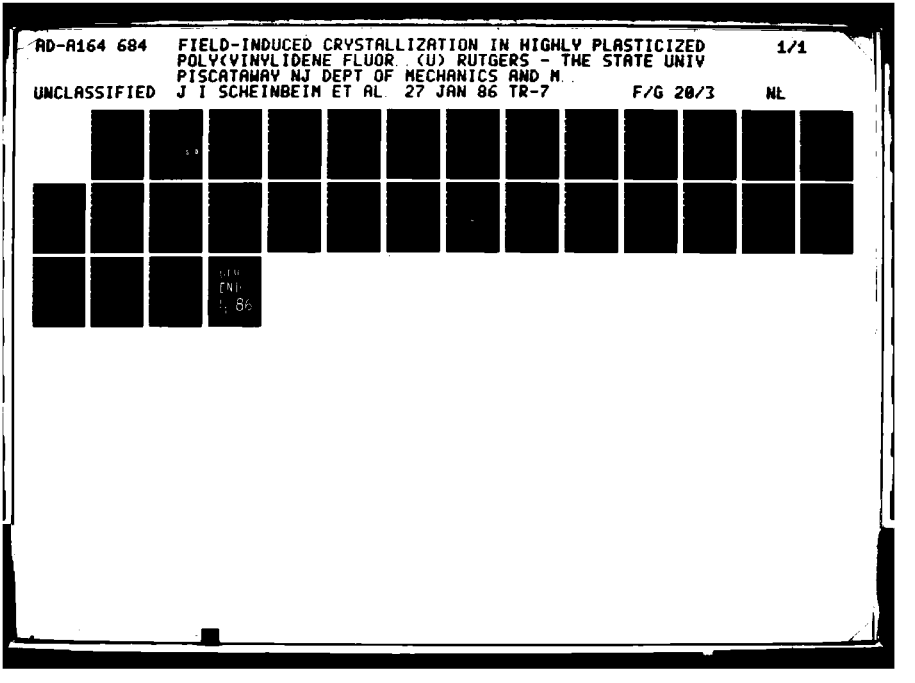

AD-A164 684 FIELD-INDUCED CRYSTALLIZATION IN HIGHLY PLASTICIZED POLY(VINYLIDENE FLUOR..(U) RUTGERS - THE STATE UNIV PISCATAWAY NJ DEPT OF MECHANICS AND M.. 1/1

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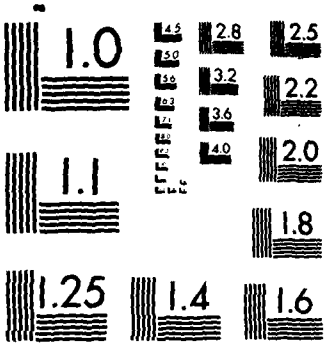

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Poly(vinylidene fluoride), pyroelectric, piezoelectric

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Highly plasticized films of poly(vinylidene fluoride) (PVF_2) containing approximately 18% by weight of tricresyl phosphate (TCP) and with initial crystallinites (phase II) of approximately 15% were poled under high vacuum. The poling temperatures were 75°C and 90°C and the poling field was 250 KV/cm. The poling time was varied from 15 minutes to 240 minutes. Weight measurements and X-ray diffraction data of poled films showed that with increasing poling time, (up to 4 hrs) that plasticizer content decreased (to a minimum of 11%) and the

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Technical Report No. 7

FIELD-INDUCED CRYSTALLIZATION IN HIGHLY PLASTICIZED POLY(VINYLIDENE FLUORIDE) FILMS

by

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Macromolecules

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INTRODUCTION

Previous studies [1, 2, 3] have shown that when phase II PVF_2 films are subjected to poling fields, field-induced crystal transitions occur by way of several stages. At relatively low fields, the non polar phase II converts to polar phase II (phase IV) and this is reflected in an increase in piezoelectric and pyroelectric response. At higher fields, phase I is formed. Furthermore, if the films are initially unoriented the threshold fields for transitions depend on the initial orientation of the crystals [4]. Preliminary studies in our laboratories have shown that addition of plasticizer to the films before poling has a strong influence on the piezoelectric and pyroelectric properties [5]. Both d_{31} and e_{31} were higher for plasticized films compared to unplasticized films poled under identical conditions. We have also shown that crystal phase transitions in the plasticized PVF_2 films take place at lower poling fields as compared to the unplasticized films under identical poling conditions [6]. At higher poling fields, the amount of phase I formed is also much greater and more highly oriented in the case of plasticized films.

An early study by Scheinbeim et al. [7], found that the effects of field on isothermal crystallization from the melt were limited, owing to the small fields which could be applied; the dielectric strength at high temperatures being very low. Recently, Stein et al. [8] have stated that, on the basis of experimental studies of isothermal crystallization of PVF_2 cooled from the melt in an electric field, the effect of the field is to lower the free energy of nucleation of phase III. They showed that at certain undercoolings and applied fields, crystallization in the polar phase III crystalline form is favored, and that the phase III spherulites produced exhibit an angularly

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dependent radial growth rate. An Infrared spectroscopic study of the crysallization behavior of PVF_2 from the melt by Hsu et al. [9], also showed that more phase III occurs in samples crystallized in the presence of an electric field.

In the case of highly plasticized PVF_2 films with low initial crystallinity, it should be possible to crystallize at lower temperatures and hence at much higher fields than those which can be applied to the melt. If we start with highly plasticized PVF_2 and apply an electric field as plasticizer is removed, then the field should bias the cold crystallization process. For the phase II TGTG' conformation, the dipole moment normal to the chain axis is 4.0×10^{-28}Ccm, while that along the chain is 3.4×10^{-28}Ccm, while for the all-trans phase I conformation, the dipole moment is 7.0×10^{-28}Ccm and lies essentially normal to the chain direction [10]. The potential energy associated with a TGTG' conformation is ~-23 KCal/mole while for an all-trans conformation, the potential energy is ~ -17 KCal/mole [11, 12]. This large difference in conformational energy implies that the presence of an electric field in the amorphous regions should have little effect on biasing chain conformations towards those found in the polar crystal forms, however, if we compare the total crystal energy of the phase I and phase II crystals obtained using the crystal structures determined by Hasegawa [13], and Doll and Lando [14], and calculated by Hopfinger [11], Bachmann and Lando [15], the energy values are very close. So in the presence of an electric field, even a small biasing effect arising from the interaction energy might result in oriented growth of the phase I form, as it would then correspond to the lowest free energy state, depending on the magnitude of the local electric field. Based on this idea, we decided to conduct a series of experiments

where heavily plasticized PVF_2 films with very low initial crystallinities are subjected to an electric field while plasticizer is allowed to evaporate and crystallization occurs.

EXPERIMENTAL

A. Sample Preparation

Kureha capacitor grade poly(vinylidene fluoride) film was dissolved in plasticizer (Tricresyl phosphate) at 185°C to obtain solution cast films with very high plasticizer content (≈ 70%) and with no crystallinity as detected by wide-angle diffractometer scans (reflection mode). These films were very difficult to handle due to the high level of plasticizer. The solution cast films were then placed in a vacuum oven at 100°C for 10 hrs. and subsequently melt pressed at 200°C between aluminum foils and then quenched in ice water. PVF_2 films containing approximately 19% by weight of tricresyl phosphate and with initial (phase II) crystallinities of approximately 13% were obtained. The film thickness was measured to be ≈ 25 μm.

B. Poling

Plasticized films were placed in the poling apparatus between two polished copper plates, connected to a high voltage dc supply, and then placed under high vacuum (~ 10^{-6} torr). Samples were poled at 250 KV/cm at 75°C and 90°C. The poling time was varied from 15 minutes to 4 hours. At 90°C, however, the samples could not be poled longer than 2 hours, due to the excessive occurrence of dielectric breakdown. The film surface was coated with silver paint to provide sample electrodes. Figure 1 shows a schematic diagram of the poling apparatus.

C. Electrical and mechanical measurements

The piezoelectric strain constant, d_{31}, piezoelectric stress constant, e_{31}, dielectric constant, and modulus were measured at 3 Hz. The silver paint was first removed from the film surfaces which were then coated with a thin layer of soft silver paste to serve as electrodes for the electrical and mechanical measurements. The pyroelectric coefficient P_y, was measured by placing a sample between two copper plates connected to an electrometer. The sample was then placed in a temperature-controlled chamber. Currents were measured for both heating and cooling cycles at a constant controlled rate of 3°C/min. The pyroelectric coefficient at room temperature was obtained using,

$$P_y = (1/A)\ [(i^+ - i^-) / 2\beta]$$

Where i^+ and i^- are the currents at room temperature during heating and cooling cycles respectively, A is the electrode area and β is the heating and cooling rate in °C/sec. By taking the difference in the measured currents, i^+ and i^-, the small but significant contribution of the background current is removed.

D. X-ray studies

A measure of the amount of phase I material produced by field induced crystallization of highly plasticized PVF_2 films was obtained from wide-angle diffractometer scans, using CuKα radiation. A measure of the total crystallinity (phase I + phase II) was determined from the reflection mode diffractometer scans by taking the ratio of the area under the phase I and phase II peaks to the area under all the crystalline diffraction peaks and the amorphous scattering. The assignment of diffraction peaks to the phase I or phase II

crystal forms was based on previous crystallographic studies [14, 16]. A measure of the amorphous contribution to the diffracted intensity was determined by subtracting the crystalline and base line intensities from the total diffracted intensity, assuming that the amorphous peak is symmetric. A measure of phase I crystallinity was determined by taking the ratio of the area under the phase I peak to the total area (crystalline + amorphous). Relative phase I crystallinity was calculated by taking the ratio of the area under the phase I peak to the area under all crystalline diffraction peaks (phase I + phase II).

The areas under the phase I "shoulders" [phase 1 (110) reflection] were obtained from the diffractometer data. The shoulder area was determined by first assuming that the phase II (110) reflection was symmetric (which is the case for an all phase II sample). After subtracting the amorphous scattering, the total area under the composite peak (phase II with its phase I shoulder) was measured. The area just under the phase II peak was then measured and subtracted from the area under the composite peak. Each measurement was performed five times. The areas obtained differed by less than $\pm$ 5% in all cases. The numerical average of the five values was used. X-ray photographs were also taken on flat film with the x-ray beam perpendicular to and parallel to the plane of the PVF_2 films both before and after poling. Although none of these measures of crystallinity or relative crystallinity are accurate in any absolute sense, they allow for some basis of comparison for films subjected to similar poling histories.

RESULTS AND DISCUSSION

Figure 2 shows the plasticizer content as a function of time while poling at 75°C and 90°C. As expected, with increased poling temperature, the plasticizer evaporated faster. After a poling time of 2 hours, the amount of plasticizer left in the PVF_2 films is 12.1% at 75°C and 10.5% at 90°C (by weight). It is also observed that the amount of plasticizer evaporated at 90°C after 2 hours is higher than the amount of plasticizer which comes out at 75°C after 4 hours. The diffractometer scans (reflection mode) of the PVF_2 films taken before and after poling at 75°C and 90°C for various poling times are presented in Figure 3 and Figure 4 and show the development of (phase I and phase II) crystallinity with poling time. The unpoled samples had an initial crystallinity of $\simeq$ 13% and after a poling time of 2 hours at 75°C and 90°C, the crystallinity was measured to be $\simeq$ 38% and 43% respectively. It is also observed that, initially, no phase I was present but with time, the phase I shoulder becomes more and more prominent. At a poling time of 2 hours at 75°C, the relative phase I crystallinity was found to peak at ~ 36% and at a poling time of 1 hour at 90°C it peaked at ~ 40%. At longer times, the phase I shoulder diminished in relative intensity indicating that little additional phase I was being formed. Reflection scans for the unpoled films with the same thermal history (1 hour at 75°C and 90°C) as that of the poled sample are shown in Figure 5. It is seen that the films crystallized under identical conditions but with no poling field contain only the non-polar phase II form. Figure 6 shows a representation of the flat film x-ray diffraction pattern obtained with the x-ray beam parallel to the plane of the PVF_2 film after being poled at 90°C for 1 hour. It can be seen that the phase I formed on field-induced crystallization is highly oriented. The (110) phase I reflection appears as an arc centered on the

meridional position. Figure 6 also shows that the intensity of the phase I (110) reflection occurs over an arc of ~ 30° to either side of the meridian, suggesting that phase I crystals within this angular range of orientation with respect to the applied field form on field-induced crystallization. Since the dipole direction of phase I is along the crystallographic b axis, the observed phase I diffracted intensity in the reflection mode diffractometer scans, Figure 3 and Figure 4, should result only from the (110) reflection and not from the (200) reflection.

The phase I (200) and (001) reflections appear as arcs in the equatorial position which indicates the orientation of the phase I crystals as: a-axis and c-axis in or close to the plane of the film and the b-axis (dipole direction) close to normal to the film surface. The diffractometer scans also showed that no phase IV (polar phase II) was formed during poling since no change in the relative intensities of the (100) and (200) phase II reflections was observed [6]. This would support an assumption that phase I forms directly by field induced crystallization and not from a poling field induced phase transformation from phase II or phase IV. The fact that the Debye-Scherrer rings from phase II are complete also supports this assumption [4]. Figure 7 schematically represents a flat-film x-ray photograph taken with the x-ray beam parallel to the plane of a PVF_2 film with the same history as a poled sample but with no poling field applied. It is clearly seen that in the absence of an electric field, no phase I formation takes place and only phase II is formed. The phase II crystallites are unoriented as is clear from Figure 7: all crystal orientations are equally present as the Debye-Scherrer rings are complete.

Figure 8 and Figure 9 show a measure of total crystallinity (phase I + phase II) and a measure of phase I crystallinity produced on poling the highly plasticized films at 75°C and 90°C. After a poling time of 2 hours, the amount of phase I formed at 90°C (17%) is higher than the amount of phase I formed at 75°C (13%). The total crystallinity (phase I + phase II) is approximately 40% after poling as shown in Figure 8. If the phase I crystals are highly oriented, the actual amount of phase I formed will be significantly lower than that determined from the diffractometer scans (reflection mode) as these scans record diffracted intensities only from the crystallographic planes parallel to the film surface. The phase I produced by crystallization during poling is clearly oriented, as it is seen that the most intense reflection for phase I (110) is an arc centered on the meridional position. This is shown in the x-ray photograph represented in Figure 6. Comparing Figure 8 and Figure 9, it can be seen that the total crystallinity (phase I + phase II) and the phase I crystallinity developed faster at a higher poling temperature. This is due to the faster rate of plasticizer evaporation at higher temperature, as shown previously in Figure 2. The total crystallinity (phase I + phase II), after a poling time of 1 hour is 41% and 32% at 90°C and 75°C respectively. A measure of relative phase I crystallinity (ratio of area under phase I peaks to area under phase I + phase II peaks) with poling time is shown in Figure 10. It is seen that after a certain poling time (1 hour at 90°C and 2 hours at 75°C) the percent of phase I relative crystallinity decreases, which indicates that during poling, both phase I and phase II are being formed, but after some time, the formation of the non-polar phase II crystals predominates.

The moduli of the poled films (poled at 75°C and 90°C) are shown in Figure 11. As expected, the modulus increased with loss of plasticizer and increased crystallinity. The increase is of course more rapid for films poled at 90°C compared to those poled at 75°C. The modulus of a film poled at 90°C (for 60 min.) is 1.3 GN/m^2, whereas it is 0.9 GN/m^2 when poled at 75°C for the same poling time. The poling time dependence of the piezoelectric strain constant, d_{31}, stress constant, e_{31}, and pyroelectric coefficient P_y, are shown in Figures 12, 13 and 14 respectively. The piezoelectric and pyroelectric response increased with increasing phase I content. At 90°C, d_{31}, e_{31} and P_y reached their maximum values of $d_{31} \approx 10$ PC/N, $e_{31} = 9.8$ mc/m^2 and $P_y = 17$ $\mu Cm^{-2}{}^{\circ}K^{-1}$ at a 1 hour poling time. At 75°C, the maximum values of $d_{31} = 8.8$ PC/N, $e_{31} = 8.6$ mc/m^2 and $P_y = 15.8$ $\mu Cm^{-2}{}^{\circ}K^{-1}$ were reached after a poling time of 2 hours. The dielectric constants of the films poled at 75°C and 90°C are very close, ≈14.7, and decrease little with increasing poling time as shown in Figure 15. The piezoelectric and pyroelectric coefficients follow the same trend as that of relative phase I crystallinity (see Figure10) which indicates that the relative amount of polar phase present is directly related to the piezoelectric and pyroelectric response [1, 6]. At longer poling times, the modulus increases, as shown in Figure 11, which accounts for the observed decrease in d_{31}. In addition, as the plasticizer content decreases and crystallinity increases, we might expect the increase in the modulus of the film to reduce the poisson's ratio and therefore the piezoelectric stress constant, e_{31} [17, 18]. This would also produce a decrease in the coefficient of thermal expansion of the poled films and hence some decrease in the pyroelectric coefficient as shown in Figure 14.

CONCLUSIONS

In the present study, we observed that poling field-induced crystallization of highly plasticized PVF_2 films (with low initial phase II crystallinity) results in the formation of highly oriented phase I crystals. At a poling field of 250 KV/cm, the amount of phase I formed increased with increasing poling temperature and initially with poling time. At a poling time of 1 hour at 90°C, a measure of relative phase I crystallinity indicates that approximately 45% of the crystalline material was in the phase I form. Since the phase I crystals are highly oriented, the actual amount of phase I is much less than 45%. At longer times, the relative percent of phase I decreased.

This study has shown that the presence of a high electric field during crystallization of PVF_2 from solution can significantly bias the free energy of crystallization towards the formation of highly oriented, polar phase I crystals. However, as crystallinity increased with increasing poling time, this bias decreases and phase II crystallization predominates.

ACKNOWLEDGEMENT

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FIGURE CAPTION

1. Schematic representation of field-induced crystallization of highly plasticized PVF_2 film.
2. Plasticizer content in poled PVF_2 films for various poling conditions.
3. Diffractometer scan (refelction mode) of PVF_2 films poled at 75°C.
4. Diffractometer scan (reflection mode) of PVF_2 films poled at 90°C.
5. Diffractometer scan (reflection mode) of PVF_2 films (unpoled) under similar thermal history as poled sample.
6. Representation of the x-ray diffraction patterns from poled PVF_2 film taken with x-ray beam in the plane of the film (poled at 90°C).
7. Representation of the x-ray diffraction patterns from the unpoled but similar thermal history as poled PVF_2 film with x-ray beam in the plane of the film (at 90°C).
8. A measure of total crystallinity of poled films.
9. A measure of phase I crystallinity produced on poling.
10. A measure of relative % phase I crystallinity produced on poling.
11. Modulus of the PVF_2 films poled at 75°C and 90°C.
12. Piezoelectric strain constant for PVF_2 films poled at 75°C and 90°C.
13. Piezoelectric stress constant for PVF_2 films poled at 75°C and 90°C.
14. Pyroelectric coefficient for PVF_2 films poled at 75°C and 90°C.
15. Dielectric constant for PVF_2 films poled at 75°C and 90°C.

POLING SET-UP

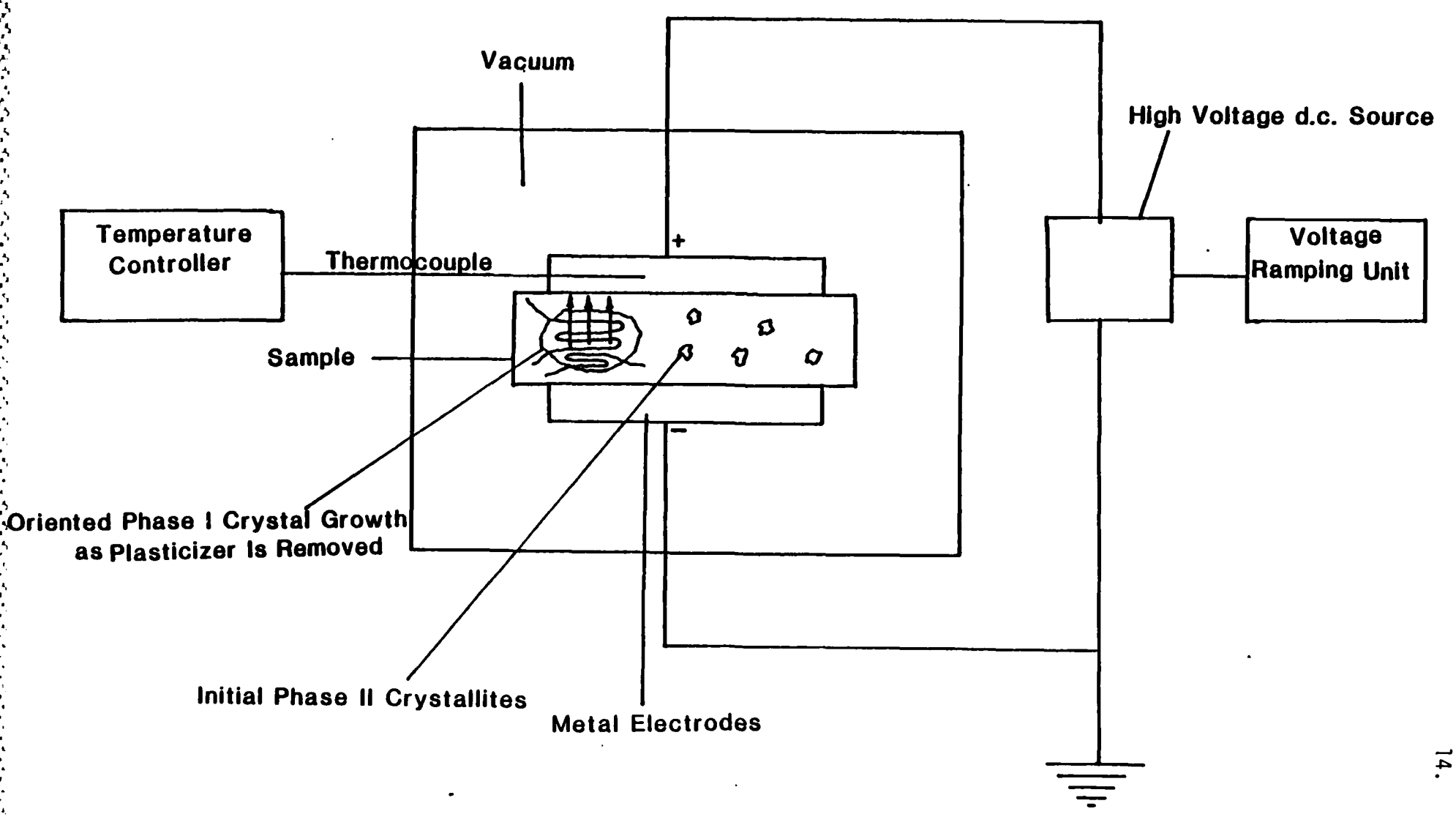

FIGURE 1

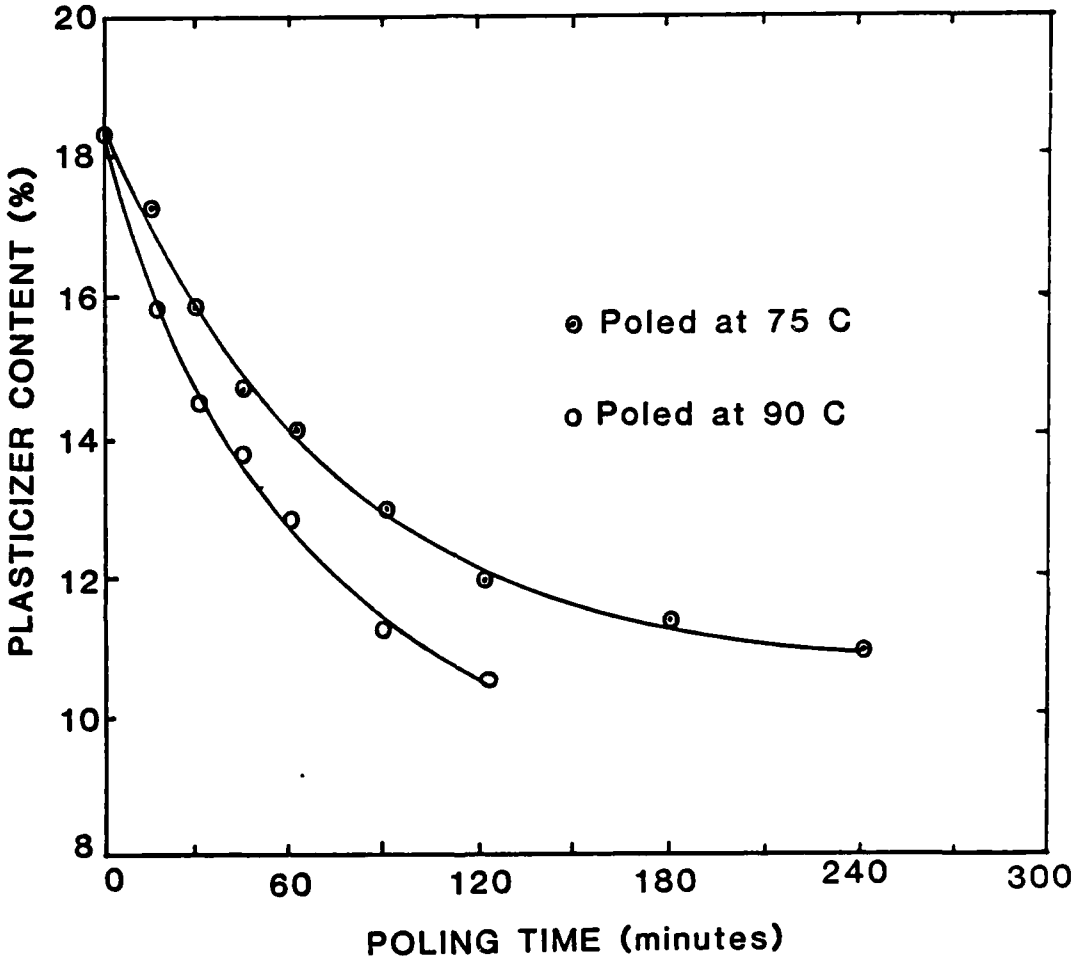
20
18
16
14
12
10
8
PLASTICIZER CONTENT (%)
0
60
120
180
240
300
POLING TIME (minutes)
Poled at 75 C
Poled at 90 C

FIGURE 2

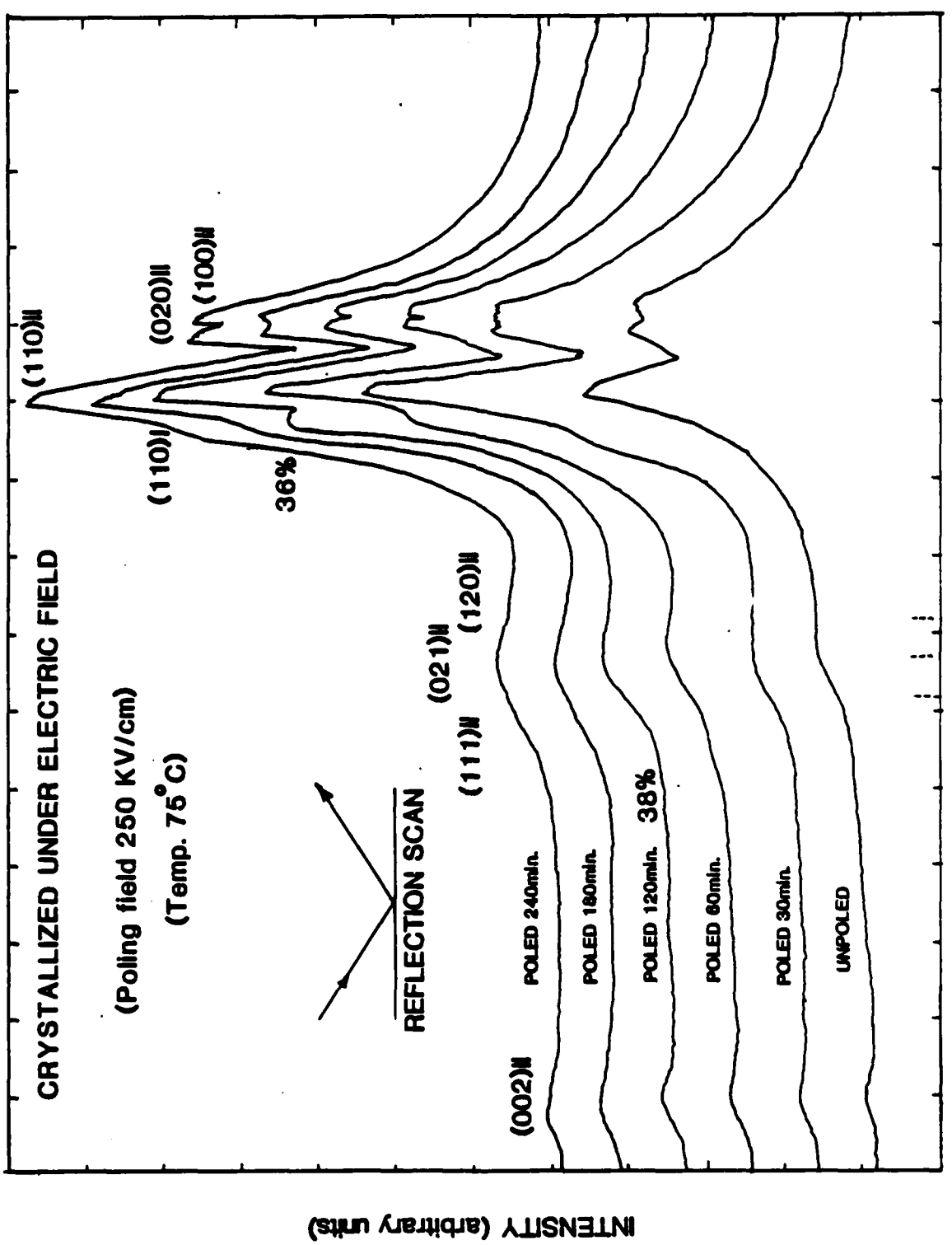
CRYSTALLIZED UNDER ELECTRIC FIELD
(Poling field 250 KV/cm)
(Temp. 75°C)
REFLECTION SCAN
(002)
(111)
(021)
(120)
(110)
(110)
(020)
(100)
36%
38%
POLED 240min.
POLED 180min.
POLED 120min.
POLED 60min.
POLED 30min.
UNPOLED
INTENSITY (arbitrary units)
38
34
30
26
22
18
14
10
DEGREES 2θ

FIGURE 3

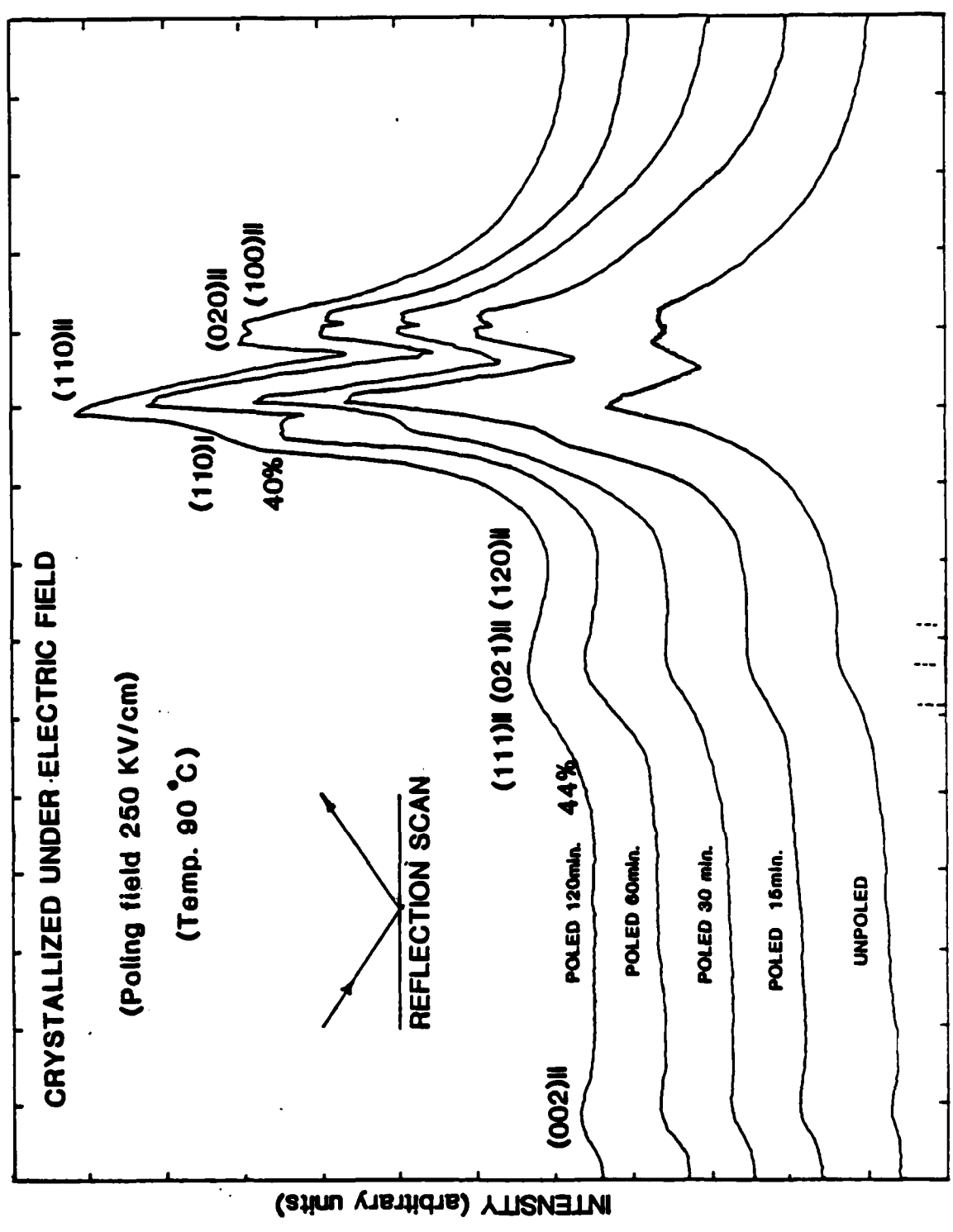
CRYSTALLIZED UNDER ELECTRIC FIELD
(Poling field 250 KV/cm)
(Temp. 90 °C)
REFLECTION SCAN
(110)II
(020)II
(100)II
(110)I
40%
(111)II (021)II (120)II
(002)II
44%
POLED 120min.
POLED 60min.
POLED 30 min.
POLED 15min.
UNPOLED
INTENSITY (arbitrary units)
10
14
18
22
26
30
34
38
DEGREES 2θ

FIGURE 4

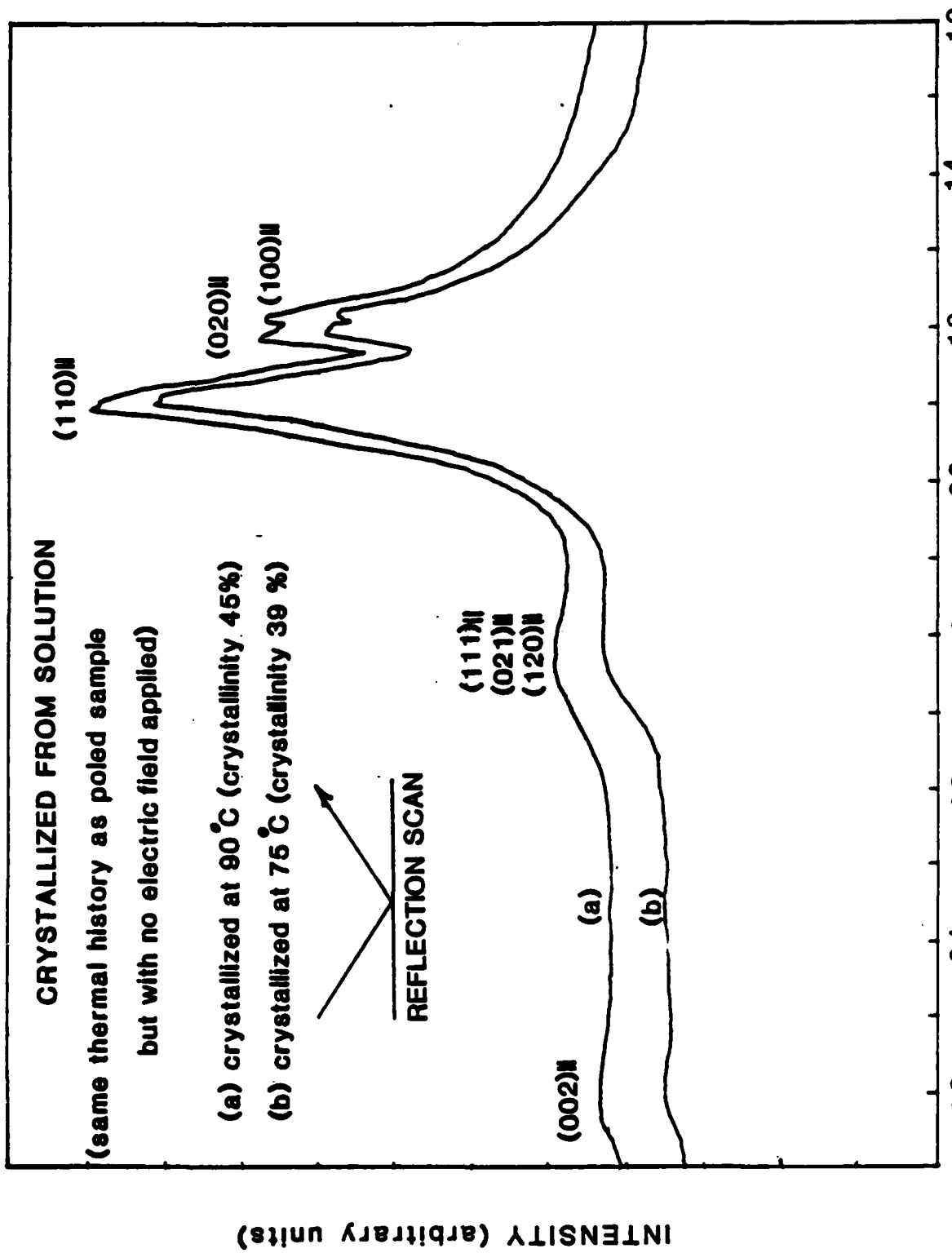
CRYSTALLIZED FROM SOLUTION
(same thermal history as poled sample
but with no electric field applied)
(a) crystallized at 90°C (crystallinity 45%)
(b) crystallized at 75°C (crystallinity 39 %)
REFLECTION SCAN
(110)II
(020)II
(100)II
(111)II
(021)II
(120)II
(002)II
(a)
(b)
38
34
30
26
22
18
14
10
DEGREES 2θ
INTENSITY (arbitrary units)

FIGURE 5

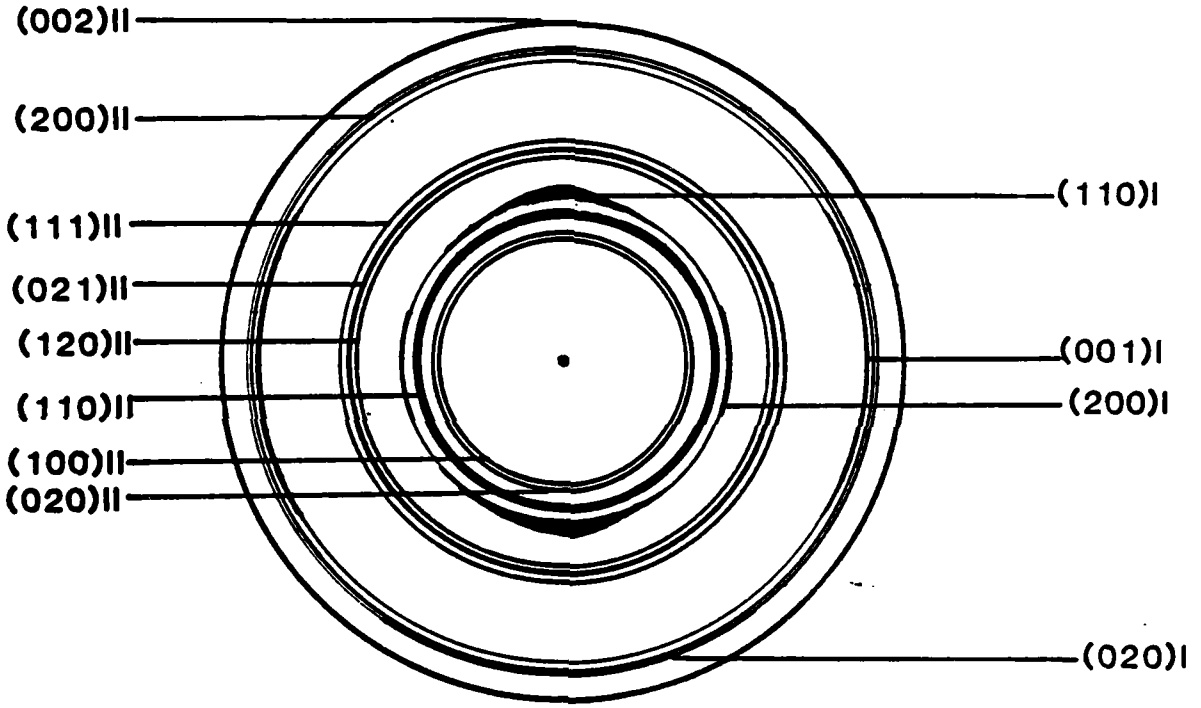
(002)II
(200)II
(111)II
(021)II
(120)II
(110)II
(100)II
(020)II
(110)I
(001)I
(200)I
(020)I

FIGURE 6

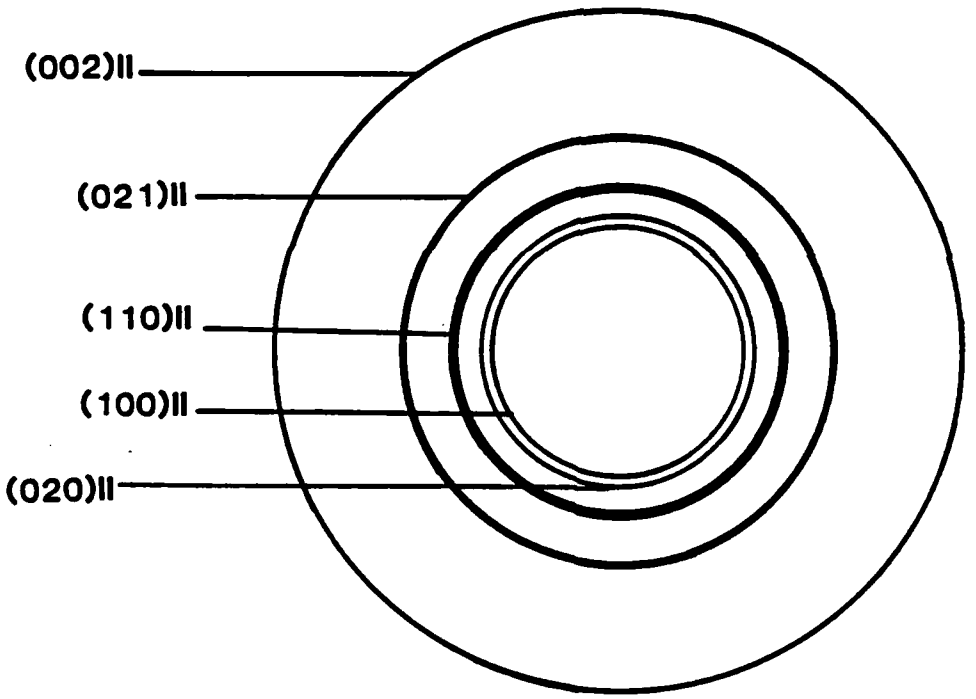

FIGURE 7

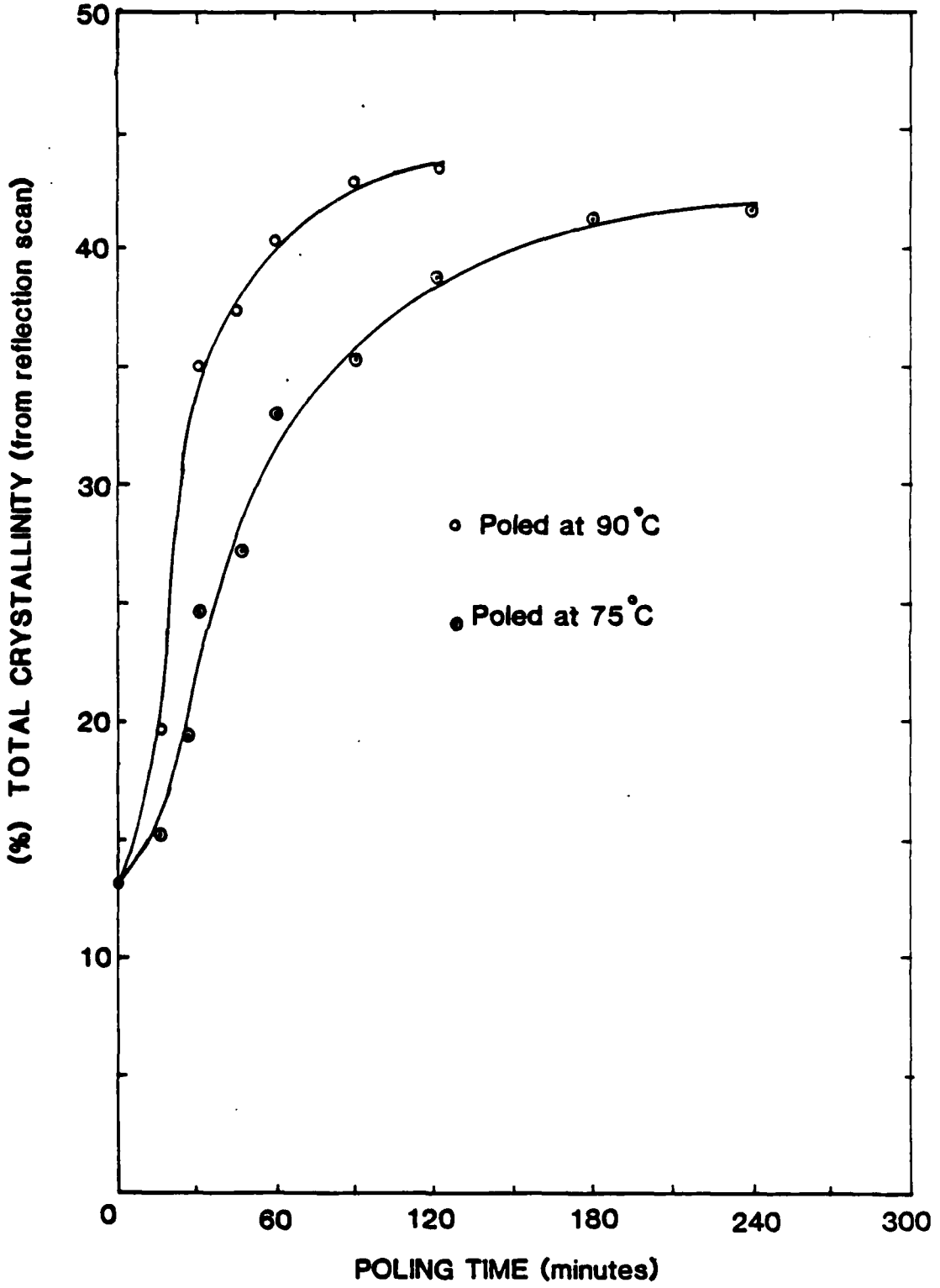
50
40
30
20
10
0
60
120
180
240
300
(%) TOTAL CRYSTALLINITY (from reflection scan)
POLING TIME (minutes)
Poled at 90°C
Poled at 75°C

FIGURE 8

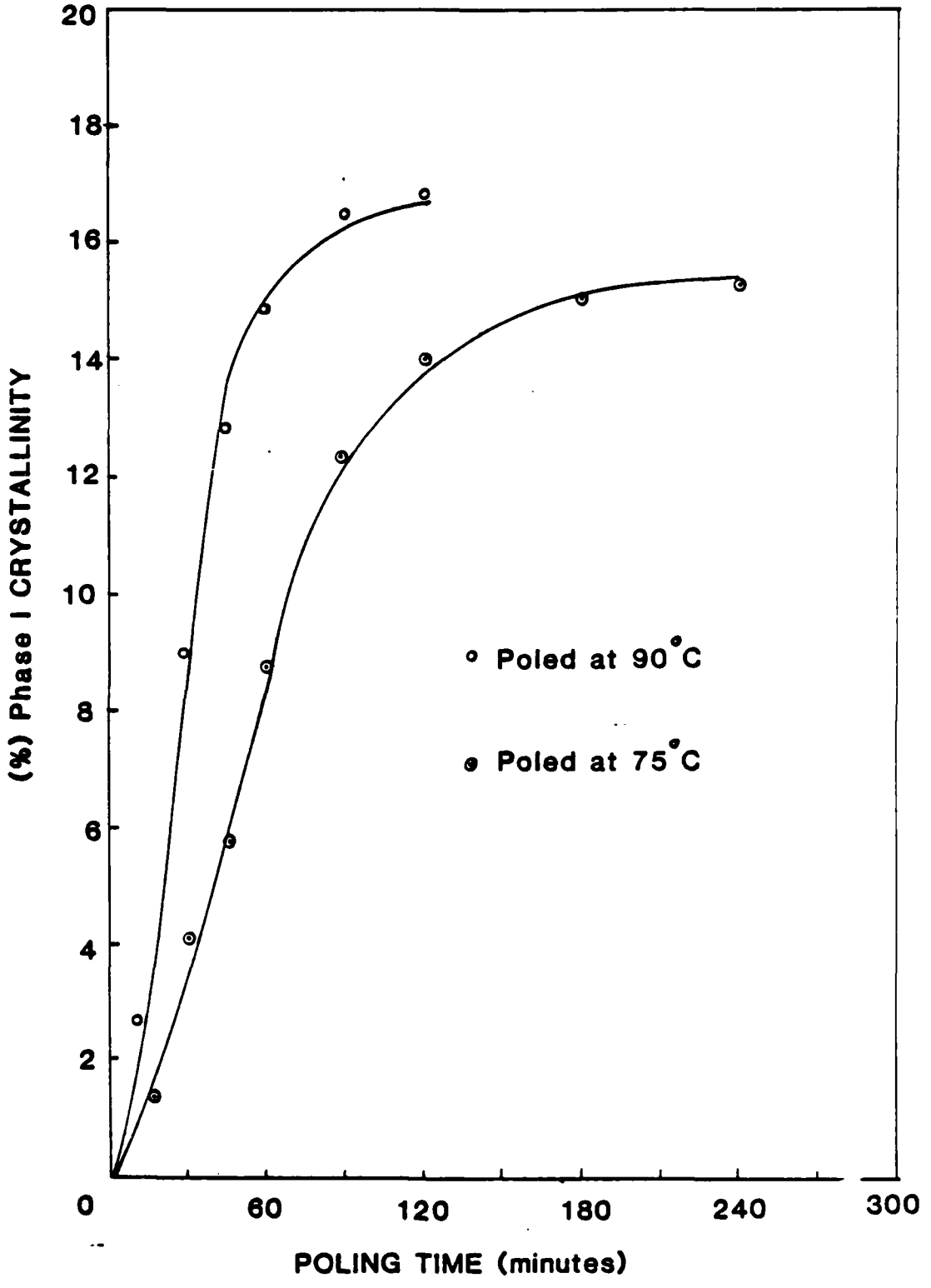
20
18
16
14
12
10
8
6
4
2
0
60
120
180
240
300
(%) Phase I CRYSTALLINITY
POLING TIME (minutes)
Poled at 90°C
Poled at 75°C

FIGURE 9

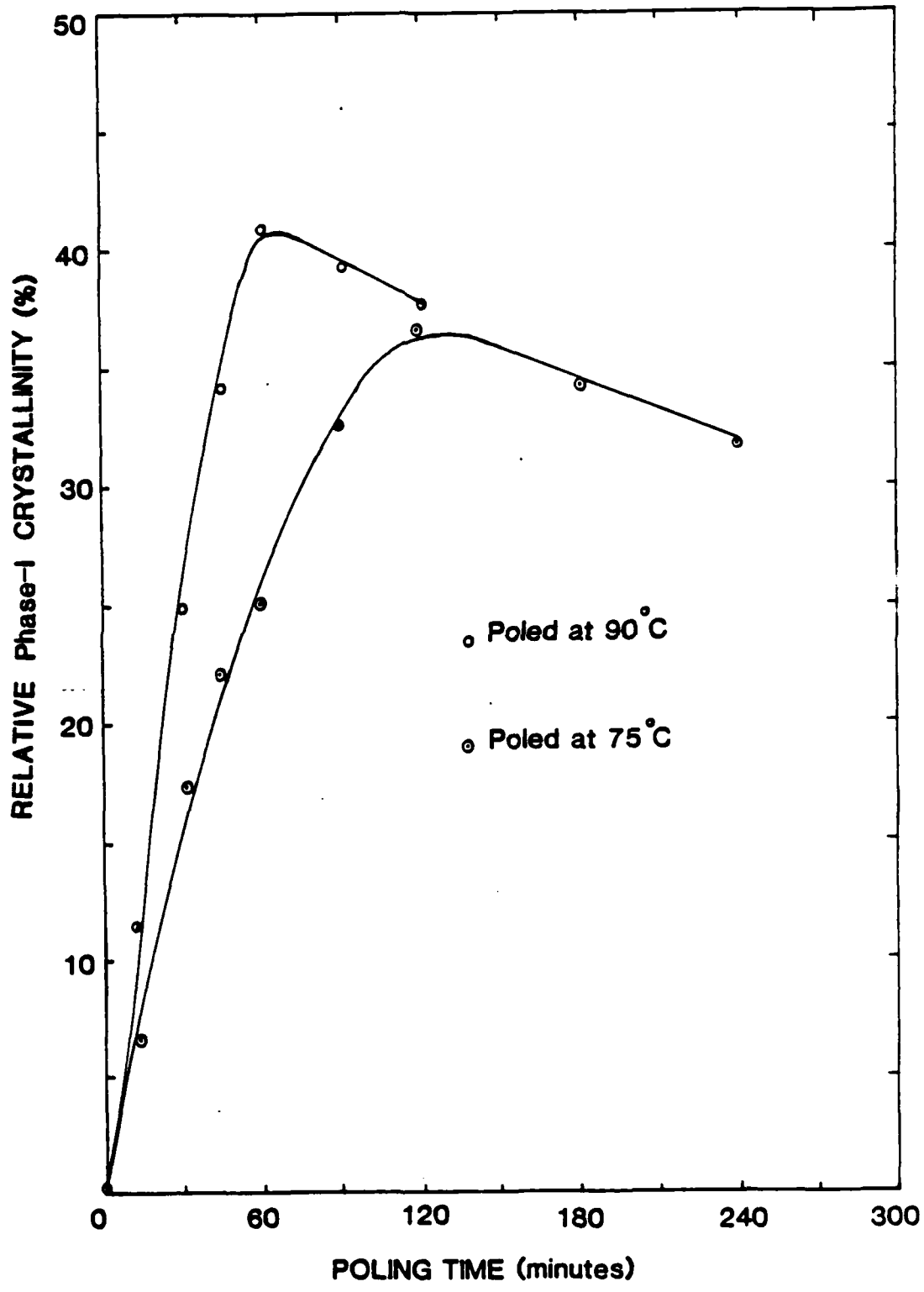
50
40
30
20
10
0
60
120
180
240
300
RELATIVE Phase-I CRYSTALLINITY (%)
POLING TIME (minutes)
Poled at 90°C
Poled at 75°C

FIGURE 10

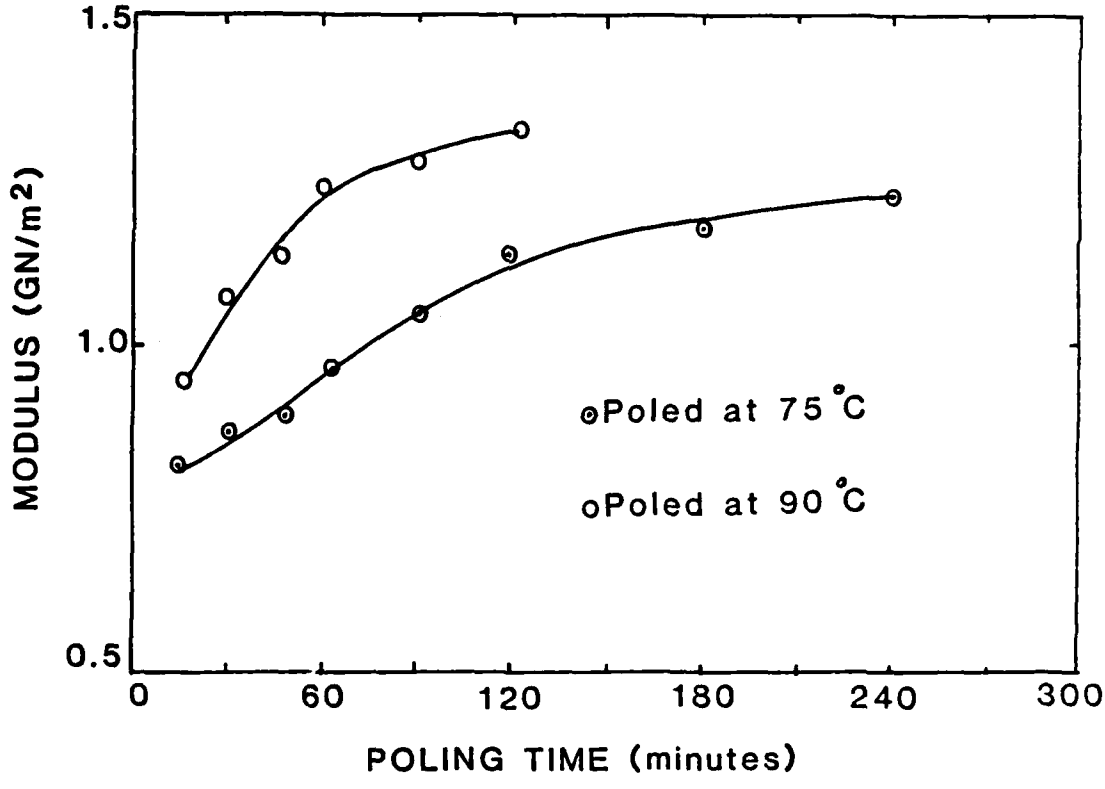
1.5
1.0
0.5
MODULUS (GN/m²)
0
60
120
180
240
300
POLING TIME (minutes)
⊙Poled at 75 °C
oPoled at 90 °C

FIGURE 11

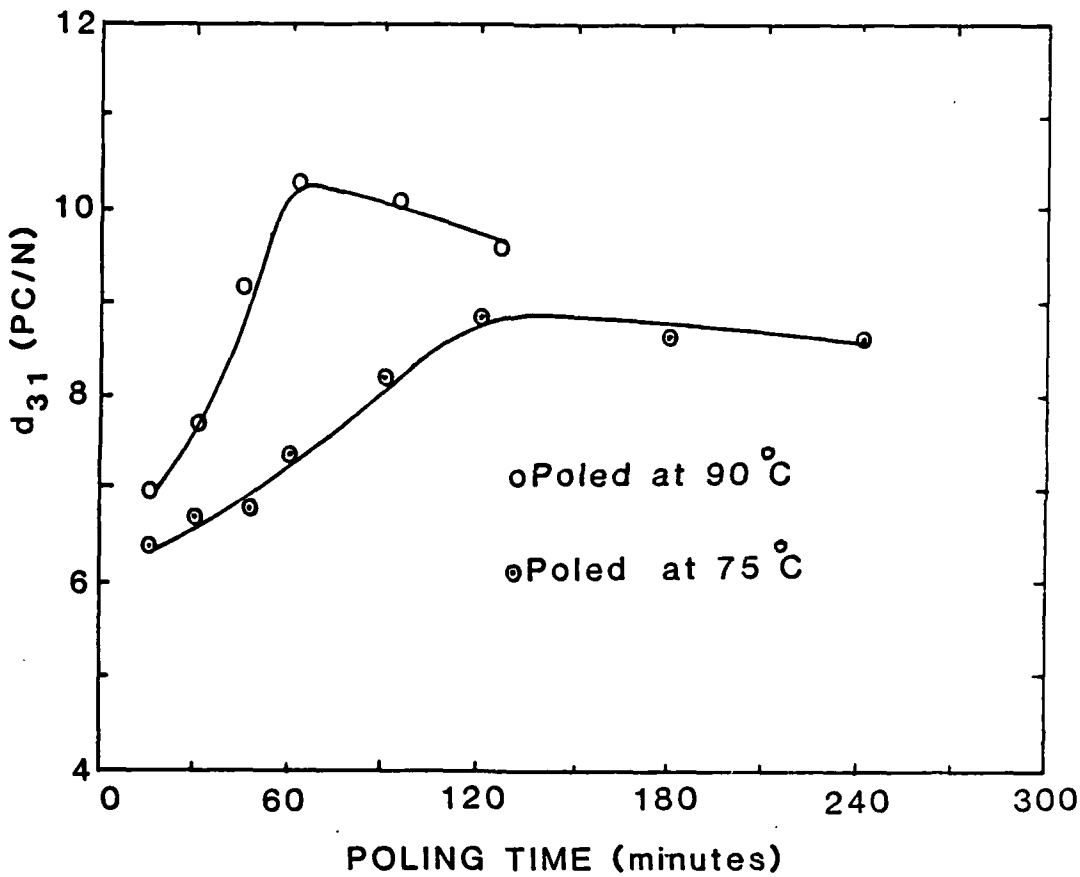
12
10
8
6
4
d_{31} (PC/N)
0
60
120
180
240
300
POLING TIME (minutes)
oPoled at 90 °C
oPoled at 75 °C

FIGURE 12

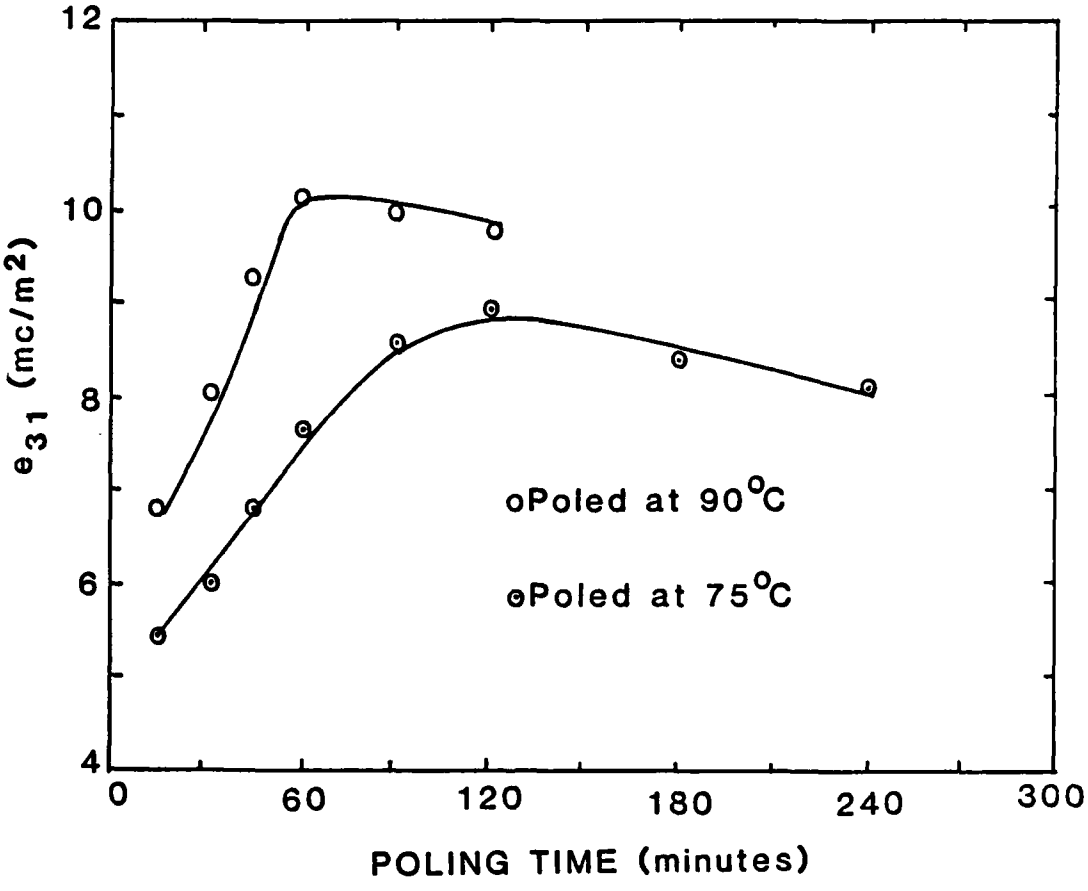

FIGURE 13

FIG. 13

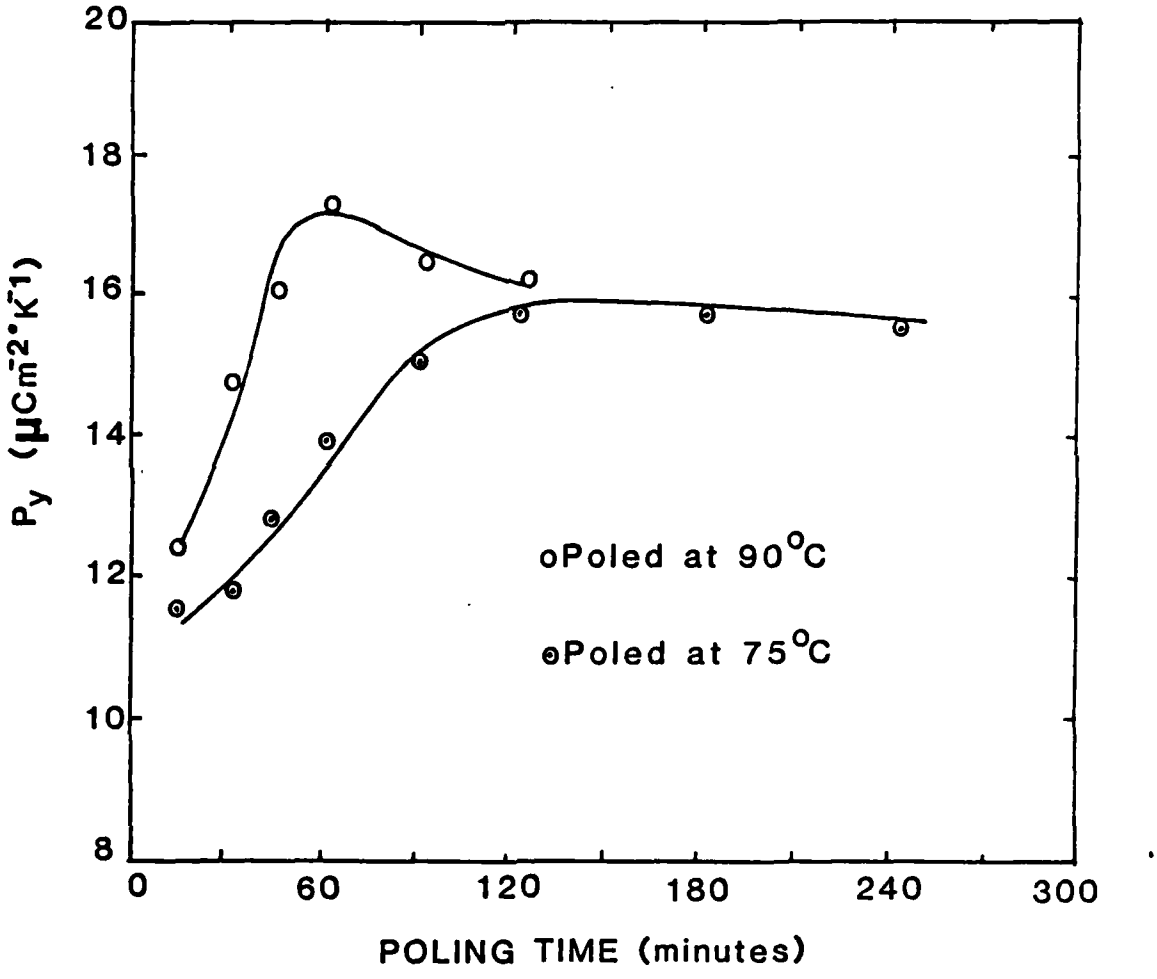
20
18
16
14
12
10
8
P_y ($\mu Cm^{-2}K^{-1}$)
0
60
120
180
240
300
POLING TIME (minutes)
oPoled at 90°C
⊙Poled at 75°C

FIGURE 14

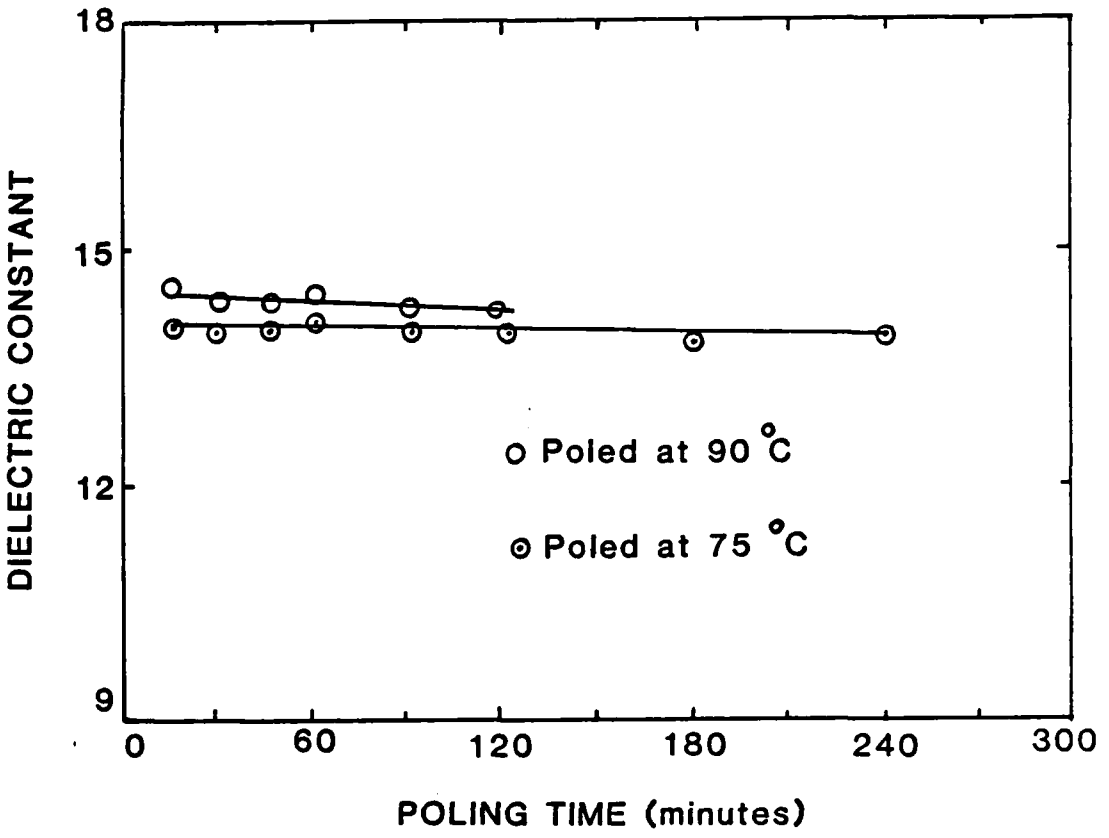
18
15
12
9
DIELECTRIC CONSTANT
0
60
120
180
240
300
POLING TIME (minutes)
Poled at 90 °C
Poled at 75 °C

FIGURE 15

DTIC

END

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