

AC Breakdown of Melt-Crystallized Isotactic Polypropylene

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Synopsis

The effect of polymer morphology on the dielectric breakdown of isotactic polypropylene was investigated under the influence of a ramped ac voltage. Samples were prepared by melt-pressing polypropylene beads into plaques and subsequently quenching or isothermally crystallizing these plaques. Some of the quenched samples were annealed at different temperatures to induce further crystallinity. The plaques were then characterized by wide-angle X-ray diffraction, density measurements, and differential scanning calorimetry. The 60 Hz, ac breakdown voltages of the samples were determined at 7°C, 23°C, and 45°C in a specially designed test cell. There was a lack of any significant effect on the ac breakdown strength of isotactic polypropylene of either the overall crystallinity or the crystallite thickness. This result can be understood in light of previous studies relating the electrical breakdown of polymers to morphology. It is postulated that the degree of crystallinity would influence the breakdown characteristics only when the amorphous regions constitute a small fraction of the total volume, that is, at very high crystallinity values—values beyond the range of the present study.

INTRODUCTION

Polymers have been replacing inorganics such as mica and ceramics as dielectrics in capacitor and insulation applications, due mainly to their higher breakdown strengths and lower densities. Ease of processing and molding is also in their favor.

Polymers lend themselves to a variety of heat treatments, which result in samples possessing a wide range of crystallinities, degrees of polymer chain orientation, and spherulitic and lamellar sizes. That these variables can influence the dielectric constant and loss tangent is well documented.^{1,2} However, the manner in which the electrical breakdown characteristics are affected by changes in the morphology is still not clear. In general, these effects are dependent on the temperature of measurement, the form of the voltage, and the polymer itself.^{3,4}

The voltage forms of interest in capacitor and insulation applications are dc, ac, or impulse. An impulse voltage is unidirectional, having fast rise times (of the order of 1 μ s) and slower decay times (approximately 50 μ s to decay to half the peak value). In general, the impulse dielectric strength of a polymer is higher than the dc dielectric strength at high temperatures, although the opposite trend is observed at lower temperatures.⁵ The higher dc breakdown strength at low temperatures has been explained by the injection and the subsequent trapping of electrons near the cathode.⁶ The existence of negative

space charges trapped (usually in the amorphous regions or in the interfaces of the crystalline and the amorphous regions) near the cathode reduces the electric field at that point and results in an increase in the apparent breakdown strength. On the other hand, under impulse conditions, there is not enough time for a space charge to develop. At higher temperatures, one finds that the space charges are able to migrate under a sustained voltage form like dc. Also thermal and/or electromechanical breakdown mechanisms may contribute to a lower dc dielectric strength when compared to the impulse strength. In contrast to all this, the ac dielectric strength of a polymer is substantially lower than the impulse or the dc dielectric strength at all temperatures due to the dissipative processes and partial discharges (electrical discharges that fall short of causing a complete breakdown) that occur during the application of alternating electrical stresses. Partial discharges during ac are more damaging and tend to occur at lower voltages than during dc stresses.⁷ The frequency of discharges during ac is also orders of magnitude higher than that with dc.⁷

There have been a number of studies (for the most part concerned with the dc or impulse dielectric strength of polyethylene) that have dealt with the effect of morphology on the electrical breakdown of polymers.⁸⁻¹⁴ The earliest such study was reported by Bird and Pelzer,⁸ who used a dc voltage to measure the dielectric strengths of two types of samples of low density polyethylene (LDPE), one quenched in cold water from the melt and the other crystallized at a slow rate from the melt. The two types of samples, possessing widely different degrees of crystallization and morphologies, exhibited almost the same breakdown characteristics over a wide range of temperatures. Miyauchi and Yahagi,⁹ on the other hand, observed that the dc dielectric strength of high-density polyethylene (HDPE) at temperatures below 80°C decreased with increasing crystallinity but above that temperature the opposite trend was seen to prevail. Cooper et al.¹⁰ found that the impulse electric strength of polyethylene at room temperature was unaffected by the density variations induced by heat treatment. However, when the polyethylene samples were prestressed with an opposing dc voltage, the impulse breakdown voltage at room temperature was found to decrease with decreasing density. Prestressing with a dc voltage of opposite polarity would create positive space charges near the impulse cathode (the dc anode), increasing the field and causing breakdown. Increasing the amorphous content of a polymer sample would increase the number of trap sites available for the space charges and therefore lower the breakdown strength. It is to be noted that the effect here is opposite to that obtained by Miyauchi and Yahagi⁹ below 80°C, where negative space charges near the cathode caused an improvement in the breakdown strength with decreasing crystallinity.

Similar studies of the effect of morphology on the dielectric strength of polypropylene have been few and inconclusive. Ceres and Schultz¹¹ reported that the times to breakdown of isotactic polypropylene samples, after the application of a dc voltage, were not affected by either the crystallinity or the crystallite thicknesses. Kaminaga et al.¹² determined that while the impulse breakdown strengths of polypropylene samples at room temperature were unaffected by the crystallite thickness, the impulse breakdown strength at 90°C increased with increasing crystallite thickness.

Wagner was the first to investigate the effect of spherulitic morphology on the breakdown behavior of polymers.¹³ Using a 60 Hz voltage, he determined that breakdown channels occurred at the boundaries of the spherulite. Kolesov¹⁴ and Ceres and Schultz¹¹ obtained similar results with dc voltages and found that the dielectric strength decreased with increasing spherulite size. This can be attributed to the fact that the breakdown channels have to follow pathways through the amorphous, interspherulitic regions of a sample.

In summary, at low temperatures, the effects of crystallinity on the breakdown characteristics of polyethylene, have been assigned to space charge modifications of the electric field. So also have the differences in the breakdown strengths measured using dc and impulse voltages. At high temperatures, space charge effects are less dominant and the breakdown processes may be dictated by electromechanical, thermal, or free volume considerations.

The objectives of the present study were to determine whether the morphology of polypropylene samples would affect its breakdown strength measured using an ac voltage of commercial power frequency and to see whether the temperature of measurement would have any influence on this score. The choice of the voltage form was dictated by the commercial importance of ac and the fact that very little work has been done on the dependence of the ac breakdown strength on the temperature of the sample.

EXPERIMENTAL

Sample Preparation

Sample plaques of 0.63 mm thickness were melt-pressed in a hydraulic press with heated plates using isotactic polypropylene beads kindly supplied by the Exxon Chemical Co. This thickness was chosen so as to avoid breakdown over the surface of the sample and to ensure that breakdown occurred through the bulk of the sample. The plaques that were formed in this manner were either quenched in cold water at 15°C or isothermally crystallized inside a vacuum oven at a high temperature. Some of the quenched samples were subjected to further annealing. Figure 1 illustrates the sample preparation procedure and lists the conditions of annealing and crystallization.

X-Ray Diffraction

Wide-angle X-ray diffractometry was employed to determine both the weight-percent crystallinities and the dimensions of the crystallites themselves. The X-ray diffraction scans were obtained using a General Electric XRD-6 unit employing a Cu-K α radiation. The 0.63 mm plaques were cut to the appropriate size and two of these plaques (having the same temperature history) were mounted back to back on the sample holder for each scan. This was done to discount the effect of sample thickness on the total intensity of the diffracted beams—an effect that would be noticeable with smaller thicknesses. The diffraction pattern obtained was that of the α form of isotactic polypropylene.¹⁵

The weight-percent crystallinity as well as the crystallite thicknesses were determined by procedures outlined by Wunderlich¹⁶ and Kakudo and Kasai.¹⁷ In the absence of a totally amorphous sample, the crystalline diffraction was

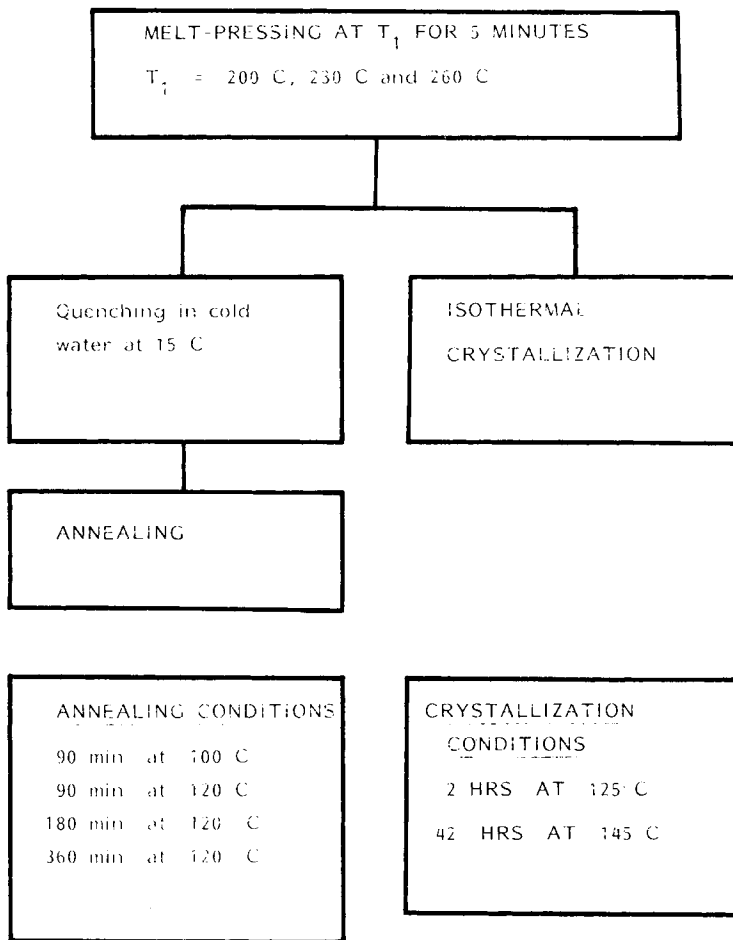


Fig. 1. Block diagram of the sample preparation procedure showing the thermal history of the various plaques.

separated from the amorphous diffraction by drawing a curve passing through the minima of the total intensity curve. Then, the weight-percent crystallinity, W_c , is given by

$$W_c = A_c / (A_c + KA_a) \quad (1)$$

where A_c = area of crystalline diffraction, A_a = area of amorphous diffraction, and $K = 0.98$ for polypropylene.¹⁶ Note that in measuring the areas, a straight line was drawn between the limits of the chosen interval of angles to separate the incoherent scattering from the rest.

The crystallite thicknesses were estimated from the diffraction peak widths using the Scherrer formula:

$$l_{hkl} = k\lambda / (\beta \cos \theta) \quad (2)$$

where k = Scherrer constant, λ = wave length of the X-ray (1.54 Å for the Cu-K_α line), θ = Bragg angle, β = half-width of the diffraction peak corre-

sponding to the $\{hkl\}$ planes, and l_{hkl} = mean dimension of the crystallites normal to the $\{hkl\}$ planes. The Scherrer constant is a number between 0.9 and 1.0. The measured half-width has to be corrected for the line-broadening caused by the finite size of the X-ray beam and by the existence of the Cu-K $_{\alpha}$ doublet.

Density and Differential Scanning Calorimetry Measurements

Density measurements were performed on some of the samples as an additional and relatively straightforward means of estimating the crystallinity. A density-gradient column filled with carbon tetrachloride and *n*-hexane was used for this purpose. Small samples cut out from the plaques were introduced into the column and the levels to which they had settled were noted after 2 h. Densities of the samples were determined from the calibration graph prepared from the positions at which glass floats of standard densities had settled.

Differential scanning calorimetry (DSC) of some of the samples was carried out to yield, among other things, the heats of fusion, the specific heats and the melting points. If the heat of fusion of the purely crystalline sample of the polymer is known, the weight-percent crystallinity can be calculated as follows:¹⁶

$$W_c = (\Delta h_f / \Delta h_{fc}) \times 100 \quad (3)$$

where Δh_f is the heat of fusion per gram of the polymer sample and Δh_{fc} is the heat of fusion per gram of the purely crystalline sample. For isotactic polypropylene Δh_{fc} is reported to be 209 J/g.¹⁸ A Dupont 1090 differential scanning calorimeter was used in the measurements. The scanning was performed under nitrogen cover at a rate of 10°C/min.

AC Breakdown Measurements

The 60 Hz, ac breakdown voltage measurements of the 0.63 mm thick samples were carried out inside a plexiglass tank filled with FC-43, a fluoro-carbon liquid from 3 M Co. During testing, the samples were held between opposing cylindrical brass electrodes of 0.63 cm diameter and the voltage raised at a rate of 1 kV/s till breakdown occurred. For measurements other than at 23°C, the plexiglass test cell was immersed in a constant temperature oil bath kept at the desired test temperature by circulating oil through a copper coil immersed in it. High voltage generation, control, and measurement were performed using a Hipotronics ac Dielectric Test Set (Model 7100 A). More details can be found elsewhere.^{19,20}

RESULTS

Table I presents the crystallinity and crystallite thickness values obtained from X-ray diffractometry. Although there is some scatter, it is evident that additional crystallinity was induced by the annealing process. Increasing the annealing temperature had the effect of increasing the crystallinity. Samples annealed at 120°C for different durations (up to 6 h), did not seem to show

TABLE I
Weight-Percent Crystallinities and Crystallite Thickness Perpendicular
to the [110] and [040] Planes

Serial no.	Thermal history	W_c	Thickness perpendicular to the [110] plane (Å)	Thickness perpendicular to the [040] plane (Å)
1	Melt-pressed at 230°C for 30 min, quenched in cold water	41%	85	92
2	Melt-pressed at 230°C for 5 min, quenched in cold water	49%	98	107
3	Same as sample 2, but annealed at 100°C for 90 min	53%	110	109
4	Same as sample 2, but annealed at 120°C for 90 min	63%	122	116
5	Same as sample 2, but annealed at 120°C for 180 min	64%	120	125
6	Same as sample 2, but annealed at 120°C for 360 min	62%	118	122
7	Melt-pressed at 200°C for 5 min, quenched in cold water, and annealed at 120°C for 180 min	58%	115	110
8	Melt-pressed at 260°C for 5 min, quenched in cold water, and annealed at 120°C for 180 min	53%	110	115
9	Melt-pressed at 230°C, isothermally crystallized at 125°C for 120 min	60%	194	150
10	Melt-pressed at 230°C, isothermally crystallized at 145°C for 42 h	61%	199	161

any increase in the crystallinity after 90 min of annealing. From Table I, it can also be seen that isothermal crystallization induced crystallinities only comparable to the quenched and annealed samples.

Furthermore, Table I shows that the crystallite thicknesses, in general, followed the trends shown by the crystallinity values with regard to the temperature and duration of annealing. The isothermally crystallized samples were found to have crystallite thicknesses which were greater than the annealed samples. This difference may be due to the fact that the slowly crystallized samples have a greater degree of perfection associated with their crystallites. In support of this, one may mention that broadening of the crystalline peaks is also known to be caused by imperfections in the crystals.

The crystallinity data obtained from the density measurements are listed in Table II. The crystallinity values were obtained using the following equation:¹⁶

$$W_c = \rho_c(\rho - \rho_a) / ((\rho_c - \rho_a)\rho) \quad (4)$$

where ρ is the density of the sample, ρ_a is the density of the completely amorphous sample, and ρ_c is the density of a completely crystalline sample of polypropylene. From Miller,¹⁸ $\rho_a = 0.854$ g/cc and $\rho_c = 0.946$ g/cc.

The data obtained from the DSC runs are presented in Table III, and these again show crystallinity trends similar to those observed with the other techniques of measurement.

TABLE II
Polymer Crystallinity Using Density Measurements

Serial no.	Temperature history	Density	Volume % crystallinity	Weight % crystallinity
1	Melt-pressed at 230°C for 5 min, quenched in cold water	0.903	49	51
2	Melt-pressed at 230°C for 30 min, quenched in cold water	0.894	36	38
3	Same as sample 1, but annealed at 100°C for 90 min	0.900	45	47
4	Same as sample 1, but annealed at 120°C for 90 min.	0.905	52	54
5	Same as sample 1, but annealed at 120°C for 180 min	0.907	56	58

TABLE III
DSC Results on Selected Samples

Serial no.	Temperature history	Melting point (°C)	ΔH_f (J/g)	Crystallinity (%)
1	Melt-pressed at 230°C for 5 min, quenched in cold water	164.1	86.9	42
2	Melt-pressed at 230°C for 30 min, quenched in cold water	166.5	86.5	41
3	Same as sample 1, but annealed at 100°C for 90 min	163.7	96.3	46
4	Same as sample 1, but annealed at 120°C for 90 min	162.9	96.8	46
5	Same as sample 1, but annealed at 120°C for 180 min	163.1	98.1	47

Table IV presents selected ac breakdown data for the samples for which the crystallinities and crystallite thicknesses were determined earlier. The major variables are the test temperature and the temperature at which the polypropylene beads were melted in the preparation of the plaques. Each of the ac breakdown voltages listed is an average of 7 to 11 determinations.

Figure 2 is a plot of the ac breakdown voltages of the samples melt-pressed at 230°C as a function of the temperature of measurement. Within the accuracy of the measurements there is no effect of temperature on the breakdown voltage. The temperature histories of the samples, as well, seem to

TABLE IV
Typical 60 Hz AC Breakdown Results

Serial no.	Thermal history	AC breakdown voltage (kV)		
		7°C	23°C	45°C
1	Melt-pressed at 230°C for 30 min, quenched in cold water	32.8	33.0	31.9
2	Melt-pressed at 230°C for 5 min, quenched in cold water	33.1	33.2	31.8
3	Same as sample 2, but annealed at 100°C for 90 min	34.0	32.4	33.0
4	Same as sample 2, but annealed at 120°C for 90 min	36.5	31.6	34.2
5	Same as sample 2, but annealed at 120°C for 180 min	35.4	32.3	34.7
6	Same as sample 2, but annealed at 120°C for 360 min	34.5	31.8	34.3
7	Melt-pressed at 200°C for 5 min, quenched in cold water, and annealed at 120°C for 180 min	33.5	33.7	33.8
8	Melt-pressed at 260°C for 5 min, quenched in cold water, and annealed at 120°C for 180 min	34.5	34.6	36.1
9	Melt-pressed at 230°C for 5 min and isothermally crystallized at 125°C for 120 min	33.4	33.8	30.2
10	Melt-pressed at 230°C for 5 min and isothermally crystallized at 145°C for 42 h	32.6	31.4	30.5

have no effect on the breakdown voltages at any of the three temperatures of measurement. This point is more clearly illustrated in Figures 3 to 5 where the breakdown voltages are plotted as a function of the weight-percent crystallinities. These plots show that within the spread of the data there does not seem to be any effect of the weight-percent crystallinity on the ac breakdown voltage of the samples.

DISCUSSION AND CONCLUSIONS

Weight-percent crystallinities were determined by three methods: wide-angle X-ray diffraction, density measurements, and DSC. A comparison of the values obtained with the different methods is shown in Table V. The crystallinity values from X-ray and density measurements are seen to match closely while those from DSC measurements are somewhat less than those obtained by the other two methods, especially at higher crystallinities. Such differences are not unusual and are observed frequently. The recrystallization that occurs during the DSC measurement may have something to do with the crystallinity values from this technique falling in a narrower range. The X-ray diffraction data appear to be more reliable and have been used throughout the discussion that follows.

The range of crystallinities achieved in the present study was 41–64%. The sample with the 41% crystallinity was obtained by melt-pressing the isotactic

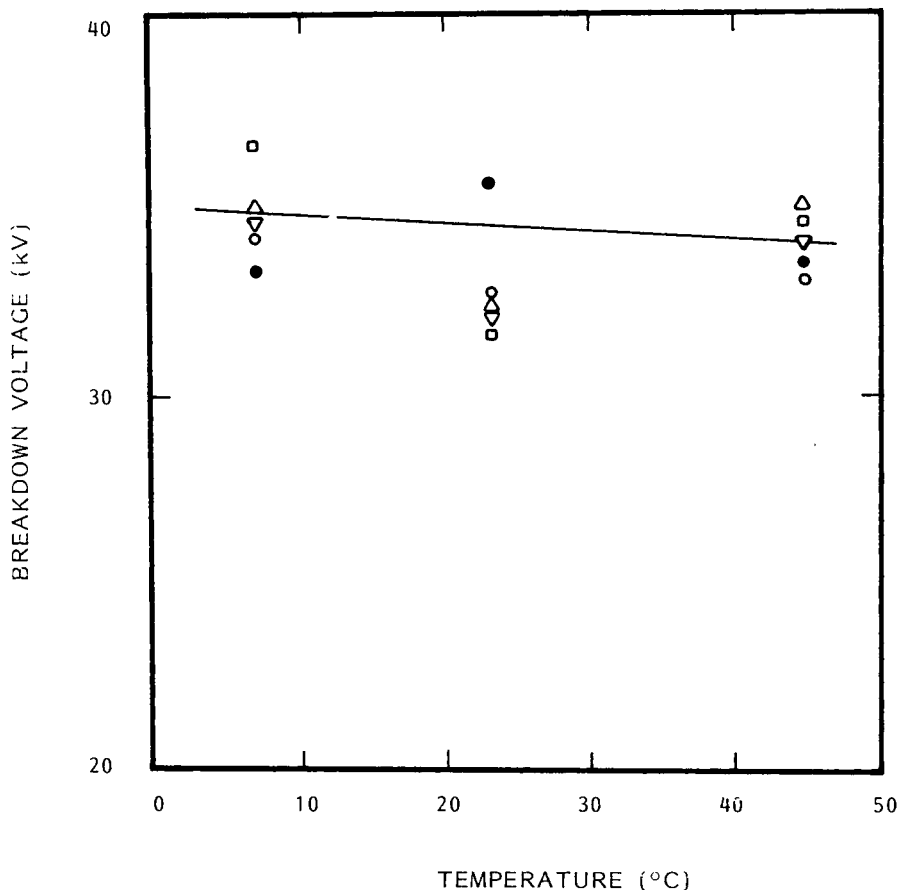


Fig. 2. Effect of temperature on the 60 Hz ac breakdown strength of isotactic polypropylene samples melt-pressed at 230°C. The different symbols denote different thermal histories: (●) quenched in cold water; (○) annealed at 100°C for 90 min; (□) annealed at 120°C for 90 min; (Δ) annealed at 120°C for 180 min; and (▽) annealed at 120°C for 360 min.

polypropylene beads at 230°C for 30 min, as opposed to 5 min for all other samples, before quenching in cold water. The difficulty in obtaining a sample of lower crystallinity is because the quench bath temperature (room temperature) is at least 30°C above the glass transition point of the polymer; crystallization can, therefore, occur even at room temperature.

In the present study, the highest degree of crystallinity obtained was 64%. In the literature, the maximum weight-percent crystallinity achievable for isotactic polypropylene has been variously reported as ranging from 65% to 80%.^{21,22} This variation may be caused partly by the differences in the measurement techniques. Ruland,²¹ for example, obtained crystallinities ranging from 31% to 65% for isotactic polypropylene samples using X-ray diffraction methods.

Blais and Manley²³ had observed that raising the annealing temperature caused a greater increase of the lamellar or crystallite thickness than an increase in the duration of annealing. This appears to be the case in the present study as well.

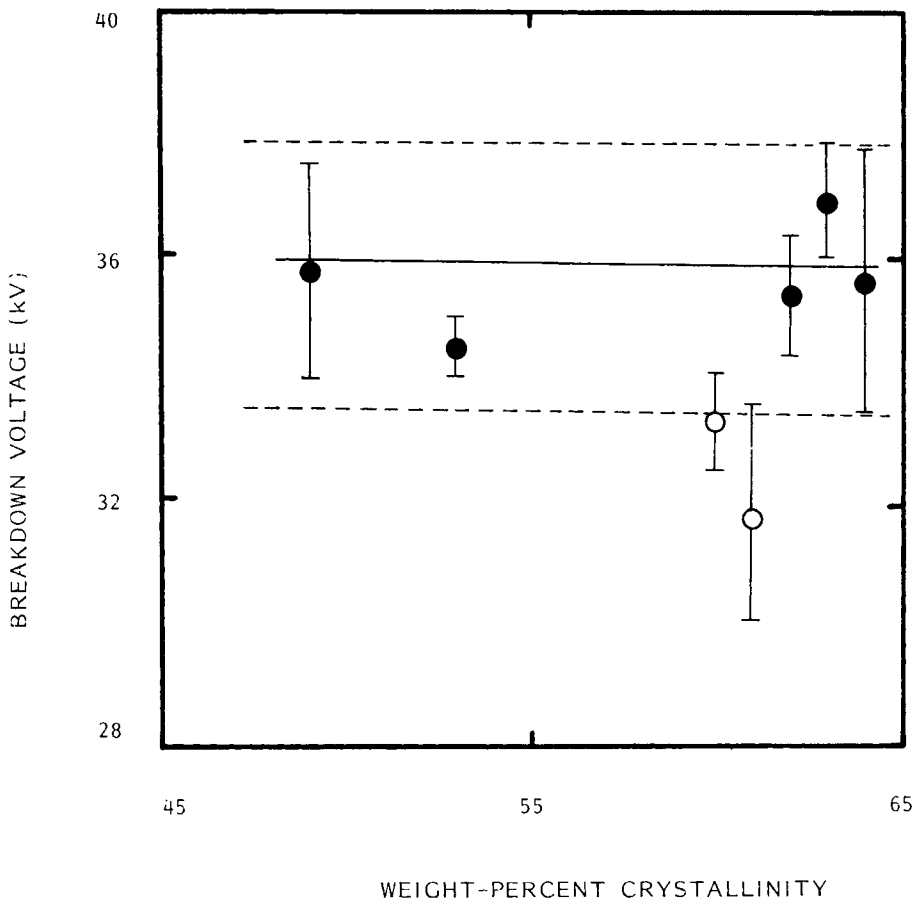


Fig. 3. Effect of weight-percent crystallinity on the breakdown strength of isotactic polypropylene. The filled symbols represent quenched and annealed samples whereas the open symbols denote isothermally crystallized plaques. The test temperature is 7°C.

For annealed samples, Figure 6 shows that the crystallite thicknesses increase linearly with the crystallinity. This is a clear indication that during annealing the increase in crystallinity is caused by an increase in the thickness of the crystallites rather than by a proliferation of the spherulites themselves.

The breakdown strengths obtained in the present study ranged from 40 to 56 kV/mm. The range of values is comparable to those that have been reported for thick samples.²² To avoid complications arising from the dependence of the breakdown strengths on the sample thicknesses, it was decided that samples having the same thickness (0.63 mm) would be used in the breakdown measurements and that breakdown voltages rather than breakdown strengths would be considered in the discussion.

In polymers, electrical breakdown is known to occur in the amorphous regions. These regions are in effect the weak links of the whole sample. At the time of initiation of this study it was thought that decreasing the amorphous fraction would increase the breakdown strength. The results of the present investigation, however, show no relation between these two quantities, and one can only speculate on the reasons for this behavior.

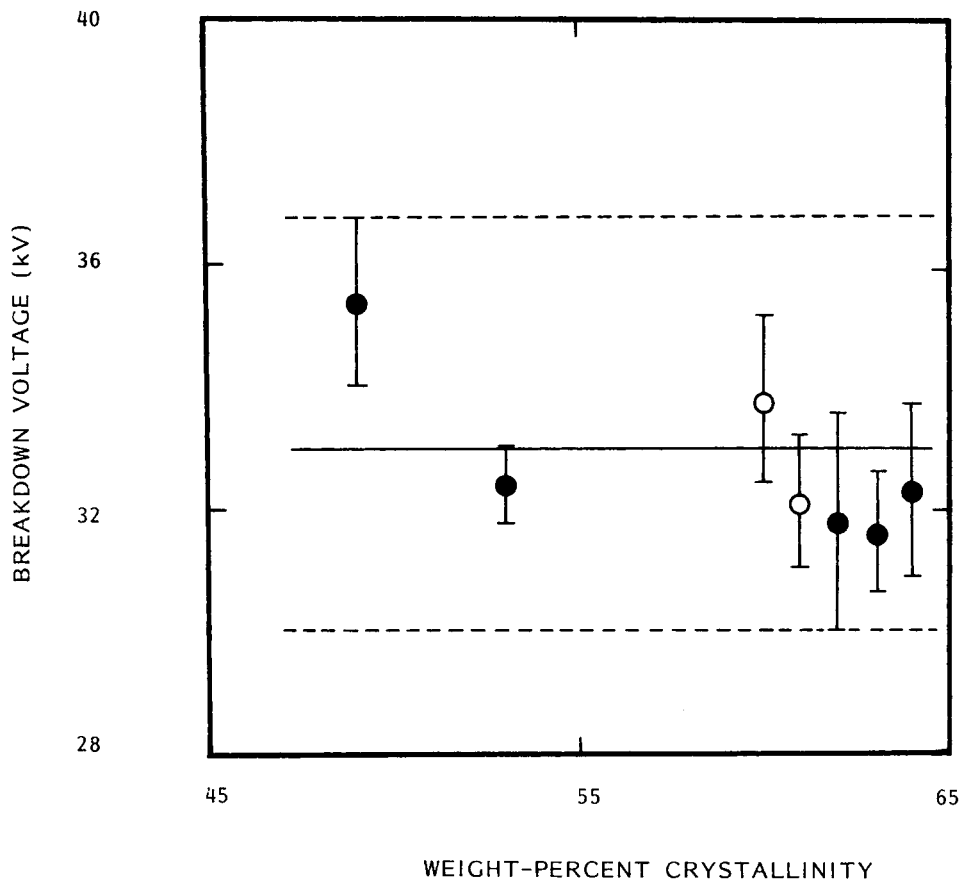


Fig. 4. Effect of weight-percent crystallinity on the breakdown strength of isotactic polypropylene. The filled symbols represent quenched and annealed samples whereas the open symbols denote isothermally crystallized plaques. The test temperature is 23°C.

Boyer²⁴ has documented the dependence of the glass transition temperature of a number of polymers on the degree of crystallinity. He found that while polymers like polyethylene, polystyrene, and polyethylene(terephthalate) exhibited an increase in the glass transition temperature (T_g) with an increase in the crystallinity, there was very little effect on the glass transition temperature of isotactic polypropylene. An increase in the T_g with increasing crystallinity is usually related to the restraints imposed by the crystalline regions on the molecular motion in the amorphous regions. A T_g independent of the crystallinity (as in isotactic polypropylene) implies a morphology wherein islands of crystalline material exist in an amorphous matrix but do not constrain the amorphous regions. If the morphology of melt-crystallized isotactic polypropylene is such that the amorphous regions are left unconstrained up to relatively high crystallinities, then electrical breakdown would also probably remain unaffected by fairly large changes in the crystallinity since, as mentioned before, breakdown is a phenomenon associated with the amorphous polymer.

In summary, there is a paucity of data on the dependence of the ac breakdown strength of any polymer on morphology. The breakdown mecha-

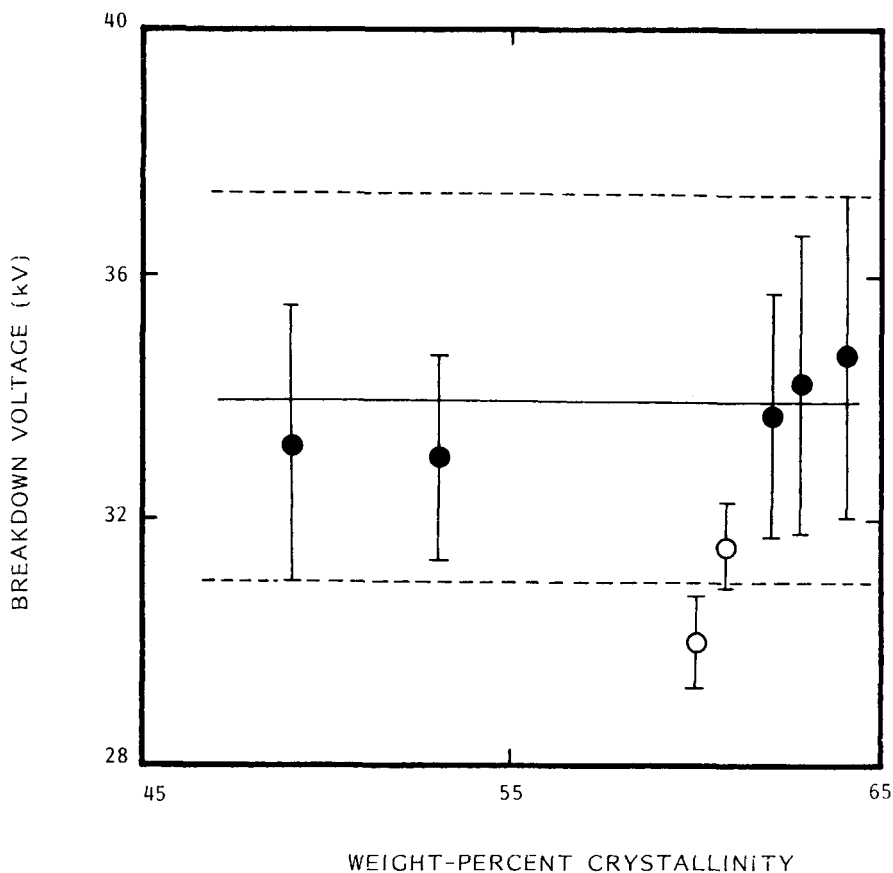


Fig. 5. Effect of weight-percent crystallinity on the breakdown strength of isotactic polypropylene. The filled symbols represent quenched and annealed samples whereas the open symbols denote isothermally crystallized plaques. The test temperature is 45°C.

TABLE V
Comparison of Crystallinity Values Using Different Techniques

Serial no.	Thermal history	Crystallinity (%)		
		X-ray	Density	DSC
1	Melt-pressed at 230°C for 5 min, quenched in cold water	49	51	42
2	Melt-pressed at 230°C for 30 min, quenched in cold water	41	38	41
3	Same as sample 1, but annealed at 100°C for 90 min	53	47	46
4	Same as sample 1, but annealed at 120°C for 90 min	63	54	46
5	Same as sample 1, but annealed at 120°C for 180 min	64	58	47

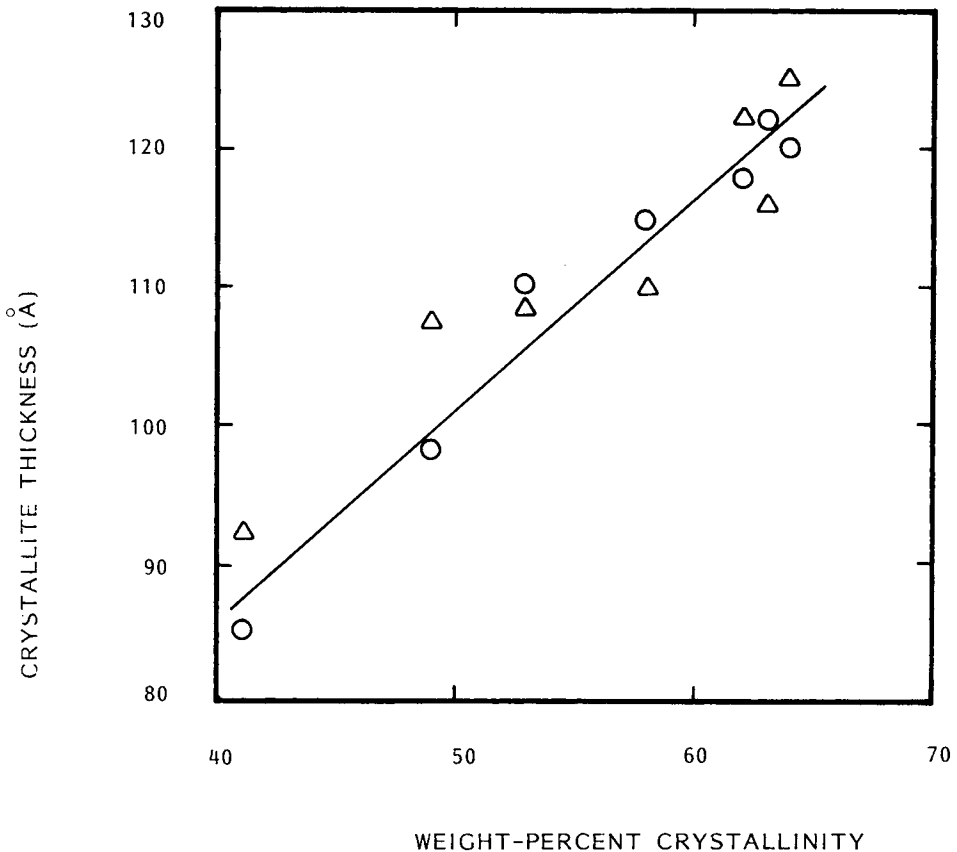


Fig. 6. Crystallite thickness as a function of weight-percent crystallinity. (○) thickness perpendicular to {110} plane; (Δ) thickness perpendicular to {040} plane.

nisms that are operative in the case of ac may be quite different from those that are operative in the dc or the impulse case. Partial discharges do more damage when they occur as a result of alternating stresses,²⁵ and these discharges can lead not only to eventual breakdown, but at higher levels can cause sudden breakdown also. The dissipative losses that accompany partial discharges are much higher than the losses that occur at low electric fields.²⁶ Thus, there may be an inherent nonideality associated with breakdowns under alternating stresses that seems to exclude considerations of morphology or structure.

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References

1. N. G. McCrum, B. E. Read, and G. Williams, *Anelastic and Dielectric Effects in Polymeric Solids*, John Wiley & Sons, New York, 1967.
2. P. J. Phillips, in *Engineering Dielectrics*, Vol. IIA, ASTM STP # 783, R. Bartnikas and R. M. Eichhorn, Eds., ASTM-Philadelphia, 1983.
3. K. Yahagi, *IEEE Trans. on Elec. Insul.*, EI-15, 241 (1980).
4. M. Nagao, M. Kosaki, and M. Ieda, in *Proceedings, 2nd International Conference on Conduction and Breakdown in Solid Dielectrics*, 1986, p. 126.

5. J. Artbauer and J. Griac, *Proc. Instit. of Elec. Eng.*, **112**, 818 (1965).
6. J. K. Nelson, in *Engineering Dielectrics*, Vol. IIA, STP 783, R. Bartnikas and R. M. Eichhorn, Eds., ASTM-Philadelphia, 1983.
7. R. J. Densley, in *Engineering Dielectrics*, Vol. I, STP 669 R. Bartnikas and E. J. McMahon, Eds., 1979.
8. D. W. Bird and H. J. Pelzer, *Inst. Elec. Eng. Part I*, **96**, 44 (1949).
9. H. Miyauchi and K. Yahagi, *Trans. Inst. Elec. Comm. Eng. Japan*, **92-A**, 36 (1972).
10. R. Cooper, B. Varlow, and J. P. White, *J. Physics*, Part D, **10**, 1521 (1977).
11. B. V. Ceres and J. M. Schultz, *J. Appl. Poly. Sci.*, **29**, 4183 (1984).
12. K. Kaminaga, K. Hirato, S. I. Irie, H. Kimura, I. Ishino, and H. Nakagawa, *Conference Records, 1986 IEEE Symp. on Elec. Insul.*, 1986, p. 170.
13. H. Wagner, 1974 *Annual Report CEIDP*, National Academy of Sciences-NRC, Washington, DC, 1975, p. 62.
14. S. N. Kolesov, *IEEE Trans. Elec. Insul.*, **EI-15**, 382 (1980).
15. A. Turner-Jones, J. M. Aizlewood, and D. R. Beckett, *Makromol. Chemie*, **75**, 134 (1964).
16. B. Wunderlich, *Macromolecular Physics*, Vol. I, Academic Press, New York, 1973.
17. M. Kakudo and N. Kasai, *X-Ray Diffraction by Polymers*, Elsevier Publishing Co., Amsterdam, 1972.
18. R. L. Miller, in *Polymer Handbook*, J. Brandrup and E. H. Immergut, Eds., Wiley-Interscience, New York, 1966.
19. B. Krishnakumar, R. K. Gupta, E. O. Forster, and J. R. Laghari, *Proc. Conf. Elect. Insul. Dielect. Phenomena, Claymont, Delaware*, 1986, p. 522.
20. B. Krishnakumar, Ph.D. thesis, Chemical Engineering, State University of New York at Buffalo, 1987.
21. W. Ruland, *Acta Crystallographica*, **14**, 1180 (1961).
22. J. Tanaka and K. Wolter, in *Engineering Dielectrics*, Vol. IIA, ASTM STP # 783, ASTM, Philadelphia, 1984.
23. J. J. B. P. Blais and R. St. J. Manley, *J. Macromol. Sci.*, **B1**, 131 (1967).
24. R. F. Boyer, in *Polymer Materials*, University microfilms International, Ann Arbor, 1986.
25. R. A. Fava, in *Treatise on Material Science and Technology*, Vol. 10B, J. M. Schultz, Ed., Academic Press, New York, 1977, p. 677.
26. R. Bartnikas, in *Engineering Dielectrics*, Vol. I, ASTM STP # 669, R. Bartnikas and E. J. McMahon, Eds., 1979.

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