above T_g of SAR (T_g of SAR \approx 297 K), the styrene segments are in a rubbery state accompanied by an increase in the free volume. At higher temperatures, the material changes its behavior from rubbery- like to plastic-like [19]. It is noted that G' at 2 kV/mm is higher than that without electric field at any temperatures, presumably due to the dipole-dipole interactions between the charges (on the acrylate group) under electric field. Figure 6(b) shows that the storage modulus response and the storage modulus sensitivity decrease monotonically with increasing temperature.

Figures 6(c) and 6(e) show G'_o and G'_{2kV/mm} vs. temperature of SIS D1112P and SBR. We can see that the storage modulus increases linearly with temperature up to 340-350 K, and it decreases beyond that. Figures 6(d) and 6(f) show the corresponding storage modulus responses and sensitivities. The responses and sensitivities increase initially and then decrease after attaining maxima at T = 320 K (SIS D1112P, T_g of styrene segment \approx 307 K [19]) and T = 330 K (SBR, T_g = 312 K). At temperatures above T_g, of the hard segment (styrene), the materials change their behavior from the rubbery-like to the plastic–like [19]. This leads to the decreases in the storage modulus. Thus, the maximum G' sensitivities of the tri-block copolymers are attained at T_g of the hard segment.

Sato et al. (1996) [19] studied the rheological properties of SIS-triblock copolymer in n-tetradecane (C14) as a function of frequency and temperature without an electric field. At low temperature (T<T_g of styrene segment), the system behavior

is still rubbery-like. But with increasing temperature ($T>T_g$ of styrene segment), the system behavior becomes plastic and viscous-like.

Table 3 lists the number of strands and the number of crosslinks per unit volume of the triblock elastomers. SIS D1112P has a higher number of strands per unit volume (2.08 x 10^{20} strands/cm³) relative to SBR, which has a lower value (1.05 x 10^{19} strands/cm³), at E = 0 kV/mm.

Under electric field, the effective number of strands per unit volume of SBR apparently increases to 2.03 x 10^9 stands/cm³. This presumably arises from the induced dipole-dipole interaction between the π -bonds of the benzene ring. On the other hand, the effective number of strands per unit volume of SIS D1112P decreases to 1.39 x 10^{20} strands/cm³; this may result from the effect of the dominant increase in the free volume over that of the dipole-dipole interaction.

3.5 The effect of dielectric constant on the electromechanical properties of the elastomers

Pelrine et al. (2000) [8] showed that the electromechanical responses of dielectric elastomers depended on many factors such as dielectric constant, hardness, and elastic properties of the materials. In our work, the effect of dielectric constant on the electromechanical properties of the elastomers was studied as a function of temperature. Figures 7(a) and 7(b) show the storage modulus response ($\Delta G'_{2kV/mm}$) of the elastomers vs. the dielectric constants (ϵ'). In Figure 7(a), the storage modulus responses of the acrylic elastomers (AR70, AR71, and AR72) increase monotonically and almost linearly with increasing dielectric constant. On the other hand, for the styrene copolymers (SAR, SIS D1112P, and SBR), as shown in Figure 7(b), the storage modulus responses increase initially, reach maxima, and then decrease with increasing dielectric constant. We may summarize that the electromechanical responses ($\Delta G'$) of the elastomers may increase or decrease with the dielectric constant depending on the elastomer chemical compositions and temperature.

. . . .

4. Conclusions

In our study, the electromechanical properties of AR70, AR71, AR72, SAR, SIS D1112P, and SBR were investigated by examining the effects of temperature, conductivity, and dielectric constant on the dynamic modulus, G', under the oscillatory shear mode at an electric field strength varying from 0 to 2 kV/mm. The dielectric constants and specific electrical conductivities of the elastomers increase with increasing temperature. In the case of the acrylic elastomers, the storage modulus responses and sensitivities increase linearly with temperature. For the styrene copolymers, the storage modulus responses and sensitivities attain their maximum at the Tg of the hard segment. The storage modulus responses of AR70, AR71, and AR72 are correlated positively with dielectric constant. On the other hand, the storage modulus responses of SAR, SIS D1112P, and SBR may increase and decrease with increasing dielectric constant, depending on whether the operating temperature is below or above the Tg of the hard segment.

5. Acknowledgements

The authors would like to acknowledge the financial support from the following: the Conductive and Electroactive Polymers Research Unit and KFAS both from Chulalongkorn University; the Petroleum Petrochemical and Advanced Materials Consortium; the Thai Royal Government Budget (Fiscal Year 2551); and the Thailand Research Fund, BRG and PhD/0182/2548. The material support from Shell in Thailand Co., Ltd., Nippon Zeon Polymix Advance Co., Ltd., and Dow Chemical Co., Ltd. is gratefully acknowledged.

6. References

- 1. G.G. Raju: Dielectrics in Electric Fields (University of Windsor, Windsor, Ontario, Canada, 2003)
- 2. S.A. Madbouly, A.A. Mansour, N.Y. Abdou: Euro. Polym. J. 43, 1892 (2007)
- 3. M.Wissler, E. Mazza: Sens. Actuator A-Phys. 120, 184 (2005)

- 4.. R. Kornbluh, R. Pelrine, Q. Pei, S. Chiba, J. Joseph: Mat. Sci. Engr. C 11, 89 (2000)
- 5. R. Palakodeti, M.R. Kessler: Mater. Lett. 60, 3437 (2006)
- 6. G. Kofod, P.S. Larsen: Sens. Actuator A-Phys. 122, 273 (2005)
- T. Ueda, T. Kasazaki, N. Kunitake, T. Hirai, J. Kyokane, K. Yoshino: Synt. Metals 85, 1415 (1997)
- 8. R.E. Pelrine, R.D. Kornbluh, J.P. Joseph: Sens. Actuator A-Phys. 64, 77 (1998)
- 9. S. Butkewitsch, J. Scheinbeim: Appl. Surf. Sci. 23, 8277 (2005)
- 10. B.G. Soares, M.E. Leyva, G.M.O. Barra, D. Khastgir: Euro. Polym. J. 42, 676 (2006)
- 11. C.C. Ku, and R. Liepins: Electrical Properties of Polymers (Hanser Publisher, New York, USA, 1987)
- 12. L.A. Cannon, R.A. Pehtrick: Polymer 43, 1249 (2002)
- 13. A.M. Bishai, A.M. Ghoneim, and A.A.M. Ward: Inter. J. Poly. Mat. 51, 793-812 (2002)
- Y.H. Choi, M. Skliar: Proceeding of American Control Conference, Anchorage, AK May 8-10, 5, 4285-4290 (2002).
- 15. A. von Hippel, L.G. Wesson: Ind. Eng. Chem. 38, 1121 (1946)
- 16. A.J. Riad, M.T. Korayem, T.G. Abdul Malik: Physica B 270, 140- (1999)
- 17. M. Rubinstein, R.H. Colby: Polymer Physics (Oxford University Press Inc., New York, USA, 2003)
- 18. W. Ma, L.E. Cross: Appl. Phys. A. 78, 1201 (2004)
- 19. T. Sato, H. Watanabe, K. Osaki: Macromolecules 29, 6231 (1996)

List of figure captions

- Figure 1 Chemical structures of the elastomers: (a) Acrylic elastomers (AR70, AR71 and AR72); (b) Styrene-acrylic copolymer; (c) Styrene-isoprene-styrene triblock copolymer; and (d) Styrene-butadiene elastomer.
- 2. Figure 2 DC specific conductivity of the elastomers vs. 1000/T.
- 3. Figure 3 (a) Dielectric constants of various elastomers vs. frequencies at T = 300 K; (b) Dielectric constants of all elastomers at various temperatures at f = 20 Hz; (c) dielectric constants of pure AR70 matrices vs. frequencies, at various temperatures, thickness ~ 0.980 mm, applied voltage E = 1 V, with silver coating.
- 4. Figure 4 Temporal response of storage modulus (G') at an electric field strengths 2 kV/mm, $\omega = 1 \text{ rad/s}$, strain 1.0%, and at 300K of (a) AR70 and (b) SAR.
- 5. Figure 5 Effect of temperature on G'($\omega = 1 \text{ rad/s}$) for the acrylic elastomers (AR70, AR71, and AR72) on: (a) the storage modulus (G') vs. temperature at E = 0 and 2 kV/mm; and (b) the storage modulus responses ($\Delta G'_{2kV/mm}$) vs. temperature and the storage modulus sensitivities ($\Delta G'/G'_0$) vs. temperature.
- 6. Figure 6 Effect of temperature on G'(ω = 1 rad/s) of the styrene copolymers: (a) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SAR; (b) the storage modulus responses (ΔG'_{2kV/mm}) vs. temperature and the sensitivities of storage modulus (ΔG'/G'₀) vs. temperature of SAR; (c) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SIS D1112P; (d) the storage modulus responses (ΔG'_{2kV/mm}) vs. temperature and the sensitivities of storage modulus (ΔG'/G'₀) vs. temperature of SIS D1112P; (d) the storage modulus (ΔG'/G'₀) vs. temperature of SIS D1112P; (e) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SBR; and, (f) the storage modulus responses (G'_{2kV/mm}) vs. temperature and the sensitivities of storage modulus responses (G'_{2kV/mm}) vs. temperature of SBR; and, (f) the storage modulus (ΔG'/G'₀) vs. temperature of SBR.
- Figure 7 Storage modulus responses (ΔG'_{2kV/mm} at ω = 1 rad/s) of the elastomers vs. dielectric constant (ε') at frequency = 20 Hz: (a) acrylic elastomers; (b) styrene copolymers.

List of table captions

- **1.** Table 1 Activation energy for conductivity (ΔE) and pre-factor (σ_o) for each elastomers
- 2. Table 2 G' Induction time and recovery times of AR70 and SAR at 300 K, E = 2 kV/mm
- **3.** Table 3 Number of strands and number of crosslinks per unit volume of the elastomers

Table 1 Activation energy for conductivity (ΔE) and pre-factor (σ_o) for each elastomers

Elastomer Types	Activation energy for conductivity (eV).	pre-factor (S/cm)		
AR70	9.18E-02	3.83E-11		
AR71	1.49E-01	1.93E-09		
AR72	8.48E-03	3.19E-12		
SAR	3.03E-01	1.24E-05		
SIS D1112P	1.22E-02	3.65E-18		
SBR	3.75E-02	7.18E-15		

Samples	Electric field	First induction time	Saturated induction time	First recovery time	Saturated recovery time	First ⊿G' _{ind}	Saturated ⊿G' _{ind}	First ⊿G' _{rec}	Saturated ⊿G' _{rec}
	(kV/mm)	(τ_{ind}) (s)	(τ_{ind}) (s)	(τ_{rec}) (s)	(τ_{rec}) (s)	(Pa.s)	(Pa.s)	(Pa.s)	(Pa.s)
AR70	2	418	405	256	239	28,936	6,842	8.655	6,316
SAR	2	756	440	697	271	21,905	8,730	14.613	7,420

Table 2 G' Induction time and recovery times of AR70 and SAR at 300 K, E = 2 kV/mm

 τ_{ind} = the time in which the storage modulus increases and attains a steady state value.

 τ_{rec} = the time in which the storage modulus decreases and reaches a steady state value.



Elastomer Types	v at E = 0 kV/mm (strands.cm ⁻³)	v at E = 2 kV/mm (strands.cm ⁻³)	Number of crosslinks per unit volume at E	Number of crosslinks per unit volume at E	
			= 0 kV/mm	= 2 kV/mm	
			(crosslinks.cm ⁻³)	(crosslinks.cm ⁻³)	
AR70	6.27×10^{19}	2.55×10^{20}	1.25×10^{20}	5.10×10^{20}	
AR71	3.54×10^{19}	9.42×10^{19}	7.08×10^{19}	1.88×10^{20}	
AR72	2.42×10^{19}	3.25×10^{19}	4.84×10^{19}	6.50×10^{19}	
SAR		-	-	-	
SIS D1112P	2.08×10^{20}	1.39×10^{20}	4.16×10^{20}	2.78×10^{20}	
SBR	1.05×10^{19}	2.03×10^{19}	2.10×10^{19}	4.06x10 ¹⁹	
	-				

Table 3 Number of strands and number of crosslinks per unit volume of the elastomers



Figure 1 Chemical structures of the elastomers: (a) Acrylic elastomers (AR70, AR71 and AR72); (b) Styrene-acrylic copolymer; (c) Styrene-isoprene-styrene triblock copolymer; and (d) Styrene-butadiene elastomer.



Figure 2 DC specific conductivity of the elastomers vs. 1000/T.





Figure 3 (a) Dielectric constants of various elastomers vs. frequencies at T = 300K; (b) Dielectric constants of all elastomers at various temperatures at f = 20 Hz; (c) dielectric constants of pure AR70 matrices vs. frequencies, at various temperatures, thickness ~ 0.980 mm, applied voltage E = 1 V, with silver coating.



(a)



Figure 4 Temporal response of storage modulus (G') at an electric field strengths 2 kV/mm, $\omega = 1$ rad/s, strain 1.0%, and at 300K of (a) AR70 and (b) SAR.



Figure 5 Effect of temperature on $G'(\omega = 1 \text{ rad/s})$ for the acrylic elastomers (AR70, AR71, and AR72) on: (a) the storage modulus (G') vs. temperature at E = 0 and 2 kV/mm; and (b) the storage modulus responses ($\Delta G'_{2kV/mm}$) vs. temperature and the storage modulus sensitivities ($\Delta G'/G'_0$) vs. temperature.





SIS D1112 at T = 300 K, G'₀ = 25,615 Pa, conductivity = 2.05×10^{-18} S/cm SIS D1112 at T = 310 K, G'₀ = 28,531 Pa, conductivity = 2.27×10^{-18} S/cm SIS D1112 at T = 320 K, G'₀ = 35,596 Pa, conductivity = 2.66×10^{-18} S/cm SIS D1112 at T = 330 K, G'₀ = 71,013 Pa, conductivity = 2.51×10^{-18} S/cm SIS D1112 at T = 340 K, G'₀ = 103,085 Pa, conductivity = 2.42×10^{-18} S/cm SIS D1112 at T = 350 K, G'₀ = 124,218 Pa, conductivity = 2.30×10^{-18} S/cm SIS D1112 at T = 360 K, G'₀ = 56,922 Pa, conductivity = 2.34×10^{-18} S/cm





Figure 6 Effect of temperature on G'($\omega = 1 \text{ rad/s}$) of the styrene copolymers: (a) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SAR; (b) the storage modulus responses ($\Delta G'_{2kV/mm}$) vs. temperature and the sensitivities of storage modulus ($\Delta G'/G'_0$) vs. temperature of SAR: (c) the storage modulus (G^{*}) at E = 0 and 2 kV/mm vs. temperature of SIS D1112P; (d) the storage modulus responses ($\Delta G'_{2kV/mm}$) vs. temperature and the sensitivities of storage modulus ($\Delta G'/G'_0$) vs. temperature of SIS D1112P; (d) the storage modulus ($\Delta G'/G'_0$) vs. temperature of SIS D1112P; (e) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SIS D1112P; (e) the storage modulus (G') at E = 0 and 2 kV/mm vs. temperature of SBR; and, (f) the storage modulus responses ($G'_{2kV/mm}$) vs. temperature and the sensitivities of storage modulus responses (SBR.





Figure 7 Storage modulus responses ($\Delta G'_{2kV/mm}$ at $\omega = 1$ rad/s) of the elastomers vs. dielectric constant (ϵ') at frequency = 20 Hz: (a) acrylic elastomers; (b) styrene copolymers.