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Three-parameter yield criterion for a brittle polyester resin

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The tensile and compressive strengths of three polyester resins were measured under superposed hydrostatic pressure extending to 300 MPa, in an attempt to establish yield criteria. The polyesters were brittle in uniaxial tension at all pressures, and accordingly, a third testing geometry, diametral compression of a disc, was employed to complete the two or three necessary parameters to establish the yield surface in stress space. From the biaxial (disc) and axial compressive test data, the atmospheric tensile yield strength (higher than the fracture strength) was computed to be ~67 MPa in comparison with the compressive strength of ~120 MPa, their ratio 0.56 being significantly less than the more common 0.75 found for thermoplastics and epoxides. The data for compressive yield strength under superposed pressure were compared with the predictions of the two-parameter pyramidal, conical and paraboloidal criteria and the fit, though reasonable for the latter, could be significantly improved if a further independent material parameter was employed to give a three-parameter pyramidal criterion (the principal stresses σ_1, σ_2 and σ_3 being measured in MPa) of the form

$$0.0150\sigma_1 - 0.0039\sigma_2 - 0.0083\sigma_3 = 1$$

1. Introduction

Thermosetting polymers are usually brittle in tension, but show considerable ductility in compression. An epoxide resin [1] which fractured in uniaxial tension at a stress of 67 MPa, for instance, was found to yield in uniaxial compression at a stress of 119 MPa. These deformation processes operating in compression are suppressed in tension as "premature" fracture occurs due to material defects and surface flaws. This behaviour is also displayed by some thermoplastics. For polymers which do yield in tension, the tensile and compressive yield strengths are generally unequal. For many polymers, both thermoplastics and thermosets, the ratio of tensile to compressive yield strengths is about 0.75 [1]. This difference in the yield strengths shows that the hydrostatic component of the applied stress influences the yield process.

This has been demonstrated experimentally both by the direct effect of hydrostatic pressure on the yield behaviour of polymers [1–5] and by experiments in which the hydrostatic component of the stress tensor was varied [6, 7]. The general effect of hydrostatic pressure on the mechanical properties of polymers is to increase the yield stress and strain, and to increase the modulus at low stresses. Most work of this type has been carried out on thermoplastics, but similar behaviour has been noted for thermosets [1, 6, 8–10]. This dependence of yield behaviour on the hydrostatic component of stress is in contrast to the behaviour of metals and means that the one-parameter yield criteria used for metals, such as those of Tresca or von Mises, will not adequately represent polymer behaviour. Several hydrostatic pressure dependent two-parameter models for polymers have been proposed.

The Tresca criterion may be modified by making the critical shear stress, τ_T, a linear function of the hydrostatic component of the stress system [11]

$$\tau_T = \tau_T^0 + \mu_T P \qquad (1)$$

where τ_T^0 is the shear yield stress in pure shear, μ_T is a material constant, and the hydrostatic component of stress

$$P = (\sigma_1 + \sigma_2 + \sigma_3)/3 \qquad (2)$$

In three-dimensional stress space, the modified Tresca yield surface is an irregular hexagonal pyramid.

Two different modifications of the von Mises criterion have been suggested. Bauwens [12] and Sternstein and Ongchin [13] proposed independently that τ_M (critical shear yield stress) or τ_{oct} (octahedral shear stress) could be linear functions of the hydrostatic component

$$\tau_M = \tau_M^0 + \mu_M P \qquad (3)$$

$$\tau_{oct} = \tau_{oct}^0 + \tfrac{2}{3}\mu_M P \qquad (4)$$

τ_M^0 is the yield stress in pure shear ($P = 0$) and μ_M is a material constant.

Raghava *et al.* [14] suggested that a more general

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pressure-modified von Mises criterion was

$$(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 - 2(C - T)(\sigma_1 + \sigma_2 + \sigma_3) = 2CT \quad (5)$$

obtained by incorporating a hydrostatic component into the von Mises criterion. C and T are the absolute values of compressive and tensile yield stress.

The difference between these two modifications is that the Bauwens/Sternstein criterion predicts a linear dependence of yield stress on hydrostatic pressure, whilst the other gives a non-linear pressure dependence. It was claimed [14] that this is more physically likely, as the yield surface will be a paraboloid in three-dimensional stress space, rather than the cone with an angular apex predicted by Bauwens and Sternstein.

A third criterion of interest for polymers is the Mohr–Coulomb, originally suggested to account for failure in soils. It states that the critical stress for yielding to occur in any plane increases linearly with the pressure applied normal to the plane

$$\tau = \tau_C + \mu_C \sigma_N = \tau_C - \mu_C P_N \quad (6)$$

where τ_C is a constant, referred to as "cohesion" in soil mechanics, μ_C is the coefficient of friction, P_N is the normal pressure on the plane and σ_N is the normal stress on the plane. The criterion predicts the stress for yielding and also the direction of yielding, which is directly related to μ_C. Both the modified Tresca and modified von Mises criteria assume the direction of yielding is that of the maximum shear stress. The locus of the Mohr–Coulomb criterion in plane stress (the $\sigma_2 = 0$ plane of stress space) is an irregular hexagon, in three dimensions a pyramid.

Work by Caddell *et al.* [15] on several thermoplastics showed very little difference when analysed in terms of the two modifications of the von Mises criterion [12–14]. They proposed that discrimination between the modifications could be achieved by using higher hydrostatic pressures, and by carrying out tests in the compression octant of stress space, and also suggested that differences would be more apparent for polymers with a ratio of compressive to tensile yield strengths greater than 1.5.

There is conflicting evidence reported by other workers. Many have found a linear variation of yield strength with hydrostatic pressure for a variety of polymers, whilst others [16] have found an initially linear relation, then a diminishing rate of increase at higher pressures.

The observed deviation of deformation bands from the direction of maximum shear, predicted by both the Tresca and von Mises criteria, can be interpreted using a Mohr–Coulomb criterion [6, 17], but Li and Wu [18] suggest it to be not sufficient alone. Ward [17] also accepted the use of the Mohr–Coulomb relation as a polymer yield criterion, but doubted whether it was capable of dealing fully with all situations. Paul [19] also supported the use of a Mohr–Coulomb criterion but pointed out that a limitation is its independence of the intermediate principal stress, σ_2. To overcome these difficulties, Paul [19] and Li and Wu [18] have suggested three-parameter yield criteria. Paul [19] proposed a generalised pyramidal yield criterion

$$X\sigma_1 + Y\sigma_2 + Z\sigma_3 = 1 \quad (7)$$

The simplest surface of this type is a single hexagonal pyramid. For the two-parameter Mohr–Coulomb criterion $Y = 0$, $X = 1/\sigma_{yt}$, and $Z = 1/\sigma_{yc}$. Two-parameter criteria are usually expressed in terms of the uniaxial compressive and tensile yield strengths, σ_{yc} and σ_{yt}, respectively. Li and Wu [18] suggested an alternative pyramidal criterion

$$|\tau| + \alpha\sigma_N + \beta\sigma_H = \tau_0 \quad (8)$$

where τ is the shear stress, and α, β and τ_0 are constants.

Wronski and Pick [1] compared the three two-parameter yield criteria and a three-parameter Mohr–Coulomb pyramid with experimental data from tensile, compressive and shear loading of epoxides under superposed hydrostatic pressure. They proposed that the third parameter could be the equibiaxial compressive yield stress, σ_{cc}. The three-parameter criterion becomes

$$\frac{P_1}{T} + \frac{E - C}{EC} P_2 - \frac{P_3}{C} = 1 \quad (9)$$

where P_1, P_2, P_3, are the normalized stresses in terms of the uniaxial compressive stress and $E = |\sigma_{cc}/\sigma_{yc}|$. They found that the three-parameter criterion was similar to the two-parameter criteria in plane stress but differences were more pronounced in biaxial and triaxial compression, where the pyramidal criterion was the most conservative and the only one to lie near the experimental data. The conical and paraboloidal models diverged from the data with increasing hydrostatic pressure. They concluded that the pyramidal was the best of the two-parameter criteria for the epoxides tested, but the three-parameter criterion fitted the data slightly better.

This paper compares the three commonly used two-parameter criteria and a three-parameter criterion for an unsaturated polyester, which was brittle in uniaxial tension. Conventional tensile, compressive and shear testing is therefore inadequate if three independent parameters are to be evaluated. Biaxial stressing was attained by using the diametral compression of disc test geometry. A further independent experimental variable was still necessary, and hydrostatic pressure was superposed on the compressive and tensile tests. It is to be noted that brittle-to-ductile transitions in uniaxial tension of brittle polymers have been reported with the application of increasing hydrostatic pressure [1, 9].

2. Experimental procedure

The resin chosen for this investigation was Stypol 40-1077. This resin is used to manufacture fibre-reinforced composites and accordingly contained all the additives necessary for that purpose, such as filler and release agent. Some unfilled resin was also made available to investigate the effect of these additives.

Study of the mechanical properties of Stypol 40-1077 formed part of a wider investigation of the mechanical properties of glass–polyester composites (fully reported in [19–21]) in which two other (matrix) resins were mechanically tested [19]. One was a variant of Stypol 40-1077 (different catalyst) and the other "Beetle" 811 (probably not fully cured). Where relevant, reference to these resins will be made.

The resins were cured in 7 mm diameter glass tubes to give cylindrical rods. Tension and compression specimens were then machined. The tensile specimens were cylindrical, with a gauge length of about 10 mm and a gauge diameter of about 2 mm. The compression specimens were cylindrical, 10 mm high, with a diameter of 5 mm. Disc specimens, 5 mm diameter, d, and 3 mm thick, t, were also prepared for the test in diametral compression. The stress system, due to load P, in terms of the principal stresses, is then

$$\sigma_1 = \sigma_D = \frac{2P}{\Pi dt}, \quad \sigma_2 = 0, \quad \sigma_3 = -3\sigma_D \tag{10}$$

Tensile and compressive tests were also carried out under superposed hydrostatic pressures extending to 250 MPa, in a Coleraine pressure cell attached to a Hedeby universal tester. Details of the testing procedure have been given elsewhere [1, 20, 21]. Compression tests were conducted at crosshead speeds of 0.05 and 0.1 mm min^{-1}, and all tensile and diametral compression tests at 0.05 mm min^{-1}. The testing speed is indicated for each set of results where relevant.

To establish the effect, if any, of the pressurizing medium, "Plexol", a synthetic diester, some test specimens (especially of "Beetle" 811 which exhibited unexpected behaviour) were coated with a rubber solution to prevent the ingress or chemical interaction between the diester and polyester. No significant effects (unlike the situation for some composites where ingress took place along the fibre–resin interfaces) were detected [20].

3. Results

The two variants of polyester resin Stypol 40-1077 yielded in compression with a pre-failure load maximum. The nominal stress at this maximum load was taken to be the yield stress. ("Beetle 811" did not show a load maximum, but the load rose linearly, deviated and then continued to rise linearly at a much slower rate. The nominal yield stress was obtained by extrapolating the two approximately straight portions of the curve and calculating the stress at the intersection [20].) All resins showed an increase in yield stress, σ_y, with increasing superposed hydrostatic pressure.

The compressive yield stress of the first Stypol variant, resin 1, tested at 0.05 mm min^{-1} was 120 $\pm$ 1 MPa at atmospheric pressure, and it rose linearly with superposed pressure, $-H$, attaining 201 $\pm$ 13 MPa at $H = -250$ MPa, i.e. with a slope of 0.33 H (Fig. 1). The unfilled resin 1 had a yield stress of 127 $\pm$ 1 MPa at atmospheric pressure. The mechanical behaviour of the other Stypol variant (resin 2)

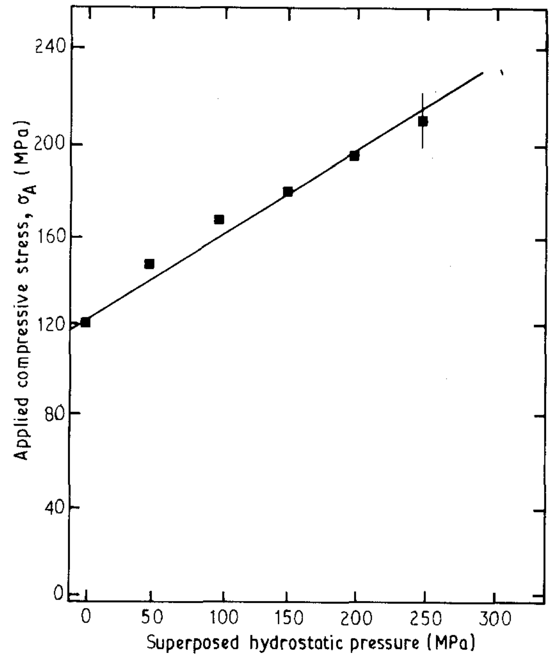

Figure 1 Applied stress at yield for polyester resin 1 specimens, tested under superposed hydrostatic pressure. (Standard deviations of less than $\pm$ 3 are not shown.)

was very similar at both test rates, e.g. atmospheric yield stress of 113 $\pm$ 3 MPa (crosshead speed of 0.1 mm min^{-1}) rose to 206 $\pm$ 5 MPa at $H = -250$ MPa, i.e. with a slope of 0.42H [19]. The yield stress for "Beetle 811" (resin 3) at atmospheric pressure was 77 $\pm$ 2 MPa (for a crosshead speed of 0.1 mm min^{-1}). This increased linearly to 101 $\pm$ 5 MPa at a superposed pressure of 150 MPa. Under superposed pressures greater than 150 MPa, the applied stress at yield was approximately constant with increasing pressure. At pressures below 150 MPa the applied stress at yield increased by 0.16 H [20].

Change in resin modulus and yield strain with increasing pressure could only be estimated. At 250 MPa superposed pressure the modulus change was approximately 25% for resin 1 and 20% for resin 2, and the increase in yield strain was about 65% for resin 1 and 50% for resin 2. (There was no apparent effect of hydrostatic pressure on the modulus and strain to yield of resin 3.)

The atmospheric tensile (fracture) strength of resin 1 was 54 $\pm$ 4 MPa, that of resin 2 was 44 $\pm$ 5 MPa. The applied tensile stress at failure for resin 1 rose approximately linearly to 86 $\pm$ 3 MPa at 200 MPa superposed pressure, i.e. by approximately $-0.16H$ (Fig. 2). All test specimens failed before any well-defined yield point could be detected. Although the loading curves showed some non-linearity [20] the failures were considered to be brittle. For resin 2 the pressure range was extended to 300 MPa, and the strength was observed to rise to $\sim$ 63 MPa, i.e. with a slope of $-0.05H$. The tensile strengths at failure for resins 1 and 2 were calculated from the maximum load and the cross-sectional area before loading. In contrast to these results, the loading curves of resin 3 specimens showed a clear deviation from linearity at atmospheric pressure, and it was possible to determine

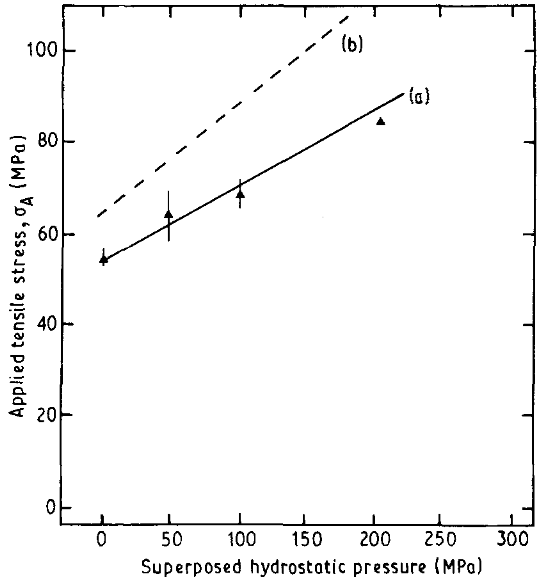

Figure 2 (a) Applied tensile stress at failure for polyester resin 1 specimens tested under superposed hydrostatic pressure. (Standard deviations of less than ± 3 are not shown.) (b) The hypothetical yield stress (in uniaxial tension), calculated according to the three-parameter pyramidal yield criterion, and its pressure dependence.

both this stress and that at maximum load. These values were 23 ± 4 and 31 ± 5 MPa, respectively. The applied tensile stress, σ_A, at fracture remained approximately constant [20] at the three pressures, 50, 100 and 150 MPa for which data were obtained. The loading curves appeared to become more linear with increasing pressure, and it was not possible to determine a stress at deviation (yield) at higher pressures.

The results of the experiments have been presented so far in terms of the applied stress, σ_A. For uniaxial tension, the principal tensile stress, under superposed hydrostatic pressure, $-H$, is $\sigma_A + H$. It should be noted that this maximum principal tensile (fracture) stress decreased with increasing pressure, and at pressures exceeding ~ 50 MPa it became compressive, such that all the principal stresses, though unequal, were compressive. Nevertheless, specimen failures were extensile; the appearance of the fracture surfaces, for example Fig. 3, at all pressures was similar to that at atmospheric pressure, where a Griffith-type mechanism is generally thought to operate. Failure origins were located at the specimen's surface, but no evidence of fluid ingress prior to failure was detected.

4. Discussion

The brittle fracture strength pressure dependence data are inconsistent with critical tensile strength, Griffith and critical strain energy criteria [23]. If the variation of the tensile modulus with hydrostatic pressure ($< 25\%$ in our experiments) is neglected, a linear dependence of σ_A on H is predicted with a slope of $2\nu - 1$, where ν is the Poisson's ratio of the polyester. The Poisson's ratio for polyesters is ~ 0.39, and thus the resultant value of -0.22 for resin 1 compares favourably with the observed value of -0.16. The pressure dependence of resin 2 was weaker ($-0.05\,H$), and if it is assumed to be independent of hydrostatic pressure, the data are then consistent with the deviatoric tensile stress or strain criteria [23]. Taking account of the pressure dependence of the tensile modulus does not markedly change these conclusions, which are in accord with data on, for example, an epoxide [24], where additionally, some effect of coating on the fracture strength under superposed pressure was noted.

The most general form of the three-parameter yield criterion is given by Equation 7 [19]. Constants X, Y and Z were calculated from the data for resin 1 tests. Because the generalized pyramidal criterion contains three independent variables, three independent equations are needed for their calculation. These equations, *if* yielding takes place in the three testing geometries used, are as follows.

From tests in uniaxial tension

$$X\sigma_T = 1 \tag{11}$$

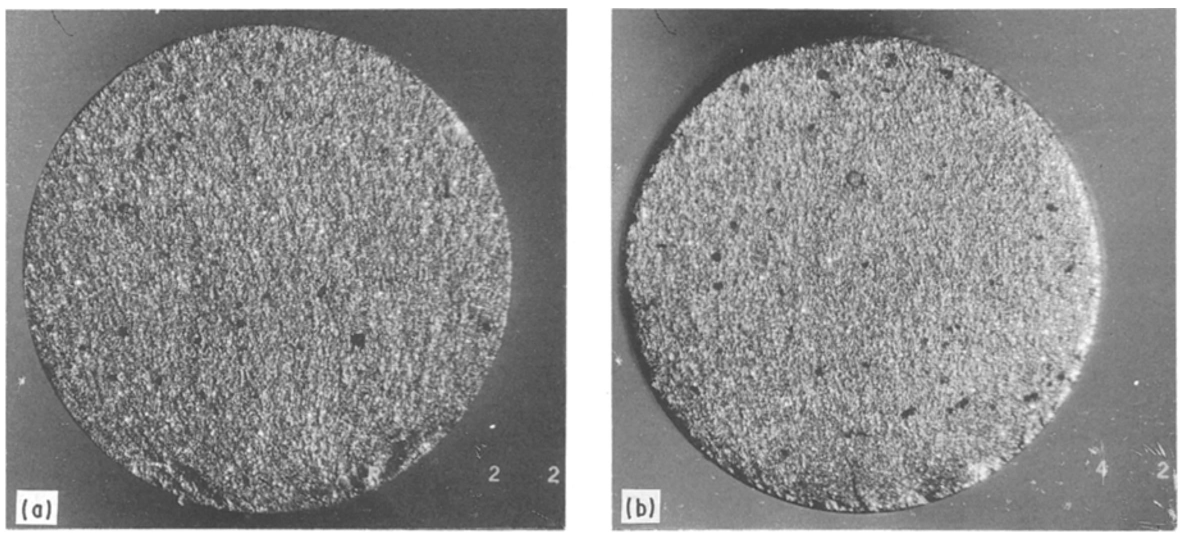

Figure 3 Fractographs of tensile specimens of resin 1, which failed (a) at atmospheric pressure, maximum principal stress of 52 MPa, and (b) under superposed pressure of 200 MPa, maximum principal stress of -88 MPa. Note extensile (cracking) failure mode operating in both cases.

From uniaxial compression

$$Z\sigma_C = 1 \quad (12)$$

From diametral compression

$$X\sigma_D - 3Z\sigma_D = 1 \quad (13)$$

The compressive yield strength for resin 1 at atmospheric pressure at a crosshead speed of $0.05\,mm\,min^{-1}$ was -120 MPa (as compressive stresses are negative). Substituting this value into Equation 11 gives:

$$Z = 1/\sigma_C = -1/120 = -0.0083\,MPa^{-1} \quad (14)$$

X cannot be determined directly from Equation 11 because resin 1 was brittle in tension. Thus, to determine X, use was made of the diametral compression test data at atmospheric pressure (Equation 13). Tests were carried out at a crosshead speed of $0.5\,mm\,min^{-1}$. The loading curves showed a deviation from linearity at a mean stress of 25 MPa, and specimens failed, in a brittle manner at a mean stress of 42 MPa. If it is assumed that the stress at deviation corresponds to the yield stress and is relatively independent of the testing speed (for a change of a factor of 10), then $X = 0.0150\,MPa^{-1}$. To evaluate the remaining constant, Y, data from the tests in compression under superposed hydrostatic pressure were used. The general form of the expression is

$$XH + YH + Z(H + \sigma_C) = 1 \quad (15)$$

where H is the superposed pressure. This predicts a linear relationship between σ_A and H of $(X + Y + Z)/Z$, and accordingly, from the slope of -0.33 (Fig. 1), Y evaluates to $-0.0039\,MPa^{-1}$.

The pyramidal yield criterion for resin 1 is thus given by

$$0.0150\sigma_1 - 0.0039\sigma_2 - 0.0083\sigma_3 = 1 \quad (16)$$

To check consistency with the data obtained in tension, the tensile yield strength can be calculated, but as resin 1 failed in a brittle manner in tension, the predicted yield strength will be "hypothetical". From Equation 11, $\sigma_T = 1/X = 66.7$ MPa. This value for the "hypothetical" tensile yield strength is greater than the observed atmospheric pressure brittle fracture stress of 54 MPa, and so the three-parameter pyramidal yield criterion is not inconsistent with the observed data. For this polyester the ratio of tensile to compressive yield would appear to be smaller than has been found for many other polymers, at about 0.56. The pressure dependence of the tensile yield strength may also be predicted from the three-parameter criterion, $X\sigma_1 + (Y + Z)H = 1$, i.e.

$$\sigma_1 = 67 + 0.773H \quad (17)$$

and this relationship is also plotted in Fig. 2. It is seen that the predicted values of the tensile yield stress are always higher than the observed brittle fracture stresses. The observed pressure dependence of the brittle fracture stress and the predicted yield stress dependence should also be noted. The two plots do not intersect, consistent with the observation that

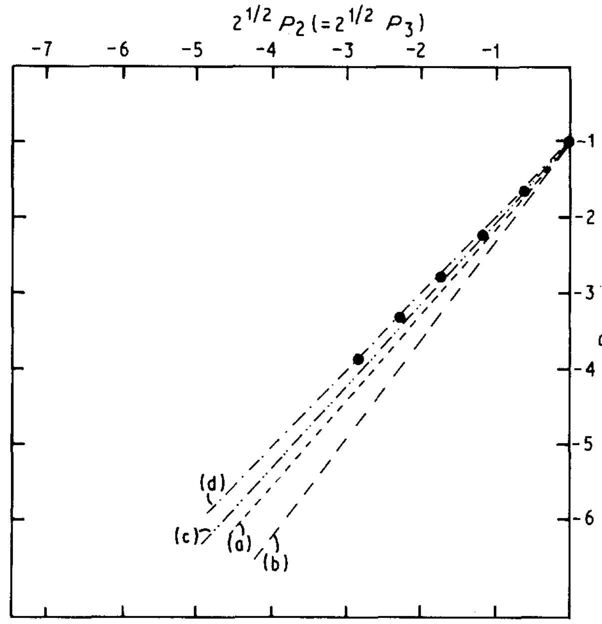

Figure 4 Plots of two-parameter compressive yield under superposed hydrostatic pressure criteria in normalized stress space: P_1, P_2, P_3. (The relevant plane is $P_2 = P_3$.) The yield envelopes were calculated according to the (a) conical, (b) two-parameter pyramidal and (c) two-parameter paraboloidal criteria based on the compressive and 'hypothetical' tensile yield stresses of polyester resin 1. (d) The experimental points and the best-fit three-parameter pyramidal criterion.

a brittle/ductile transition with increasing pressure is not observed in this resin.

The three-parameter criterion can be compared with the three commonly used two-parameter criteria, by drawing their yield envelopes in the compression quadrant of stress space, and then comparing them with the data obtained for resin 1 in compression under superposed hydrostatic pressure. It has been pointed out by Paul [19] that if σ_1 is plotted against $2^{\frac{1}{2}}\sigma_2 (= 2^{\frac{1}{2}}\sigma_3)$ the relevance of triaxial testing to yield surface determination is stressed. The data for resin 1 are plotted in Fig. 4 in normalized stress space, along with the yield envelopes for the three two-parameter and one three-parameter criteria. The three-parameter pyramid appears to fit best the experimental data for resin 1, and the best two-parameter criterion is the paraboloid. Three-parameter criteria for resins 2 and 3 were not independently derived, but if normalized compressive yield stresses are plotted they lie close to the resin 1 data [20]. For these polyesters, of the simple two-parameter criteria, the best correlation with experimental data is with the paraboloid. However, if a three-parameter hexagonal pyramidal yield surface is considered, the fit is improved.

Similarities in the yielding behaviour of the polyesters with epoxies and thermoplastics are thus to be noted [1, 18]. Noteworthy differences are the (predicted) low tensile/compressive yield ratio, < 0.6, compared to the more common ~ 0.75, continued brittle behaviour of the polyester resins under superposed pressures extending to 300 MPa, and the behaviour of "Beetle 811". This (possibly undercured) resin either shows a transition in compressive behaviour with increasing pressure or resembles polyoxymethylene as reported by Sardar [16]. The data further indicate that, should polymers be used under complex loading (the most common examples being pipes or pressure vessels), it is important to ascertain – for each material – the actual yielding criterion and consider the relative difficulties of yielding and fracture.

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