

Yield Behavior of Unoriented and Oriented Polycarbonate and Polypropylene as Influenced by Temperature

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(Received April 13, 1979; in revised form July 19, 1979)

SUMMARY

The yield behaviors of an amorphous polymer (polycarbonate) and a crystalline polymer (polypropylene) were investigated over certain ranges of temperature. Both polymers were used in an unoriented (isotropic) and an oriented (anisotropic) condition. By using proposed yield criteria for the two structural conditions various theoretical yield loci are predicted; these are then compared with experimental findings based on a number of uniaxial and biaxial stress states. With a few exceptions that seem amenable to rational explanation the comparison between theory and experiment is most promising. The onset of yielding is defined by two methods: using a 0.3% offset and using the concept of plastic work. Similar findings result. Finally, for the range of parameters used in this study it is possible to compare individual results for a given material condition with a single yield locus, regardless of the temperature at which the tests were conducted.

1. INTRODUCTION

The macroscopic yield behavior of unoriented (isotropic) and oriented (anisotropic) polymers has been investigated in previous studies [1 - 5]. A comparison of experimental findings and theoretical predictions has been made for several unoriented polymers using a modification of the von Mises yield criterion which takes into account the influence of the mean normal stress and the difference that is

usually observed between the tensile and compressive yield strengths [1, 2]. Hill's [6] modification of the von Mises criterion, which includes the influence of anisotropy, has been further modified [3] for use with oriented polymers, and subsequent studies [4, 5] have provided comparisons between experimental results and theoretical predictions. In all these earlier investigations comparisons have resulted by the development of yield loci (first and fourth quadrant points only) based on yielding caused by uniaxial and biaxial stress states at room temperature (nominally 25 °C). Other studies, e.g. refs. 7 and 8, consider the effects of pressure on yielding.

To pursue further the applicability of the yield criteria for unoriented [1] and oriented [3] polymers, Woodliff [9] has considered the effects of temperature on the yield loci of oriented and unoriented polypropylene (PP) and polycarbonate (PC). His major findings constitute the basis of this paper.

2. ANALYTICAL FORM OF PROPOSED YIELD CRITERIA

For brevity full details of the developments leading to the proposed yield criteria are not included here; they can be found elsewhere [2, 3]. In terms of principal stresses, which are pertinent to this paper, the criterion for isotropic polymers is

$$(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 + 2(C - T)(\sigma_1 + \sigma_2 + \sigma_3) = 2CT \quad (1)$$

where C and T are the absolute values of the compressive and tensile yield strengths measured at atmospheric pressure.

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For anisotropic polymers eqn. (1) is modified to the form

$$H(\sigma_1 - \sigma_2)^2 + F(\sigma_2 - \sigma_3)^2 + G(\sigma_3 - \sigma_1)^2 + K_1\sigma_1 + K_2\sigma_2 + K_3\sigma_3 = 1 \quad (2)$$

where the parameters H , F and G characterize the state of anisotropy [6] and K_1 , K_2 and K_3 account for the pressure dependence.

Since the scope of this paper is limited to biaxial stress states (*i.e.* $\sigma_3 = 0$ throughout), eqns. (1) and (2) reduce to

$$\sigma_1^2 + \sigma_2^2 - \sigma_1\sigma_2 + (C - T)(\sigma_1 + \sigma_2) = CT \quad (3)$$

for the isotropic case and

$$(H + G)\sigma_1^2 + (H + F)\sigma_2^2 - 2H\sigma_1\sigma_2 + K_1\sigma_1 + K_2\sigma_2 = 1 \quad (4)$$

for the anisotropic case.

It was found helpful to use a normalized form of eqns. (3) and (4) for comparative purposes; this was done by defining the following parameters for use with eqn. (3):

$$R_1 = \sigma_1/T \quad R_2 = \sigma_2/T \quad (5)$$

The normalized form of eqn. (3) becomes

$$R_1^2 + R_2^2 - R_1R_2 + \left(\frac{C}{T} - 1\right)(R_1 + R_2) = \frac{C}{T} \quad (6)$$

In the anisotropic case, since T is not independent of direction the normalizing parameters used are

$$R_1 = \sigma_1/T_1 \quad R_2 = \sigma_2/T_1 \quad (7)$$

The normalized form of eqn. (4) becomes

$$(H + G)R_1^2 + (H + F)R_2^2 - 2HR_1R_2 + \frac{K_1R_1}{T_1} + \frac{K_2R_2}{T_1} = \frac{1}{T_1^2} \quad (8)$$

Now the various parameters, all of which depend on the absolute values of the tensile and compressive* yield strengths in the three principal directions, are defined as

*Note that, for example, $C_1 = |C_1|$ etc. For simplicity the absolute value signs are omitted.

$$H + G = \frac{1}{C_1T_1}$$

$$F + H = G + F = \frac{1}{C_2T_2} = \frac{1}{C_3T_3} \quad (9)$$

$$K_1 = \frac{C_1 - T_1}{C_1T_1}$$

$$K_2 = K_3 = \frac{C_2 - T_2}{C_2T_2} = \frac{C_3 - T_3}{C_3T_3}$$

It has been found [4, 5] that, for the particular polymers studied, deliberate orientation in say the 1 direction leads to the observation that $C_2 = C_3$, $T_2 = T_3$ but $C_2 \neq T_2$. This produces the results in eqn. (9) plus the observation that $H = G$. From these relationships, eqn. (8) can then be expressed as

$$R_1^2 + \frac{H + F}{H + G}R_2^2 - R_1R_2 + \frac{K_1R_1}{T_1(H + G)} + \frac{K_2R_2}{T_1(H + G)} = \frac{1}{T_1^2(H + G)} \quad (10)$$

To simplify eqn. (10) new parameters are introduced as follows:

$$X = \frac{H + F}{H + G} = \frac{C_1T_1}{C_2T_2} \quad (11a)$$

$$W = \frac{K_2}{T_1(H + G)} = \frac{C_1}{C_2} \left(\frac{C_2}{T_2} - 1 \right) \quad (11b)$$

$$Z = \frac{1}{T_1^2(H + G)} = \frac{C_1}{T_1} \quad (11c)$$

Equation (10) can then be written as

$$R_1^2 + XR_2^2 - R_1R_2 + (Z - 1)R_1 + WR_2 = Z \quad (12)$$

For the remainder of this paper eqns. (3) and (6) are used for those studies concerned with unoriented PP and PC; only C and T need be evaluated at the test temperature to predict a theoretical yield locus for various stress ratios. Equations (4) and (12) will be used for the oriented polymers; in this case, at the temperature of concern it is necessary to determine the values C_1 and T_1 of the compressive and tensile yield strengths in the direction of orientation as well as the yield

strengths C_2 and T_2 in a direction perpendicular to the orientation. From these basic findings a theoretical yield locus can then be predicted for the test temperature. Theory and experiment can be compared if enough experiments are performed using various biaxial stress ratios.

3. MATERIALS

Solid cylindrical rods of PC (one 35 mm and one 51 mm in diameter) and PP (one 35 mm and one 64 mm in diameter) were obtained from the Westlake Plastics Company. The smaller rods were used for the unoriented studies and the larger ones for the oriented tests. Except for producing the orientation required for such experiments, all the rods were used in the as-received condition.

Both microscopic and macroscopic tests were conducted to determine the isotropy of these commercial rods. The details of the tests have been presented earlier [5]. As found in our earlier study [5], all the rods exhibited initial isotropy to a degree that was fully acceptable for the purposes of the present investigation.

To produce a highly oriented structure, large tensile specimens were produced from the larger bar of each polymer. These were pulled until a stable neck had formed and had been allowed to propagate for an adequate length. The procedural details and precautions concerning such use of PC have been presented in detail elsewhere [4, 5, 9].

4. TEST SPECIMENS

It is to be understood in what follows that various specimens of both polymers were used in both oriented and unoriented structural conditions.

4.1. Uniaxial compression

Right circular cylinders were machined such that their axis of revolution was either parallel or perpendicular to the axis of the original or oriented bars. Unoriented specimens of PC were 6.35 mm in diameter and 12.70 mm long; the PP specimens were 8.89 mm in diameter and 15.24 mm long. To take full advantage of the machine capacity

both unoriented and oriented PP specimens used at temperatures lower than 0 °C were reduced in diameter to 6.35 mm while oriented PP specimens used above 0 °C were 7.62 mm in diameter. Note that the unoriented materials provided the basic value of C in eqn. (6) (which was found to be effectively independent of direction) while the values of C_1 and C_3 for use in eqn. (12) differed according to the "direction" in which the specimens had been machined.

4.2. Uniaxial tension

Specimens having a uniform gauge length of 51 mm were machined parallel to the axis of the unoriented rods. The gauge diameter of the PC specimens was 6.35 mm; that of the PP specimens was 8.89 mm (7.62 mm for tests below 0 °C). The uniaxial tension tests gave the value of T in eqn. (6). To determine values of T_1 and T_2 for use in eqn. (12), tubes were made from the oriented materials. Use of an unpressurized tube loaded in uniaxial tension gave T_1 ; T_2 was obtained using an "open-ended" tube test (details are given elsewhere [1, 4, 5, 9]).

4.3. Thin-walled tubes (biaxial conditions)

The test section of these tubes was 51 mm long, 22.6 mm in outer diameter (17.3 mm for oriented PP) and 20.6 mm in inner diameter (15.9 mm for oriented PP). Tubes made from the oriented materials were machined so that the entire test section consisted of material that lay within the stable neck (see Section 3).

5. TEST PROCEDURE

All tests were conducted on a 500 kgf Instron machine using a constant cross-head speed of $8.47 \mu\text{m s}^{-1}$. The specimens were located inside an Instron environmental chamber of nominal temperature range from -73 °C to 204 °C. A carefully developed temperature calibration curve was used to provide more accurate temperature measurements than those indicated roughly by the control knob on the chamber. Exploratory studies indicated that a period of 30 min was sufficient to bring about thermal equilibrium throughout the unit and the test specimens. This warm-up period was used in all the

individual tests except those at room temperature. Compression specimens were loaded between two stainless steel platens; comparisons between lubricated and unlubricated interfaces indicated that frictional effects were negligible. An extensometer which closed up during loading was adapted to the platens and except for negligibly small deflections of these steel members its displacement gave the decrease in height of the specimen as loading proceeded. Signals due to changes in both the load and the length were fed through an amplifier and were then displayed on an X-Y-Y recorder to provide a continuous plot of load *versus* displacement. A test was concluded at the first observable sign of barrelling. Tensile tests were run with the extensometer adapted to the gauge section of the specimen; all other details were as for the compression tests. The thin-walled tubes were subjected to internal fluid pressure and simultaneous axial loading in either tension or compression; such combinations provided a number of conditions for various constant stress ratios (*i.e.* σ_1/σ_2). In earlier studies [1, 4, 5] pressure had been controlled by a hand pump. This had caused some concern since it was impossible to achieve a truly constant loading line, although deviations were quite small. To avoid this problem in the present study we built a specially designed feedback pressure-control device. This receives the signal from the load cell and in essence adjusts the pressure to the desired level. The resulting loading line provides an almost perfectly constant stress ratio that can be adjusted to give any desired loading line. Dow Corning 510 silicone oil was used to produce the internal tube pressure since it reduces the chance of crazing and remains serviceable from -58°C to 204°C . Changes in length were again detected with a standard extensometer; a second extensometer was modified to measure the outer tube diameter during loading. Signals from these two devices plus that from the load cell were displayed on the X-Y-Y recorder.

6. CONVERSION OF RAW DATA

All the load-displacement data from the uniaxial tension and compression tests were converted to values of stress and strain. For

the tube tests, values of true strain in the 1 and 2 directions were computed directly from the changes in length and diameter. Because of difficulties in measuring changes in wall thickness the volume was assumed to be constant to compute the thickness strain. This has been shown to be a reasonable assumption [1]. Using the computed value of the thickness strain, instantaneous values of wall thickness were computed, and these constituted the unknown dimension that permitted calculations of the true stress in the axial direction. The equations used in these calculations involving tube tests are

$$\begin{aligned}\epsilon_1 &= \ln\left(\frac{l}{l_0}\right) & \epsilon_2 &= \ln\left(\frac{D}{D_0}\right) \\ \epsilon_3 &= -(\epsilon_1 + \epsilon_2) = \ln\left(\frac{t}{t_0}\right)\end{aligned}\quad (13)$$

where l , D and t are instantaneous values of length, outer diameter and wall thickness and the subscript zero indicates the initial values of these three quantities. The axial stress σ_1 and the hoop or tangential stress σ_2 were determined from

$$\sigma_1 = \frac{Pd^2 + 4L/\pi}{4t(d+t)} \quad \sigma_2 = \frac{Pd}{2t} \quad (14)$$

where P is the internal pressure, L is the axial load (tension or compression) and $d = D - 2t$ is the inner diameter.

7. EVALUATION OF YIELDING

As yet there is no universally accepted definition of the onset of yielding in polymers. As mentioned in earlier studies [1, 4, 5, 10] the maximum load prior to the onset of neck formation cannot be consistently used since not all polymers show this behavior. We therefore relied on the traditional offset method. Admittedly the choice of offset used is arbitrary; the major advantage is consistency. In this study a 0.3% offset was chosen as one method of defining yielding. To use this method when biaxial stress states are involved, the two stresses must be plotted against a common strain function. For this reason we used the effective strain function associated with the von

Mises criterion. Assuming a constant volume the effective strain $\bar{\epsilon}$ is given by

$$\bar{\epsilon} = \left\{ \frac{4}{3} (\epsilon_1^2 + \epsilon_1\epsilon_2 + \epsilon_2^2) \right\}^{1/2} \quad (15)$$

It should be mentioned that in all such tests the ratio of σ_1 to σ_2 at yielding was almost identical with the loading ratio of these stresses for each individual test (Fig. 1).

Yielding was also defined using the relation between the effective strain and the plastic work. The plastic work per unit volume in terms of principal stresses and strain increments is

$$dW = \sigma_1 d\epsilon_1 + \sigma_2 d\epsilon_2 + \sigma_3 d\epsilon_3 \quad (16)$$

or in terms of the effective stress and strain is

$$dW = \bar{\sigma} d\bar{\epsilon} \quad (17)$$

The use of the effective strain to define σ_1 and σ_2 at yielding for different states of stress on a yield locus tacitly assumes that the plastic work is equivalent in all cases. This is shown schematically in Fig. 2 where the tension test is used as a basic reference and the tensile yield strength is defined by a 0.3% offset. Yielding under compression or biaxial stress is assumed to require an amount of plastic work that is equivalent to that determined from the tensile test. The areas in Fig. 2 are related by $A_T = A_C = A_1 + A_2$. Using this method it was found that the

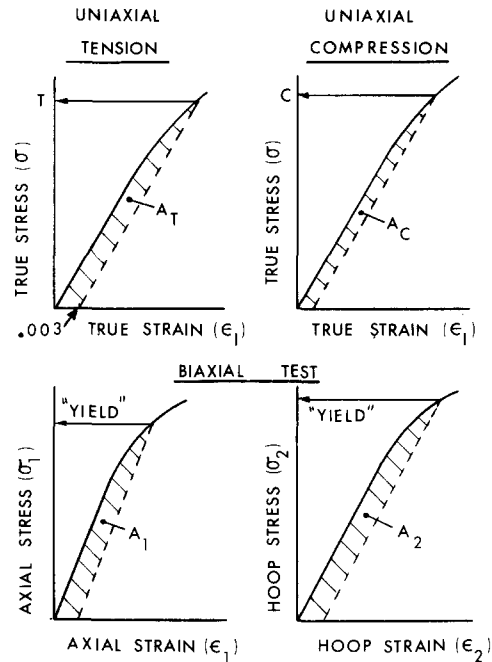


Fig. 2. The equivalence of plastic work to define yielding due to different stress states.

equivalence of plastic work for these three general situations does not usually lead to equivalence in the offset that results.

8. TEST RESULTS

In a complementary study it was discovered that both oriented PP and oriented PC show distinct changes in their stress-strain behavior at annealing temperatures as low as 50 °C. Thus it was concluded that studies involving these two structures must be limited to temperatures at or below that at which orientation was induced. The four discrete values of temperature used were 25, 0, -25 and -50 °C.

With regard to the unoriented materials, PC was studied at temperatures of 100 and -50 °C* whereas PP was subjected to temperatures of 50, 25, 0 and -50 °C.

Figures 3 and 4 show the experimental results compared with the theoretical yield loci based on eqn. (3). Figures 5 and 6 show the test points and the loci given by eqn. (4). Because the various loci expand in a fairly

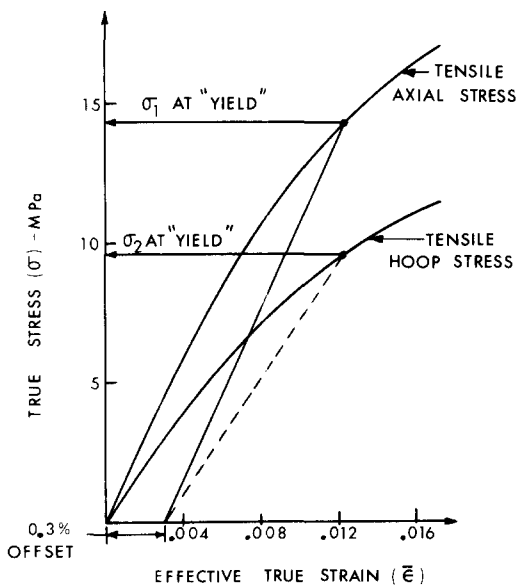


Fig. 1. Biaxial stress against effective strain for a PP tube tested at 25 °C, with a stress ratio σ_1/σ_2 of 1.49 and using a 0.3% offset to define yielding.

*A few tests at 25 °C corresponded very closely to earlier work [1, 4, 5] so a repetition of tests at this temperature seemed unnecessary.

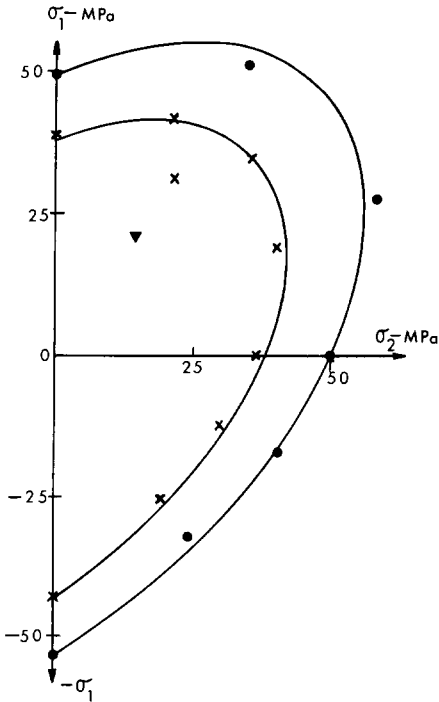


Fig. 3. A comparison between yield loci based on eqn. (3) and experimental results for unoriented PC at two temperatures: x, 100 °C; •, -50 °C; ▼, hydraulic oil.

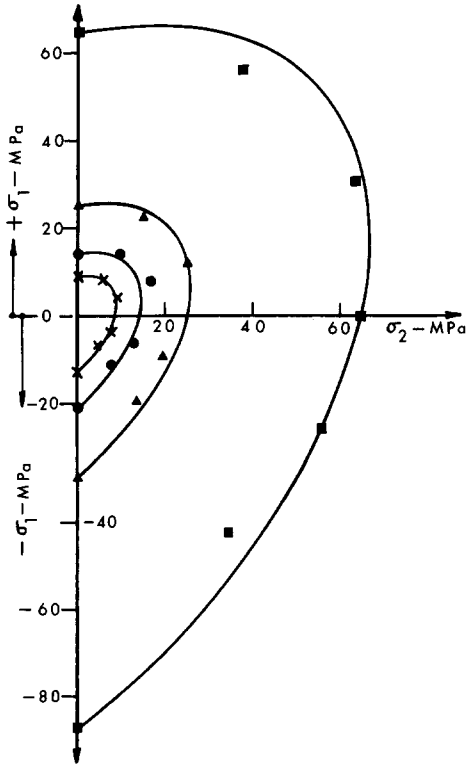


Fig. 4. A comparison between yield loci based on eqn. (3) and experimental results for unoriented PP at various temperatures: x, 50 °C; •, 25 °C; ▲, 0 °C; ■, -50 °C.

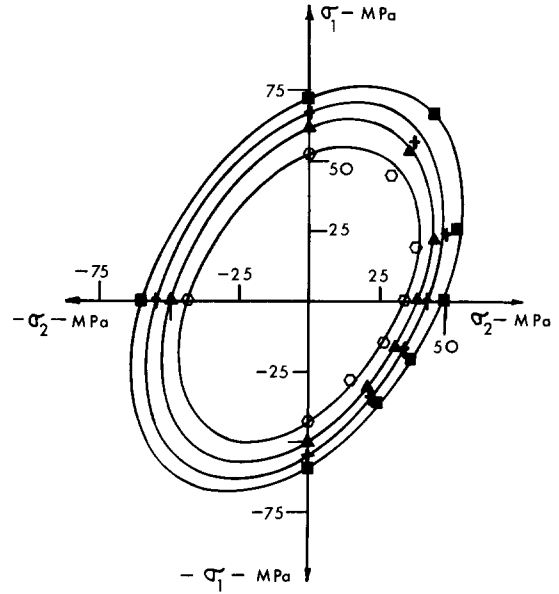


Fig. 5. A comparison between yield loci based on eqn. (4) and experimental results for oriented PC at various temperatures: ○, 25 °C; ▲, 0 °C; +, -25 °C; ■, -50 °C.

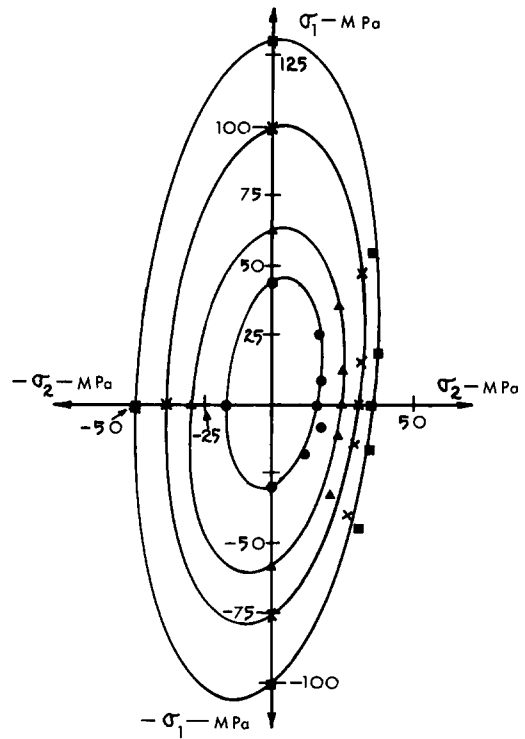


Fig. 6. A comparison between yield loci based on eqn. (4) and experimental results for oriented PP at various temperatures: •, 25 °C; ▲, 0 °C; x, -25 °C; ■, -50 °C.

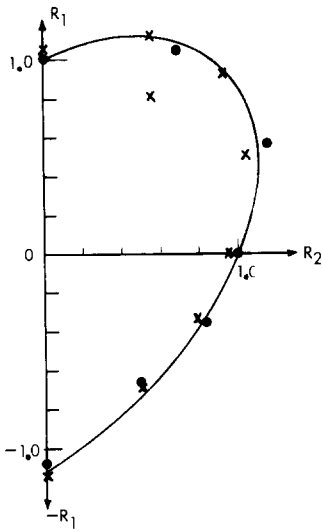


Fig. 7. A normalized yield locus based on eqn. (6) and experimental results for unoriented PC at two temperatures: x, 100 °C; ●, -50 °C.

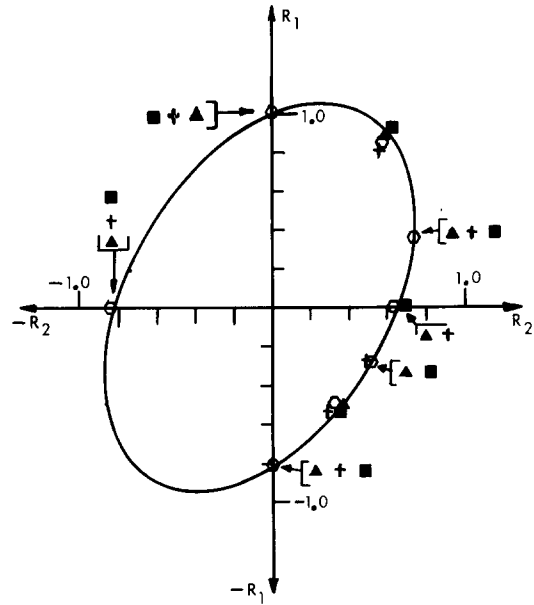


Fig. 9. A normalized yield locus based on eqn. (12) and experimental results for oriented PC at various temperatures: ○, 25 °C; ▲, 0 °C; +, -25 °C; ■, -50 °C.

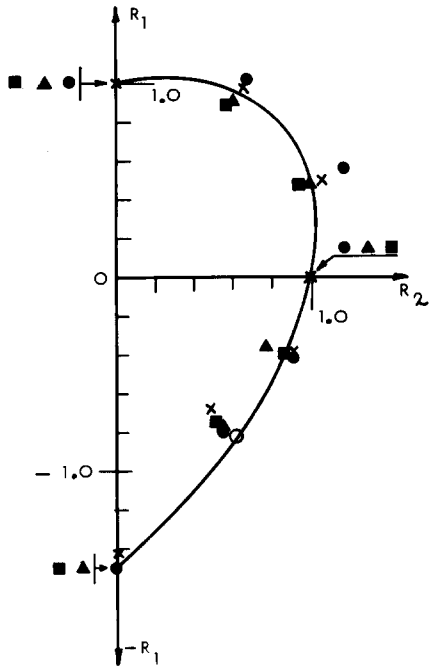


Fig. 8. A normalized yield locus based on eqn. (6) and experimental results for unoriented PP at various temperatures: x, 50 °C; ●, 25 °C; ▲, 0 °C; ■, -50 °C; ○, 25 °C (thick tube).

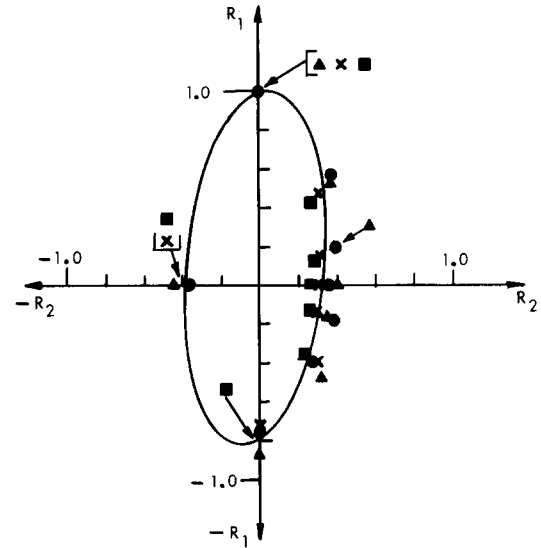


Fig. 10. A normalized yield locus based on eqn. (12) and experimental results for oriented PP at various temperatures: ●, 25 °C; ▲, 0 °C; x, -25 °C; ■, -50 °C.

uniform manner as the temperature is decreased it was found that the data on each of these four figures could be plotted on a single yield locus using the concept of normalization. Figures 7 - 10 show the results when the theoretical loci in Figs. 3 and 4 were developed using eqn. (6) and the theoretical loci in Figs. 5 and 6 were developed using eqn. (12).

The normalizing parameters were determined for each material-temperature combination and were then averaged; they are given in Table 1.

To provide further comparison, Fig. 11 is a normalized yield locus for unoriented PC based on data from the present study (at -50 and 100 °C) and results obtained by Raghava [1] at 25 °C. Although the material used in the earlier study at 25 °C came from an

TABLE 1
Normalized yield loci parameters

	X	W	Z
Unoriented PC ^a	1.00	0.11	1.11
Unoriented PP ^a	1.00	0.48	1.48
Oriented PC	1.53	0.266	0.820
Oriented PP	5.967	0.389	0.769
Combined oriented PC	1.556 ^b	0.271 ^b	0.747 ^b

^aIn the isotropic case where eqn. (6) applies, $X = 1$, $W = C/T - 1$ and $Z = C/T$. Equation (12) is used for the oriented (anisotropic) case.

^bThese values are based on results from the present study and from refs. 4 and 5.

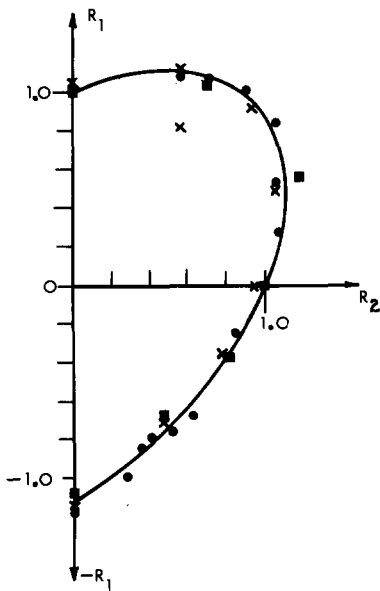


Fig. 11. A normalized yield locus based on eqn. (6) and results for unoriented PC from the present work and from ref. 1: x, 100 °C (present work); ■, -50 °C (present work); ●, 25 °C (ref. 1).

entirely different batch of PC, excellent agreement can be seen. Figure 12 shows a similar comparison of earlier results [4, 5] concerning oriented PC with data from the current study; again the overall correlation is excellent. Table 1 includes the average of the normalizing parameters used in connection with Fig. 12.

Figures 13 and 14 concern the use of the concept of plastic work to define yielding. They show results for oriented PC and PP tested at 25 °C and include test points based on yielding as defined by the effective strain function and plastic work. In each of the figures the yield locus was drawn on the basis

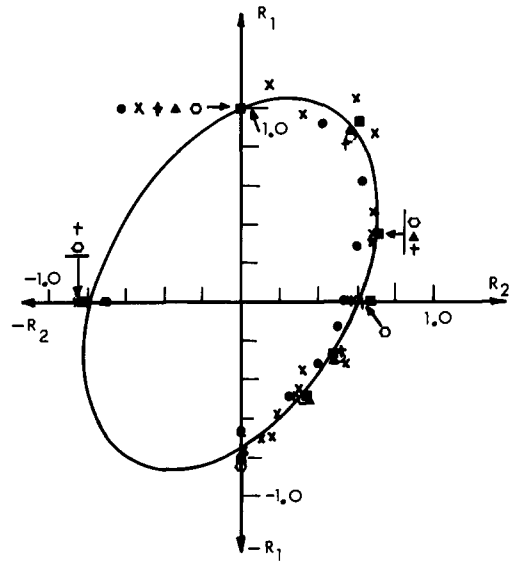


Fig. 12. A normalized yield locus based on eqn. (12) and results for oriented PC from the present work and from refs. 4 and 5: ○, 25 °C (present work); ▲, 0 °C (present work); +, -25 °C (present work); ■, -50 °C (present work); x, 25 °C (ref. 4); ●, 25 °C (ref. 5).

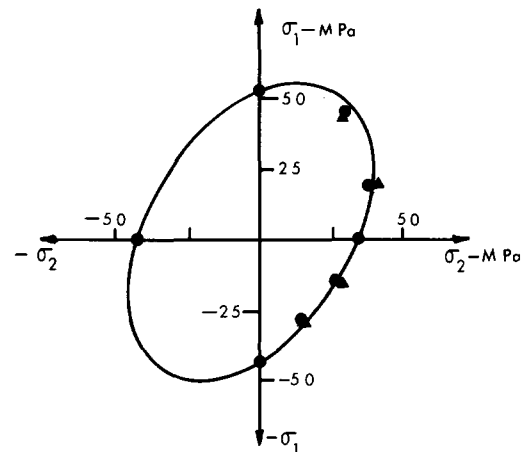


Fig. 13. A yield locus of oriented PC at 25 °C compared with results based on two methods of defining yielding: ●, effective strain definition; ▲, plastic work definition.

of the values of yielding given by the effective strain function. The discrepancy is larger for oriented PP. This might be expected since the shape of the locus for PP is very different from the von Mises ellipse; the PC locus has a shape more like that ellipse.

9. DISCUSSION OF RESULTS

Because of the manner in which the yield loci expand as the temperature is decreased

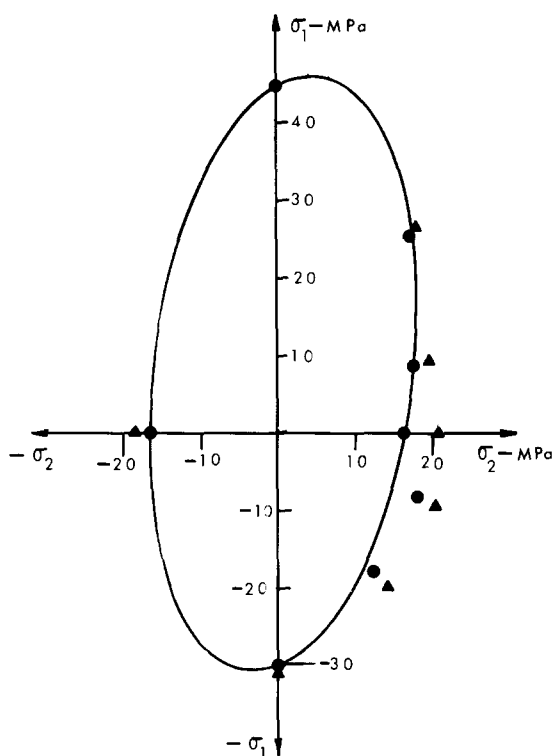


Fig. 14. Yield locus of oriented PP at 25 °C compared with results based on two methods of defining yielding: ●, effective strain definition; ▲, plastic work definition.

it is possible to use the normalized eqns. (6) and (12) to plot all data points for a given material on a single yield locus; Figs. 7 - 10 show such plots. In general the experimental points fit the theoretical yield loci well, with two exceptions. In Figs. 3, 7 and 11 the test using a stress ratio of 1.5 at 100 °C produces a point that lies well inside the yield locus; this test was repeated three times with almost identical results. As shown in Fig. 3, an additional test using hydraulic oil rather than silicone oil for internal pressurization led to an even greater discrepancy. Visual inspections showed clearly that crazing had occurred in all these cases. This possible mode of "yielding" has been pointed out by others [11]. It is surprising that the use of stress ratios of 1.0 and 2.0 with silicone oil and a temperature of 100 °C did not appear to cause crazing and the test points matched the theoretical locus almost perfectly. In addition, two tests with a stress ratio of 1.5 at 25 °C gave equivalent results that also fell on the theoretical locus. It is reasonable to conclude that the combination of high temperature and tensile stresses led to crazing, which was more

pronounced in the presence of hydraulic oil. Just why this should occur with a stress ratio of 1.5 and not with ratios of 1.0 and 2.0 remains unresolved.

The second disparity between experiment and theory concerns the stress ratio of -1.5 at all temperatures for the unoriented PP (see Figs. 4 and 8). All four points fell inside the predicted locus and buckling of the pressurized tube loaded in axial compression was suspected as the cause. Two analyses were conducted; the first indicated that Euler buckling did not occur. From suggestions by others [12, 13] it appeared that local surface buckling could be influential. To test this hypothesis a PP tube with an outer diameter of 23.7 mm and an inner diameter of 20.6 mm was subjected to a stress ratio of -1.5 at 25 °C to determine the significant values of σ_1 and σ_2 . Although the third principal stress σ_3 is no longer assumed to be zero, as in the approximation for thin-walled tube given in eqn. (14), its value was ignored in order to compare these findings with those shown in Fig. 8. This single test result is indeed closer to the theoretical locus, which supports the contention that the other tests were influenced by buckling. This problem of buckling during testing in the "fourth quadrant" of yield locus studies has been encountered by others [2, 5, 14, 15].

10. CONCLUSIONS

(1) Yield criteria proposed for unoriented and oriented polymers provide a most reasonable description of the yield behavior of both amorphous (PC) and crystalline (PP) polymers; in the latter case agreement was observed even through a glass transition region.

(2) For each material and condition of structure, a decrease in temperature caused a reasonably uniform expansion of the yield locus. This allowed the use of normalization and the results from different test temperatures for any one material could then be plotted on a single yield locus.

(3) In view of conclusion (2) it appears that the yield behaviors of PC and PP subjected to biaxial stress states can be predicted by first determining the tensile and compressive yield strengths as functions of temperature and by then determining the yield

strengths (in pertinent directions if the material is anisotropic) at any one convenient temperature. From such limited measurements critical normalizing parameters (see Table 1) can be readily computed for use in either eqn. (6) or eqn. (12).

(4) Crazeing or buckling may lead to yielding at stress states inside the yield locus of concern. However, such results seem to be the exception rather than the rule.

ACKNOWLEDGMENT

This material is based on work supported by the National Science Foundation under Grant DMR-74-10355. That support is gratefully acknowledged.

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