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6. AUTHOR(S)
J. Su, Z. Ma, J.I. Scheinbeim and B.A. Newman

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Polymer Electroprocessing Laboratory
Department of Mechanics and Materials Science
College of Engineering, Rutgers University
P.O. Box 909
Piscataway, NJ 08855-0909

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Polymer Electroprocessing Laboratory
Department of Mechanics and Materials Science
College of Engineering
Rutgers University
Piscataway, NJ 08855-0909

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August 1994

Ferroelectric and Piezoelectric Properties of Nylon 11/ Poly(Vinylidene Fluoride) Bilaminate Films

J. SU, Z. Y. MA, J. I. SCHEINBEIM,* and B. A. NEWMAN

Polymer Electroprocessing Laboratory, College of Engineering, Rutgers-The State University of New Jersey, Piscataway, New Jersey 08855-0909

SYNOPSIS

The ferroelectric and piezoelectric properties of a new class of polymer ferroelectric and piezoelectric materials, nylon 11/polyvinylidene fluoride (PVF$_2$) bilaminate films, prepared by a co-melt-pressing method, is presented. The bilaminate films exhibit typical ferroelectric D-E hysteresis behavior with a remanent polarization, P_r, of about 75 mC/m^2, which is higher than the value of 52 mC/m^2 observed for PVF$_2$ or nylon 11 films measured under the same conditions. The coercive field, E_c, of the bilaminate films is $\sim$ 78 MV/m, which is higher than that of either PVF$_2$ or nylon 11 films. Measurements of the temperature dependence of the piezoelectric strain coefficient, d_{31}, and the piezoelectric stress coefficient, e_{31}, were also carried out. The bilaminate films exhibit a piezoelectric strain coefficient, d_{31}, of 41 pC/N at room temperature, which is significantly higher than the PVF$_2$ films (25 pC/N) and the nylon 11 films (3.1 pC/N). When the temperature is increased to 110°C, d_{31} of the bilaminate films reaches a maximum value of 63 pC/N, more than five times that of PVF$_2$ (11 pC/N) and more than four times that of nylon 11 (14 pC/N) at the same temperature. The piezoelectric stress coefficient, e_{31}, of the bilaminate films shows a value of 109 mC/m^2 at room temperature, almost twice that of the PVF$_2$ films (59 mC/m^2) and about 18 times that of the nylon 11 films (6.2 mC/m^2). Measurement of the temperature dependence of the hydrostatic piezoelectric coefficient, d_h, of the bilaminate films also shows an enhancement with respect to the individual components, PVF$_2$ and nylon 11. © 1995 John Wiley & Sons, Inc.
Keywords: ferroelectricity • piezoelectricity • bilaminate • nylon 11 • PVF$_2$

INTRODUCTION

As the only known class of ferroelectric polymers, polyvinylidene fluoride (PVF$_2$) and its copolymers have been extensively studied since the initial discovery of significant piezoelectric activity by Kawai more than 20 years ago.[1-6] Investigations of the origins of the ferroelectric, piezoelectric, and pyroelectric properties of this class of polymers have been carried out using various methods, leading to an understanding of the phenomena in terms of polar crystal structures, crystal phase transitions, and dipole switching phenomena.[7-9] In a series of recent papers,[10-14] Scheinbeim, Newman, and others in our laboratory have reported the discovery of a new class of ferroelectric polymers: the odd-numbered nylons. After appropriate electroprocessing, films of nylons 11, 9, 7, and 5 can be made to exhibit their ferroelectric nature as shown in measurements of the electric displacement (D) versus electric field (E). These nylons exhibit typical ferroelectric hysteresis behavior. In addition, a relatively large piezoelectric response can also be observed, especially when the temperature is elevated above the glass transition temperature of the nylons. For poled, melt-quenched, and cold-drawn nylon 11 films, which show a glass transition at around 70°C, the piezoelectric strain constant, d_{31}, increases from 1.9 pC/N at room temperature to 14 pC/N at 100°C,

* To whom correspondence should be addressed at Rutgers University Polymer Electroprocessing Laboratory, Department of Mechanics and Materials Science, P.O. Box 909, Piscataway, NJ 08855-0909.

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whereas the piezoelectric stress constant, e_{31}, increases from 6 to 21 mC/m^2. These results indicate that the odd-numbered nylons may be particularly valuable for piezoelectric polymer applications at higher temperatures. For example, nylon 5 shows stable piezoelectric response at 250°C.

Based on our discovery of the second class of ferroelectric and piezoelectric polymers, it occurred to us that we could synthesize a new type of electroactive polymer film by constructing a two-layer composite of PVF_2 and an odd-numbered nylon. Because both components of the bilaminate films, the odd-numbered nylon and PVF_2, are ferroelectric and piezoelectric, we believed that the bilaminate films should also exhibit ferroelectric and piezoelectric behavior.

We also believed that the ferroelectric and piezoelectric properties of each component, in the presence of the other, would add, to give resultant properties for the composite, but wondered what would be the effects of the introduced interface between the two components of a composite system.

We decided to first examine the electroactive properties of a two-layer composite (bilaminate) of PVF_2 and nylon 11. Investigations of the ferroelectric switching and piezoelectric properties of these bilaminate films not only confirmed our beliefs but also produced a material with a significant enhancement in these properties compared to either PVF_2 or nylon 11 films.

EXPERIMENTAL

Bilaminate Film Preparation

The materials used in the preparation of nylon 11/PVF_2 bilaminate films were poly(vinylidene fluoride) powder supplied by Soltex Polymer Corporation (HM 1012) and nylon 11 pellets supplied by Rilsan Co. First, PVF_2 and nylon 11 films were individually produced by melt-pressing at 190°C and 210°C, respectively, followed by quenching into an ice-water bath. The PVF_2 and nylon 11 films were then co-melt-pressed between aluminum foils at 210°C, followed by quenching into an ice-water bath to obtain the nylon 11/PVF_2 bilaminate films. Appropriate spacers were used to control the thickness of the bilaminate films. The thickness ratio of the two components was 1 : 1. The co-melt-pressed-quenched bilaminate films were then uniaxially oriented by drawing to a ratio of 3.5 : 1 at room temperature. The drawn bilaminate films were then clamped at fixed extension for 30 min. The final thicknesses of

the bilaminate films produced were 25–50 μm. The films were examined by both optical microscopy and x-ray diffraction methods. We observed that a well-connected interface clearly existed between the nylon 11 and PVF_2 layers of the bilaminate films and that significant crystal orientation existed. As the films were kept at elevated temperature for only a few minutes and because the two polymers are thermodynamically incompatible, we expect no significant mixing to occur at a molecular level across the interface. The well-connected interface results from the adhesive properties of nylon 11, which is often used as a hot melt adhesive. The molecular chains in the PVF_2 component were highly oriented along the draw direction whereas the molecules of nylon 11 were doubly oriented, with the chains oriented in the draw direction and the hydrogen-bonded sheets parallel to the surface of the films. For purpose of comparison, individual nylon 11 and PVF_2 films were prepared using an identical melt-quenching and cold-drawing process: the draw ratio for PVF_2 was 3.7 : 1 and for nylon 11 it was 3.2 : 1.

Gold electrodes were then evaporated on opposing surfaces of the drawn films. The evaporation process was carried out carefully to protect the films from being overheated or annealed.[13] The shape of the electrodes was rectangular and the area of the electrodes was about 10 mm^2 (2 × 5 mm). The processing techniques used to produce the bilaminate films are illustrated in Figure 1.

Poling Treatments and Measurements

Poling of samples was carried out at room temperature by application of a triangular-shaped electric
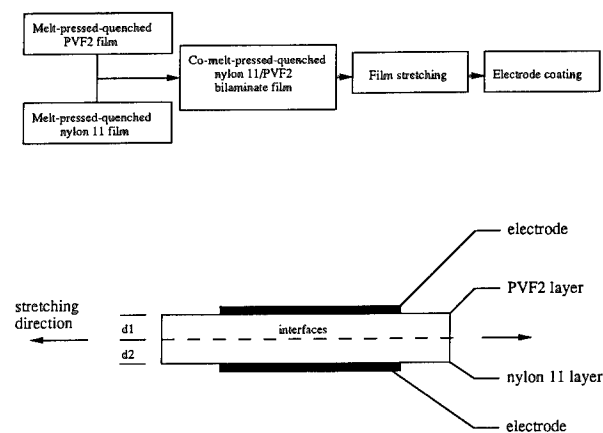

Figure 1. Fabrication of nylon 11/PVF_2 bilaminate ferroelectric film. Block diagram for the procedure and illustration of the sample film.

field waveform. The period of the triangular high voltage waveform was 1000 seconds and the maximum amplitude of the applied field was 160 MV/m. The samples were immersed in a silicone oil bath during the poling treatment to prevent surface conduction and discharge. The current density versus electric field (J-E) characteristics were measured using a picoammeter (Keithley 485) connected in series with the samples during the poling process. The J-E measurements were controlled using a microcomputer. Conductivity effects on the J-E measurement were observed and corrected, using a previously described relationship[13] between the measured current density and the applied electric field:

$$J = aE + b \exp(cE) \tag{1}$$

where E is the applied electric field, J is the measured current density, and a, b, and c are constants, which can be obtained from J versus E and log J versus E plots, respectively.[13] Within the limits of the experimental conditions, the numerical values of a, b, and c were obtained in the range of 10^{-12}, 10^{-5}, and 10^{-8}, respectively.

The temperature dependence of the piezoelectric strain coefficient, d_{31}, piezoelectric stress coefficient, e_{31}, and dielectric constant, ε, were measured using a Rheolograph Solid® (Toyoseiki, Japan). During the measurement, the force was applied in the draw direction, which is the 1 direction, whereas the electric signal was measured in the direction perpendicular to the electrode-coated surface, which is the 3 direction. The measurement temperature ranged from $-150°$C to $+140°$C and the heating rate was $2°$C/min. The measurement frequency was 104 Hz.

Figure 2b.   Corrected curves of current density, J, versus applied electric field, E, for (a) nylon 11/PVF$_2$ bilaminate, (b) nylon 11, and (c) PVF$_2$ films.

The hydrostatic piezoelectric coefficient, d_h, and its temperature dependence were measured from room temperature (25°C) to 140°C using a heating rate of 2°C/min. The measurement frequency was 10 Hz. The d_h measurements were performed using a current amplifier to receive the current change induced by internal pressure change that was excited by a piezoceramic buffer while the sample was placed in a silicone oil-filled, sealed copper chamber.

RESULTS AND DISCUSSION

J-E and D-E Hysteresis Characteristics

Figure 2a shows the as-measured current density, J, versus applied electric field, E, characteristics at room temperature for nylon 11, PVF$_2$, and nylon 11/PVF$_2$ bilaminate films, respectively. The corrected J vs. E curve, shown in Figure 2b, of the nylon 11/PVF$_2$ bilaminate film shows a clear current switching peak as is the case for the individual nylon 11 or PVF$_2$ films. This indicates that the polarization reversal phenomenon observed in the homopolymer films also occurs in the bilaminate film under the applied electric field. From the curves of Figure 2, features of the bilaminate film can be compared with the behavior of individual nylon 11 or PVF$_2$ films: the position of the current density peak (the apparent coercive field) shifts to a higher applied electric field, the symmetry of the peak is lost, and the broadness of the peak is in between those of the nylon 11 and PVF$_2$ films. The coercive field for nylon 11 is 64 MV/m and for PVF$_2$ is 69 MV/m, whereas the coercive field for the bilaminate is 78 MV/m.
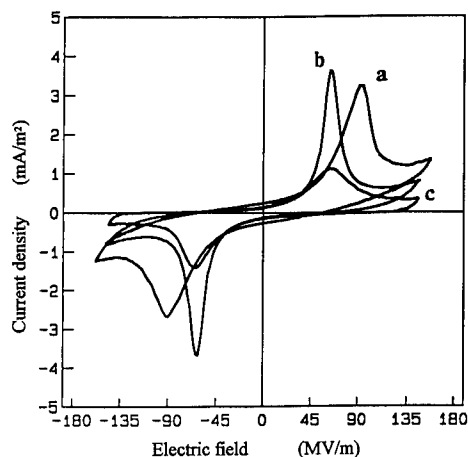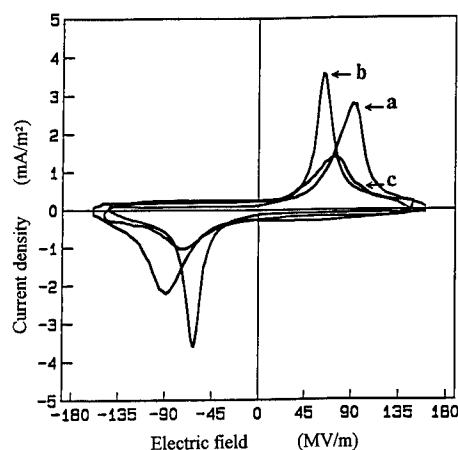

Figure 2a.   As-measured curves of current density, J, versus applied electric field, E, for (a) nylon 11/PVF$_2$ bilaminate, (b) nylon 11, and (c) PVF$_2$ films.

The electric displacement, D, versus applied electric field, E, characteristics for the three samples, nylon 11/PVF_2 bilaminate, nylon 11, and PVF_2, were obtained by integrating the corrected current density curves shown in Figure 2b with respect to time and are shown in Figure 3. It is apparent that the nylon 11/PVF_2 bilaminate film exhibits a *clear D-E* hysteresis loop, characteristic of what is observed in typical ferroelectric materials. From the hysteresis loop, the remanent polarization, P_r, of the bilaminate film can be obtained. The value of P_r for the bilaminate is 75 mC/m^2, which is 44% higher than the 52 mC/m^2 of either nylon 11 or PVF_2 films produced using an identical procedure.

Piezoelectric Properties and Their Temperature Dependence

Figures 4 and 5 show the temperature dependence of the piezoelectric strain coefficient, d_{31}, and the piezoelectric stress coefficient, e_{31}, of the bilaminate and homopolymer films. A very large enhancement of the piezoelectric properties of the nylon 11/PVF_2 bilaminate film is obvious at both room temperature and elevated temperatures. The d_{31} value of 41 pC/N observed for the bilaminate film at room temperature is about 62% higher than the d_{31} value of 25 pC/N observed for PVF_2, and 13 times higher than the d_{31} value of 2.9 pC/N observed for nylon 11, under identical measurement conditions. When the temperature is increased to about 110°C, the d_{31} of the bilaminate films reach their peak value, 63 pC/N, which is more than five times that of the PVF_2 films (11 pC/N) and about seven times that of the nylon 11 films (9 pC/N). A very large enhancement
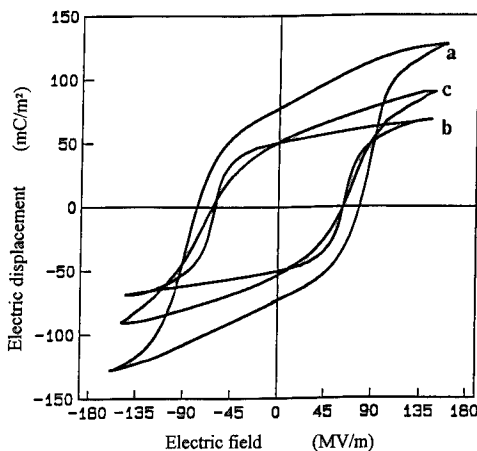

Figure 4. Temperature dependence of the piezoelectric strain coefficient, d_{31}, of (a) nylon 11/PVF_2 bilaminate, (b) PVF_2 films, and (c) nylon 11. The measurements were carried out at a frequency of 104 Hz.

is also observed in the piezoelectric stress coefficient, e_{31}. The value of e_{31} at room temperature is 109 mC/m^2 for the bilaminate, whereas PVF_2 and nylon 11 films show e_{31} values of 59 and 6.2 mC/m^2, respectively. At higher temperature, the e_{31} value of nylon 11 increases from 6.2 mC/m^2 at room temperature to 22 mC/m^2 at $\sim$145°C, and the e_{31} value of PVF_2 decreases from 60 mC/m^2 at 40°C to a value of 3 mC/m^2 at 140°C. A decrease in the value of e_{31} for the nylon 11/PVF_2 bilaminate film is also observed, starting at 40°C, but even when the temperature is increased to 145°C, the value of e_{31} (25 mC/m^2) for the bilaminate film is still higher than that of the nylon 11 film.

The results of the measurement of the temperature dependence of the hydrostatic piezoelectric coefficient, d_h, for nylon 11, PVF_2 and the nylon 11/

Figure 3. Curves of electric displacement, D, versus applied electric field, E, for (a) nylon 11/PVF_2 bilaminate, (b) nylon 11, and (c) PVF_2 films.
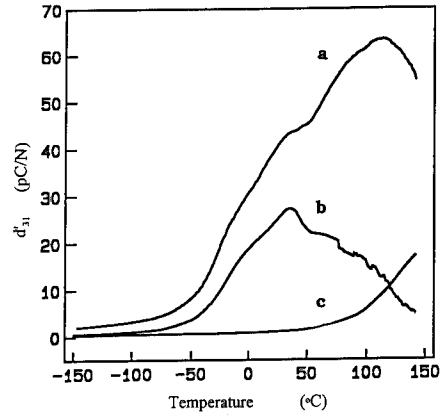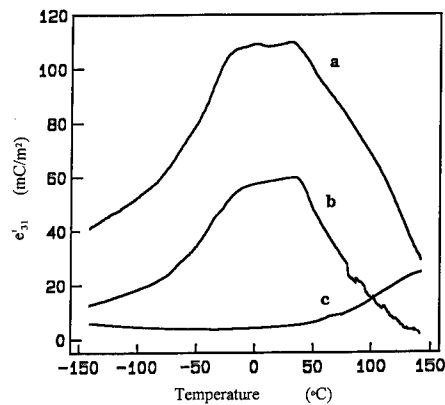

Figure 5. Temperature dependence of the piezoelectric stress coefficient, e_{31}, of (a) nylon 11/PVF_2 bilaminate, (b) PVF_2 films, and (c) nylon 11. The measurements were carried out at a frequency of 104 Hz.
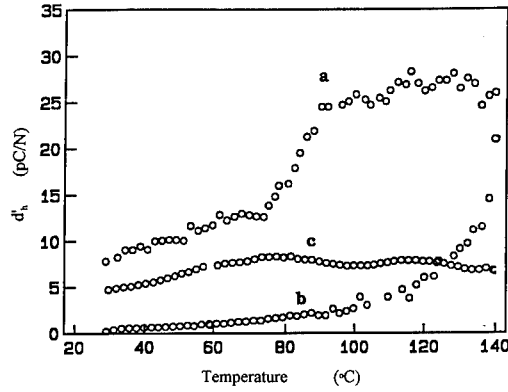
Figure 6. Temperature dependence of the hydrostatic piezoelectric coefficient, d_h, of (a) nylon 11/PVF$_2$ bilaminate, (b) nylon 11, and (c) PVF$_2$ films. The measurements were carried out at a frequency of 10 Hz.

PVF$_2$ bilaminate films are shown in Figure 6. The d_h vs. T curve for the bilaminate shows characteristics similar to those observed in the d_{31} vs. T curve obtained for the bilaminate. At room temperature, the d_h value of 7.6 pC/N for the bilaminate film is obviously higher than those of the PVF$_2$ and nylon 11 films. In addition, the bilaminate exhibits a maximum d_h value of 25 pC/N at 110°C, which is consistent with the high temperature value of the peak position shown in the d_{31} vs. T curve of the bilaminate film shown in Figure 4. A continuous increase in the hydrostatic piezoelectric coefficient, d_h, of the nylon 11 film at higher temperature is observed. It should be noted that the temperature-dependent properties observed for the bilaminate films suggest that in higher temperature applications these films have significant advantages in comparison with PVF$_2$.

The experimental results presented show that the nylon 11/PVF$_2$ bilaminate films are ferroelectric and exhibit clear D-E hysteresis characteristics. In addition, the piezoelectric properties of the bilaminate are greatly enhanced compared with those of either nylon 11 or PVF$_2$ films. Some important measured data, such as remanent polarization, P_r, coercive

field, E_c, piezoelectric strain coefficient, d_{31}, piezoelectric stress coefficient, e_{31}, dielectric constant, ε, mechanical (Young's) modulus in the drawn direction, C, and hydrostatic piezoelectric coefficient, d_h, of the nylon 11/PVF$_2$ bilaminate and nylon 11 and PVF$_2$ homopolymer films are tabulated (see Table I).

In Table I, it should be noticed that the value of the mechanical (Young's) modulus of the bilaminate films, 2.27×10^9 N/m^2, is very close to the value of 2.24×10^9 N/m^2 obtained from a simple calculation based on the relationship for obtaining the modulus of a laminated composite film:

$$C = f_1 C_1 + f_2 C_2 \qquad (2)$$

where C is the Young's modulus of the bilaminate and C_1 and C_2 are the Young's moduli of the component films in the draw direction, where the subscript 1 is for PVF$_2$ and the subscript 2 is for nylon 11. f_1 and f_2 are the volume fractions of the two components, which are $d_1/(d_1 + d_2)$ and $d_2/(d_1 + d_2)$, respectively. However, when the dielectric constant is calculated using this simple model, such a relationship does not exist between the bilaminate and its components. The experimental value of the dielectric constant of the bilaminate (13.8) is far from the calculated value (6.92), if a similar two-layer model is applied to the dielectric response:

$$1/\varepsilon = f_1/\varepsilon_1 + f_2/\varepsilon_2 \qquad (3)$$

where ε is the dielectric constant of the bilaminate, and ε_1 and ε_2 are the dielectric constants of the two components. This is possibly due to a space charge build up at the interface between the two layers of materials having different dielectric constants. The interfacial behavior may be modeled by using a Maxwell-Wagner two-layer capacitor,[15,16] which gives the dielectric constant as:

$$\varepsilon = \varepsilon_\infty + \frac{\Delta\varepsilon}{1 + (\omega\tau)^2}, \qquad (4)$$

Table I. Comparison of Data Taken at 25°C

	E_c MV/m (0.001 Hz)	P_r mC/m^2 (0.001 Hz)	d_{31} pC/N (104 Hz)	e_{31} mC/m^2 (104 Hz)	ε (104 Hz)	C 10^9 N/m^2 (104 Hz)	d_h pC/N (10 Hz)
Bilaminate	78	75	41	109	13.8	2.27	7.6
Nylon 11	64	52	2.8	6.2	4.7	2.14	0.5
PVF$_2$	69	52	25	59	13.1	2.34	4.6

where

$$\varepsilon_\infty = \frac{\varepsilon_1 \varepsilon_2}{f_2 \varepsilon_1 + f_1 \varepsilon_2}, \tag{5}$$

$$\Delta \varepsilon = \frac{f_1 f_2 (\varepsilon_2 \sigma_1 - \varepsilon_1 \sigma_2)^2}{(f_2 \sigma_1 + f_1 \sigma_2)^2 (f_2 \varepsilon_1 + f_1 \varepsilon_2)}, \tag{6}$$

and

$$\tau = \varepsilon_o \frac{f_1 \varepsilon_1 + f_1 \varepsilon_2}{f_2 \sigma_1 + f_1 \sigma_2}. \tag{7}$$

Here, ε_1 (13.1), σ_1 ($1.88 \times 10^{-9}\ \Omega^{-1}\ m^{-1}$), and ε_2 (4.7), σ_2 ($3.81 \times 10^{-9}\ \Omega^{-1}\ m^{-1}$) are the measured dielectric constants and conductivity of PVF_2 and nylon 11, respectively, ω is the measurement frequency, and τ is the relaxation time of the interfacial polarization. When the equations for the Maxwell-Wagner bilayer model, in which the interfacial effect is considered, are applied, the calculated dielectric constant is 7.57, which is higher than the value obtained by the simple model but still not close to the experimental value of 13.8. This appears to indicate that a space charge-dependent interfacial polarization may exist, but the mechanisms of the interfacial effects are not as simple as described in the Maxwell-Wagner model. In the Maxwell-Wagner model, the two constituents are isotropic and they do not possess a remanent polarization. In our real system, both constituents, PVF_2 and nylon 11, are anisotropic and possess a significant remanent polarization. To understand the difference between the model and the experimental results, these distinguishing points should be noted.

If these interfacial effects are significant, they would be important contributors to the observed enhancement in the ferroelectric and piezoelectric properties. Other factors, due to differences in the heterogeneity of these semicrystalline polymers and in their Poisson's ratios, would also affect the ferroelectric and piezoelectric properties of the bilaminate. Further investigations on the ferroelectric and piezoelectric enhancement demonstrated by the nylon 11/PVF_2 bilaminate are being carried out, and other results, including a study of the effects of the relative thickness fraction on these properties, will be reported in the future.

Finally, it should be mentioned that preliminary x-ray diffraction studies of bilaminate films show a field-induced reorientation of crystallites in each component of the bilaminate, which is consistent with observations of homopolymer films poled separately. This is especially important with regard to the nylon component, because in the initial orientation the nylon 11 polymer chains are in the draw direction and the hydrogen-bonded sheets (and therefore the dipoles) are parallel to the film surface. Poling the bilaminate reorients the hydrogen-bonded sheets (and therefore the dipoles) in the nylon 11 compound perpendicular to the film surface, as previously observed in nylon homopolymer films.[12] These results, along with the effects of moisture, will be presented in a separate publication.

CONCLUSIONS

From measurements of electric displacement (D) versus electric field (E) at room temperature, the bilaminate films exhibited typical ferroelectric D-E hysteresis behavior, with a remanent polarization, P_r, of about 75 mC/m^2 and a coercive field, E_c, of 78 MV/m. The piezoelectric strain coefficient, d_{31}, of the bilaminate films showed values of 40.9 pC/N at room temperature and 63 pC/N, as a maximum, at 110°C, whereas the piezoelectric stress coefficient, e_{31}, of the films showed a value of 109 mC/m^2 at room temperature. The enhancements in P_r, d_{31}, and e_{31} are obvious if these results are compared with typical values of remanent polarization and piezoelectric strain and stress coefficients observed for nylon 11 or PVF_2 films, measured under identical conditions. The hydrostatic piezoelectric coefficient, d_h, for the bilaminate films was also measured as a function of temperature from room temperature to 140°C. The results obtained from this experiment also exhibited an enhancement in the bilaminate films' response similar to those observed in the temperature dependence of the piezoelectric strain and stress coefficients.

The basis for the very large enhancement in piezoelectric response for the bilaminate films, compared with the homopolymer films, is being investigated at the present time. However, it is instructive to consider the bilaminate films as a simple two-layer capacitor and to carry out some simple modeling of the mechanical and dielectric properties of these films. Further investigations on the mechanisms of the ferroelectric and piezoelectric enhancement demonstrated by the nylon 11/PVF_2 bilaminate are being carried out in our laboratory at this time, and the results will be reported in a future publication.

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