Fractography and Failure Mechanisms of Particulate-Filled Thermoplastic Polyester

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SYNOPSIS

Fractography has been used for the postfailure analysis of a filled thermoplastic polyester. The five fracture modes that were previously defined on the basis of macroscopic stress–strain behavior were distinguished by certain fractographic features. These features were characteristic of the fracture mode and did not depend on filler type or filler content. The Mode A ductile fracture surface consisted of two regions: a pullout region of slower crack growth and a rosette region of faster crack growth. The Mode B ductile fracture surface contained only a ductile pullout texture. The Mode C quasi-brittle fracture surface exhibited secondary fracture features that sometimes included the herringbone pattern. The Mode D quasi-brittle fracture surface consisted of a stress-whitened dimple region and a brittle fracture region. The Mode E fracture surface exhibited primarily the rough texture characteristic of brittle fracture. The failure mechanisms inferred from analysis of the fracture surfaces confirmed a microscopic failure model of the ductile-to-quasi-brittle transition in filled PETG that is based on the strain-hardening strength of the polymer ligaments between debonded filler particles. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

Previous studies of filled thermoplastic polyester described the ductile-to-quasi-brittle transition that occurs with increasing filler content.[1,2] The transition was accompanied by a sharp drop in fracture strain when there was a change from fracture during strain-hardening or neck propagation to fracture during neck formation. A simple model based on a microscopic failure condition was used to predict the ductile-to-quasi-brittle transition. In this model, the critical filler content was determined by the strain-hardening strength of the polymer ligaments between debonded filler particles.

The previous studies also led to the identification of five fracture modes based on the macroscopic stress–strain behavior: Modes A and B were ductile fracture modes where fracture occurred during strain-hardening (Mode A) or neck propagation (Mode B), Modes C and D were identified with quasi-brittle fracture during neck formation, whereas Mode E was the brittle fracture mode.

Various ductile fracture modes have been described for polymeric materials from the characteristic features of the fracture surface.[3,4] Since it is often possible to infer microscopic failure mechanisms from analysis of fracture surfaces, a fractographic analysis was made of the five fracture modes of filled thermoplastic polyester. The analysis was used to test the validity of the microscopic failure model of the ductile-to-quasi-brittle transition.

MATERIALS AND METHODS

The polymer used in this study was an amorphous copolyester (PETG) from Eastman Kodak (Kodar Copolyester 6763). Two types of fillers were used: an organic filler, calcium terephthalate (CaT), and an inorganic filler, calcium carbonate ($CaCO_3$). Three $CaCO_3$'s and two CaT's were used in the study; they differed in particle size and size distribution. The fillers were described previously, as were

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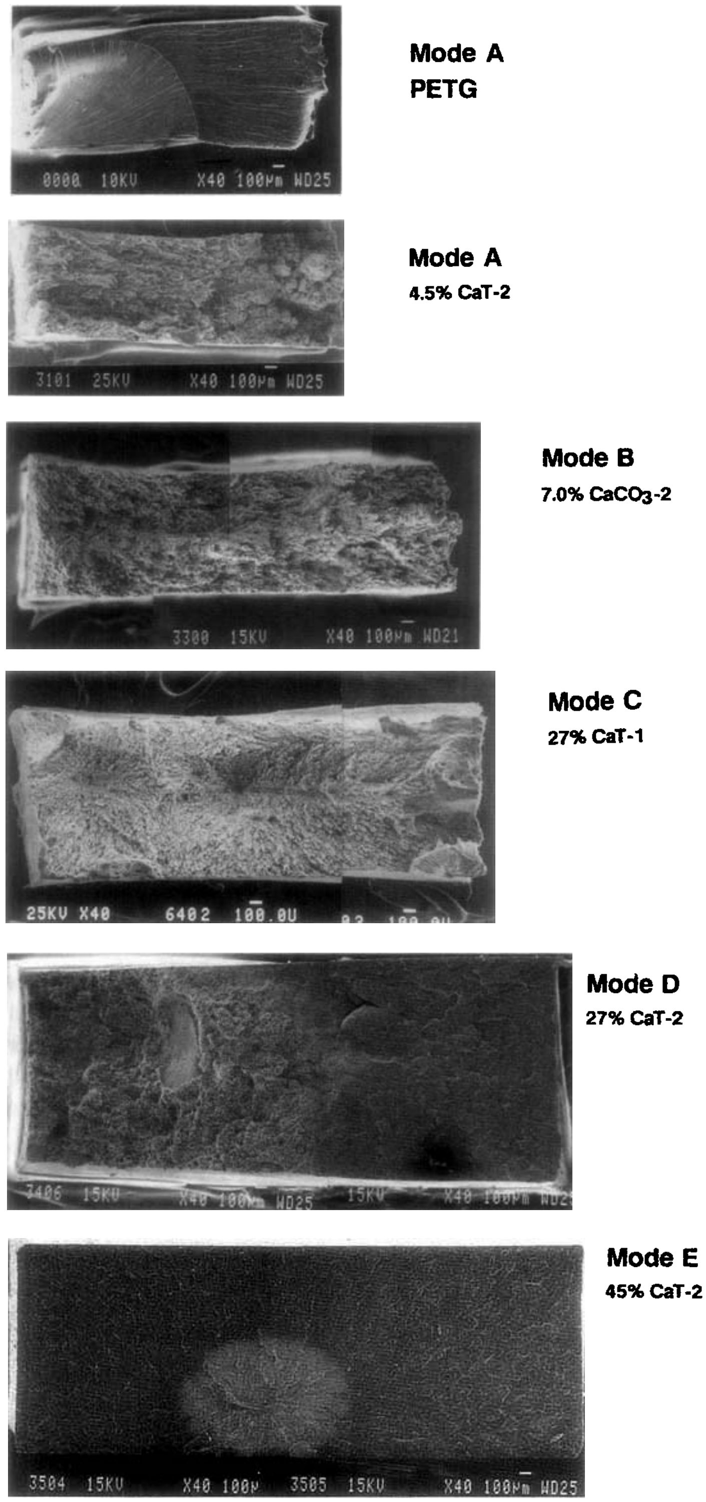


Figure 1 Typical fracture surfaces. Mode A fracture surfaces of PETG and PETG with 4.5 vol % CaT-2, Mode B fracture surface of PETG with 7 vol % $CaCO_3$-2, Mode C fracture surface of PETG with 27 vol % CaT-1, Mode D fracture surface of PETG with 27 vol % CaT-2, and Mode E fracture surface of PETG with 45 vol % CaT-2.

the blending conditions and the compression-molding conditions.[2] The PETG was blended with 5, 15, 30, and 50 wt % of filler, which corresponded to 4.5, 13.5, 27, and 45 vol % CaT and 2.4, 7.0, 14, and 24 vol % $CaCO_3$.

Tensile specimens were cut from compression-molded plaques according to the ASTM D1708 geometry. The specimens were fractured in tension in an Instron testing machine with a crosshead speed of 2 mm/min, which corresponded to a strain rate of 9% per minute. Four specimens were tested for each composition. Some of the tensile fracture surfaces were coated with 90 Å of gold and examined in the JEOL JSM-840A or JEOL JSM-35CF scanning electron microscope (SEM). Others were subsequently fractured in liquid nitrogen through the thickness parallel to the loading direction to reveal the internal morphology. The cryogenic fracture surfaces were coated with 90 Å of gold and the region close to the fracture surface examined in the SEM.

RESULTS AND DISCUSSION

Fracture Behavior

Five fracture modes were previously described for filled PETG on the basis of the macroscopic stress–strain behavior.[2] At low filler content, a stable neck formed and the filled PETG subsequently fractured during strain-hardening (Mode A) or during neck propagation (Mode B). Both Modes A and B were considered to be ductile fracture modes. As the filler content increased, quasi-brittle fracture occurred during neck formation while the engineering stress dropped from the upper yield stress to the draw stress (Modes C and D). Mode C fracture occurred

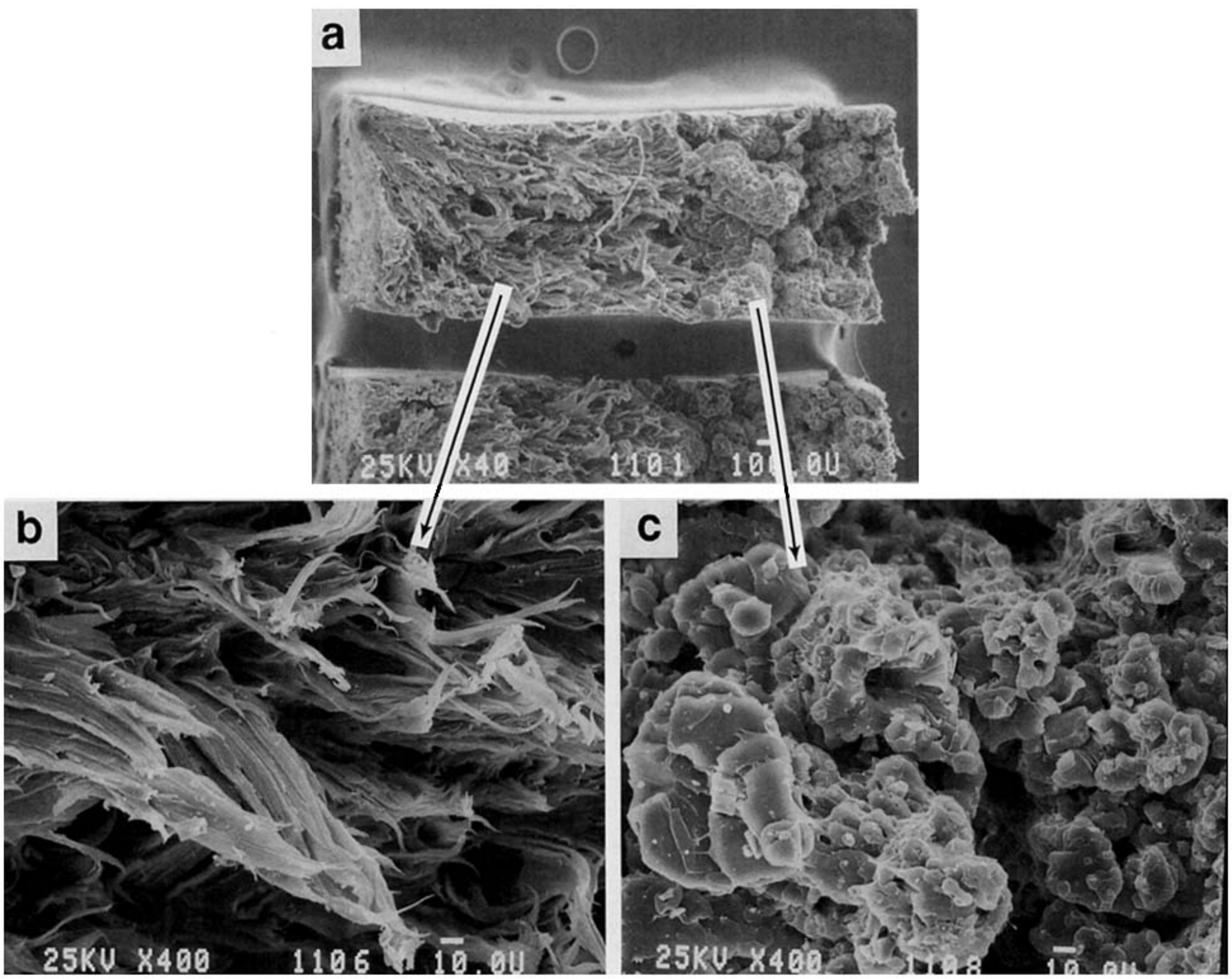


Figure 2 Mode A fracture surface of PETG with 2.4 vol % $CaCO_3$-2: (a) the entire fracture surface; (b) higher magnification of the pullout region; (c) higher magnification of the rosette region.

through the thin region at the site of incipient neck formation when the stress had dropped almost to the draw stress. In Mode D, specimens fractured shortly after the yield maximum on the stress–strain curve and fracture occurred through the macro-shearband that formed across the specimen in the beginning of the necking process. With further increase in filler content, specimens fractured in a brittle manner perpendicular to the loading direction before reaching the yield point (Mode E). The ductile-to-quasi-brittle transition was accompanied by a sharp drop in fracture strain when the fracture mode changed from Modes A and B, when specimens fractured during strain-hardening or neck propagation, to Modes C and D, when specimens fractured during neck formation. This transition was always observed with increasing filler content.

Typical fracture surfaces of each mode are shown in Figure 1. Since the initial cross-sectional area was always the same, the differences in the area of the fracture surfaces reflected the changes in strain at the fracture site as the fracture mode changed from Mode A to Mode E. The Mode A fracture surfaces had the smallest cross-sectional area since fracture occurred at the largest strains in the strain-hardening region of the stress–strain curve. In comparison to Mode A, the Mode B fracture surface had a somewhat larger cross-sectional area because this

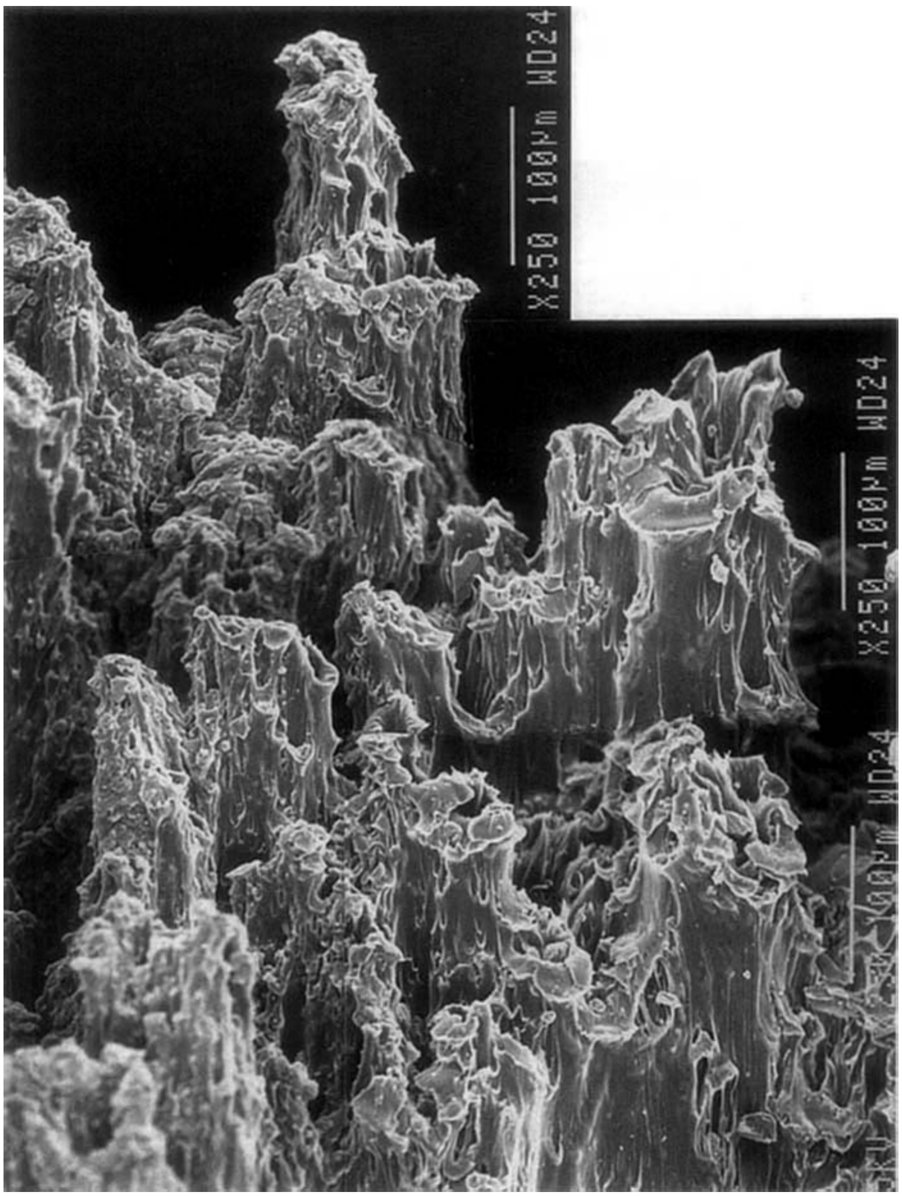


Figure 3 A side view of the rosette region on the fracture surface of PETG with 2.4 vol % $CaCO_3$-2.

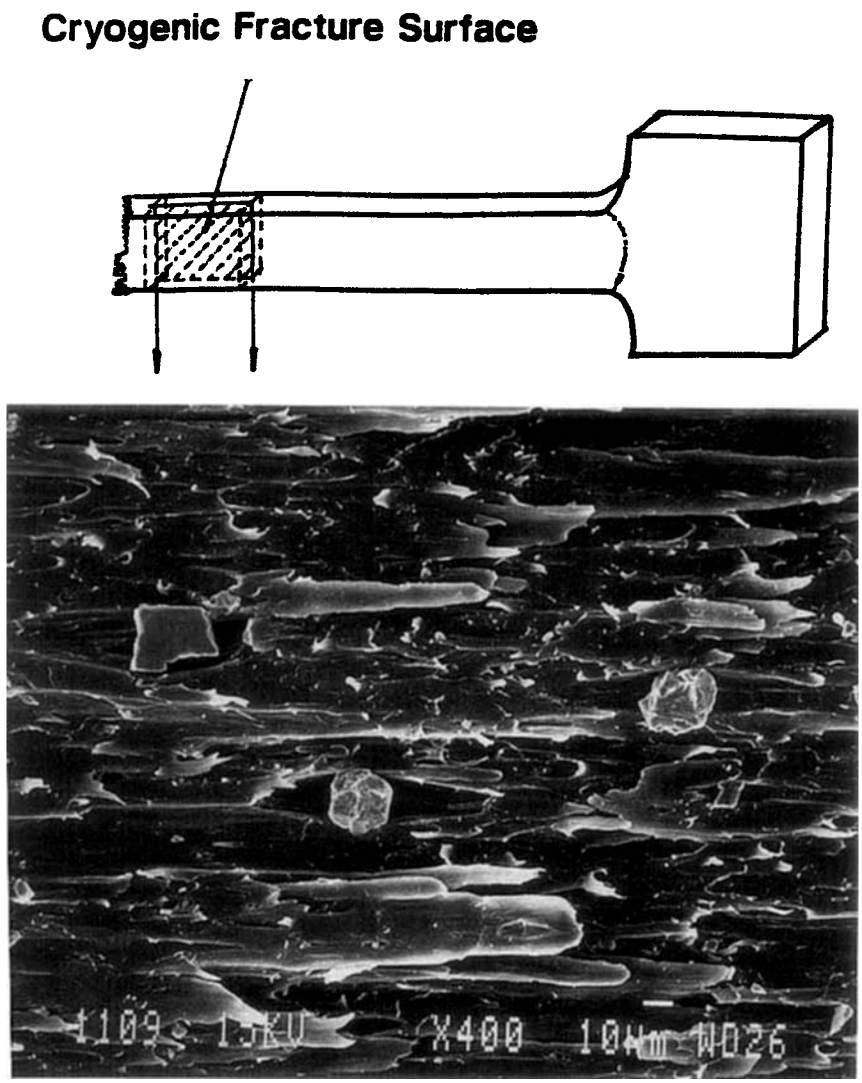


Figure 4 The necked region of PETG with 2.4 vol % $CaCO_3$-2 close to the fracture site. The specimen was cryogenically fractured parallel to the loading direction.

specimen fractured in the neck during neck propagation without experiencing the additional reduction in cross section that occurred during strain-hardening. Filled PETG with Mode C fractured in the thinned region when neck formation was almost completed. As a result, the cross-sectional area was similar to that of Mode B. The fracture surface of Mode D was somewhat thinned at one end where fracture propagated through the macro-shearband, while the width of the remaining surface was unchanged. For brittle Mode E fracture, the cross-sectional area was essentially the same as the undeformed specimen. The major features of the filled PETG fracture surfaces were the same, regardless of filler or filler content, as long as the fracture mode was the same.

Mode A fractures initiated from one edge and the resulting fracture surfaces consisted of two regions: a more ductile region of slower crack growth that included the fracture initiation site and a more brittle region of faster crack growth. A distinct boundary separated the two regions. The primary difference in the appearance of the PETG and filled PETG Mode A fracture surfaces was the profuse stress-whitening of the filled PETG that was absent from PETG. The Mode A fracture surface of PETG in Figure 1 was relatively smooth; the concave slow crack growth region contained numerous tear lines that radiated outward from the initiation site. An abrupt change in surface texture from tearing to cleavage marked the transition to fast crack growth. The Mode A fracture surface of filled PETG also consisted of two regions, but cavitation resulted in surface textures that were somewhat different from those of unfilled PETG. The Mode A fracture surface of filled PETG consisted of a pullout region where the crack propagated by ductile tearing and a rough region with a rosette pattern that was created by a quasi-cleavage mechanism.

The Mode B fracture surface contained only a ductile pullout texture. Fracture initiated from either the edge or the interior of specimens. In the example in Figure 1, fracture initiated at the upper left corner, and the matrix material was pulled out in the crack propagation direction.

The Mode C fracture surface in Figure 1 had the characteristic spine and rib pattern called herringbone or chevron.[3,4] The main crack initiated in the center of the specimen. Repeated interaction of the main crack with secondary cracks that initiated along the center line created a series of ridges that pointed back toward the site of fracture initiation. Two herringbone regions were observed on the fracture surface in Figure 1: one that initiated in the center and a second that initiated near the upper left corner of the specimen.

The Mode D fracture surface consisted of two regions: a stress-whitened region and a brittle fracture region. The crack always initiated from the edge at one end of the macro-shearband that formed across the specimen at the beginning of neck formation and propagated through the shearband at an angle to the loading direction. As the crack speed increased, the crack path usually deviated from the shearband and proceeded in a direction that was more nearly perpendicular to the loading direction. In the example in Figure 1, fracture initiated at the left edge. Propagation through the macro-shearband resulted in the stress-whitened region that in this example extended about halfway across the fracture surface. The place where the crack path deviated from the shearband was clearly discerned by the boundary that marked the end of the stress-whitened region and the beginning of a brittle fracture region.

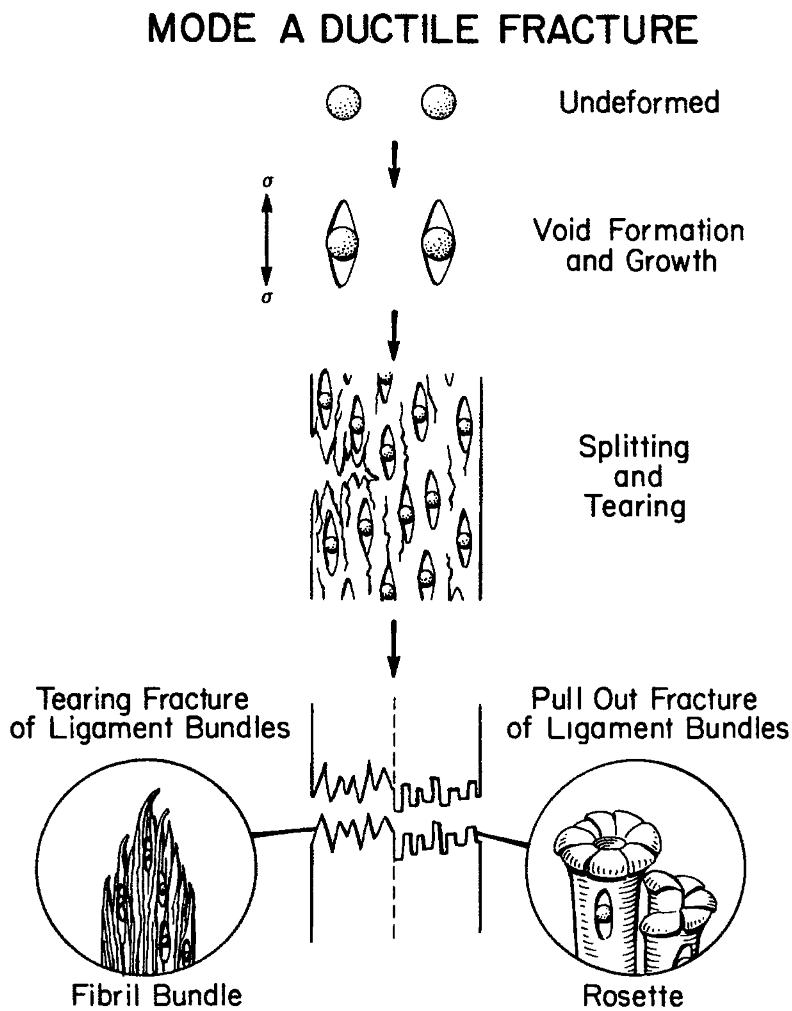


Figure 5 Schematic representation of Mode A fracture.

The otherwise featureless brittle fracture surface of Mode E contained a slightly stress-whitened region that included the flaw where fracture initiated. Aside from the slight stress-whitening, the texture of the crack initiation region in Figure 1 was the same as the rest of the brittle fracture surface.

Mode A Ductile Fracture

Strain-hardening enables PETG to sustain loads significantly larger than the draw stress. The amount of polymer in the cross section can be reduced to a certain level and the matrix is still able to support the external load without fracture. At a very low filler content, such as the compositions that exhibit Mode A behavior, the amount of matrix material in the ligaments between the filler particles is sufficient to support both neck propagation through the entire gauge length as well as some amount of strain-hardening. The tensile properties, such as fracture stress and fracture strain, are very close to those of unfilled PETG.

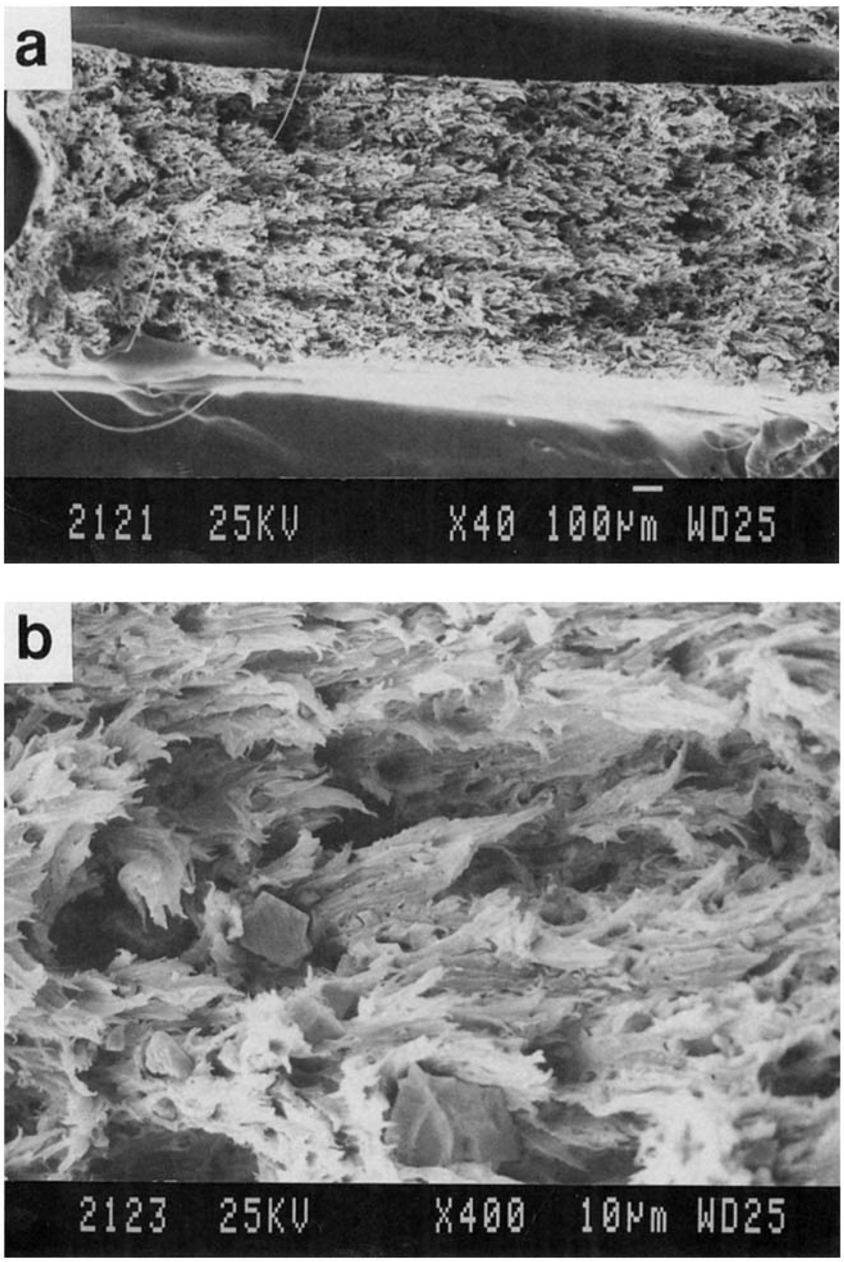
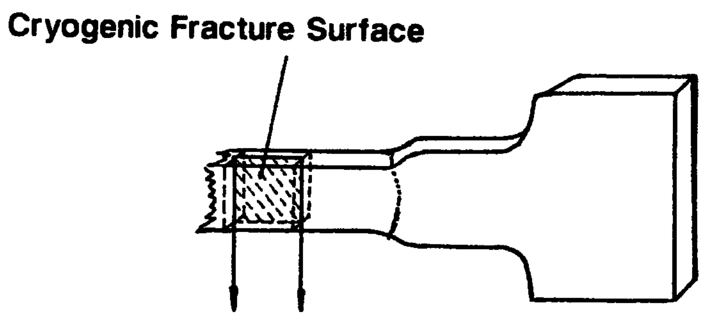


Figure 6 Mode B fracture surface of PETG with 7 vol % $CaCO_3$-2: (a) the entire fracture surface; (b) higher magnification.

Mode A tensile fracture surfaces always consisted of a pullout region and a quasi-cleavage rosette region. Figure 2(a) shows a Mode A fracture surface of PETG with 2.4 vol % $CaCO_3$-2. Fracture initiated at the upper left corner and proceeded through the pullout region. Highly drawn fibril bundles were observed in this region; the 30–100 μm bundles consisted of 2–5 μm fibrils [Fig. 2(b)]. Elongated voids that contained debonded particles were visible on the exposed sides of the fibril bundles. As the crack speed increased, the fracture mode changed abruptly near the center of the fracture surface to quasi-cleavage fracture [Fig. 2(c)]. Protrusions on one of the very rough fracture surfaces corresponded to holes on the matching surface. A side view of the protrusions showed that they fractured at many different levels (Fig. 3). A group of rosettes typically decorated the top of the 30–100 μm protrusions. Voids that often contained debonded particles were visible on the sides of the protrusions.

The necked region close to the fracture site of PETG with 2.4 vol % $CaCO_3$-2 was cryogenically fractured parallel to the loading direction to reveal the internal structure (Fig. 4). Particles were debonded from the matrix and elongated voids had formed around the particles with the long axis parallel to the loading direction. The length of the ellipsoidal voids was about two to three times the particle diameter, while the maximum width of the voids was equal to the diameter of the particles. In this composition, the amount of polymer in the ligaments between filler particles was large enough that the voids grew independently without interacting laterally with nearby voids.

The Mode A fracture process is schematically represented in Figure 5. Filler particles debonded from the matrix during yielding and voids formed around the particles as the polymer was plastically deformed during neck propagation. The width of the ligaments between the particles decreased during neck propagation, while the width of the voids remained constant and equal to the particle diameter.



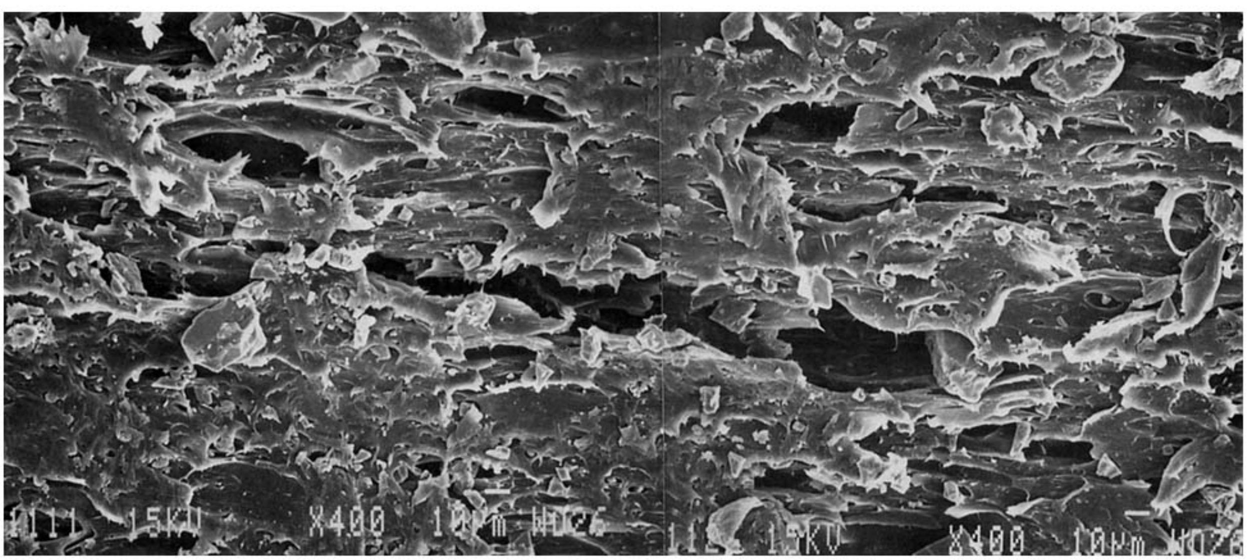


Figure 7 Necked region of PETG with 7 vol % $CaCO_3$-2 close to the fracture site. The specimen was cryogenically fractured parallel to the loading direction.

Because of the low filler content, the ligaments between voids were thick enough that they could undergo thinning during necking without fracturing, and thereby lateral coalescence of the voids was avoided. Close to the ultimate fracture strain, longitudinal splitting through the voids created bundles of ligaments. Mode A fracture occurred in the neck when the local strain in the ligaments reached the fracture strain of the polymer. Fracture started at the edge of the neck and, initially, the crack propagated slowly by a ductile tearing mechanism. The ligament bundles were pulled out into bundles of fibrils during tearing fracture. As the crack accelerated, fibrillation of the ligament bundles was replaced by a quasi-cleavage fracture mechanism. The rough fracture surface resulted when each ligament bundle fractured at a different level. The quasi-cleavage fracture mode produced a group of rosettes on top of each fractured ligament bundle. A single rosette formed when multiple microcracks initiated from one of the elongated voids. Impingement of the microcracks as they propagated away from the void produced the tear ridges that emanated outward from the void. Since each ligament bundle contained multiple voids that could be the initiation site of a rosette fracture, a group of rosettes was typically observed on the fractured ligament bundle. This kind of quasi-cleavage fracture requires a certain amount of matrix material around the void and was only observed with the lowest filler contents.

The rosette fracture of filled PETG was very similar to the classical rosette fracture of metals that is initiated by spherical voids.[5] Since the voids were elongated rather than spherical, the multiple microcracks that created the rosette could all initiate and grow out from the debonded filler particle in approximately the same plane or they could initiate on slightly different planes and spiral around the elongated void. Both types of rosette fracture can be observed in the quasi-cleavage region of the Mode A fracture surfaces in Figure 2(c) and Figure 3.

Mode B Ductile Fracture

Compositions that fractured during neck propagation at relatively large deformations were categorized as Mode B. These specimens fractured in the necked region before the neck had propagated through the entire gauge section and differed from Mode A specimens by not experiencing strain-hardening. Fracture could initiate either from the edge or from the center. Fracture of the specimen of PETG with 7 vol % $CaCO_3$-2 in Figure 6 initiated from the left edge. Mode B fracture surfaces did not contain a rosette fracture region; instead, a pullout texture covered the entire surface. The pulled-out fibril bundles were shorter and smaller in diameter than in Mode A, and the fibrils that composed the bundles were also thinner.

Cryogenic fracture of the neck parallel to the loading direction, near the tensile fracture site of PETG with 7 vol % $CaCO_3$-2, revealed highly elongated voids around the filler particles (Fig. 7). Coalescence of the voids in the longitudinal direction was more prevalent than in Mode A fractures and the resulting longitudinal cracks were larger, 100–200 μm in length and typically 10 μm in width. Occasionally, lateral coalescence of the voids was also seen as in Figure 7. This occurred in places where the ligaments between filler particles were especially thin and the ligaments fractured when they experienced further thinning during necking. Lateral coalescence of voids could have created a critical size flaw from which catastrophic fracture initiated.

Mode B failure is represented schematically in

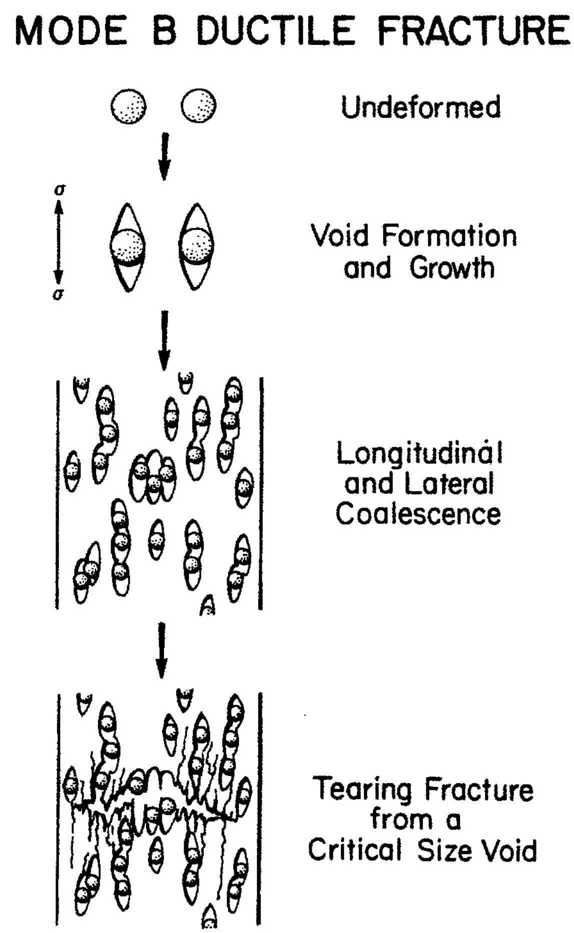


Figure 8 Schematic representation of Mode B fracture.

Figure 8. Filler particles were closer together than in Mode A, and as a result, the ligaments between particles were thinner. Most of the ligaments were thick enough to extend during necking without fracturing. The elongated voids coalesced longitudinally; the resulting longitudinal cracks were larger and more numerous than in Mode A. In some places, especially where the filler was not well dispersed,

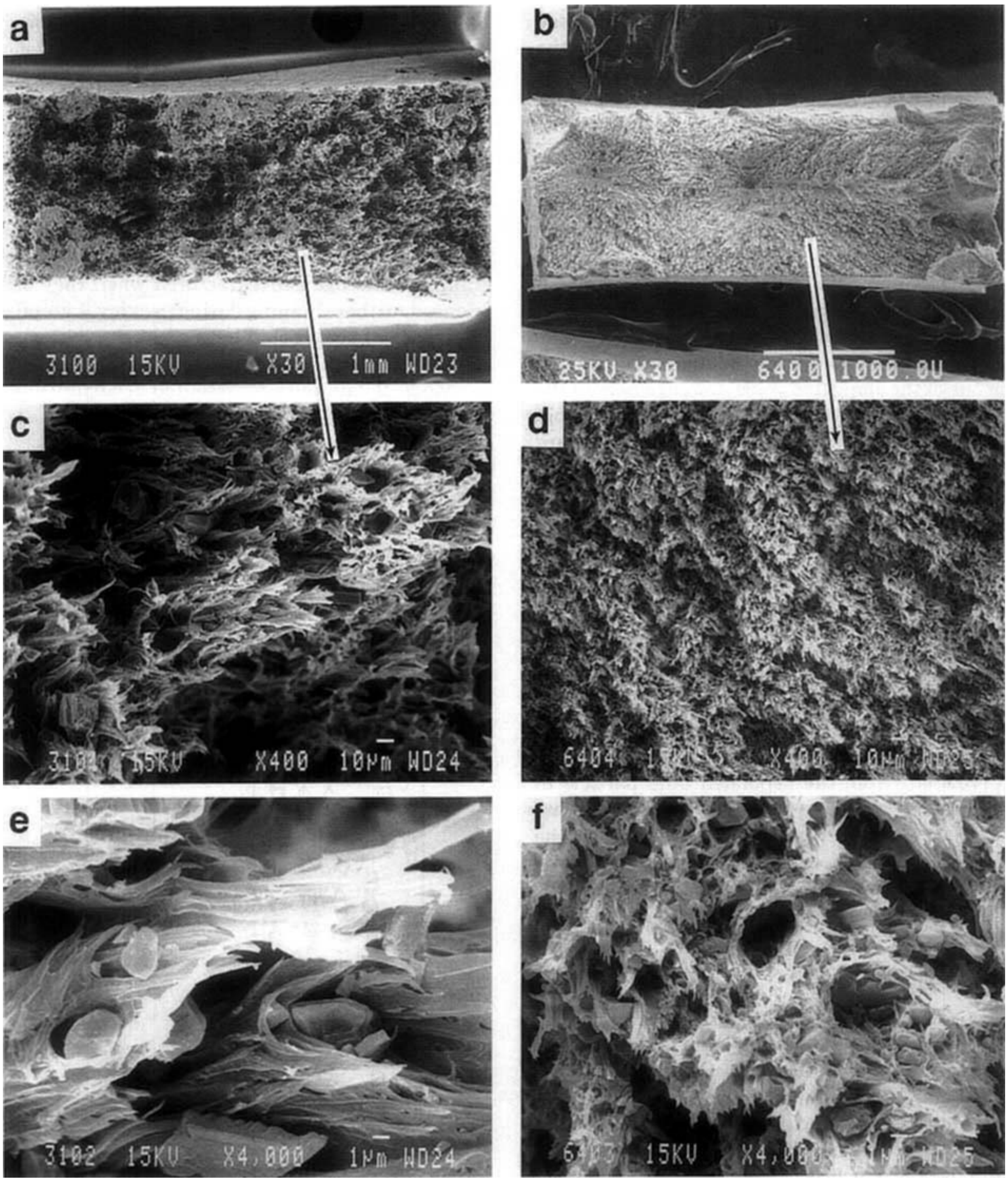


Figure 9 Mode C fracture surface: (a) the fracture surface of PETG with 14 vol % $CaCO_3$-2; (b) fracture surface of PETG with 27 vol % CaT-1; (c, e) higher magnifications of the fracture surface in (a); (d, f) higher magnifications of the fracture surface in (b).

the ligaments were sometimes thin enough that necking caused them to fracture. When this occurred, the voids coalesced laterally to create a larger void. In Mode B compositions, catastrophic fracture ensued when lateral coalescence of voids created a critical-size flaw. The critical flaw was a singularity that occurred when the local concentration of filler particles was unusually high; otherwise, there was enough matrix material in the cross section to support stable propagation of the neck. The critical flaw could have formed either in the center or near the edge. Subsequent crack growth through the necked material occurred by ductile tearing as in the initial phase of Mode A fracture, although the pulled-out fibril bundles were smaller and the individual fibrils were thinner.

Mode C Quasi-brittle Fracture

The change from Mode B to Mode C fracture marked the transition from ductile-to-quasi-brittle fracture. Quasi-brittle fractures occurred during necking. Specifically, Mode C was identified with fracture through the thinned region at the site of incipient neck formation. Secondary fracture was a characteristic feature of Mode C fracture surfaces. Sometimes, as in Figure 9(a) of PETG with 14 vol % $CaCO_3$-2, secondary fracture appeared more or less randomly. Other times, as in the example in Figure 9(b) of PETG with 27 vol % CaT-1, the presence of a herringbone pattern indicated that secondary cracks initiated sequentially along the

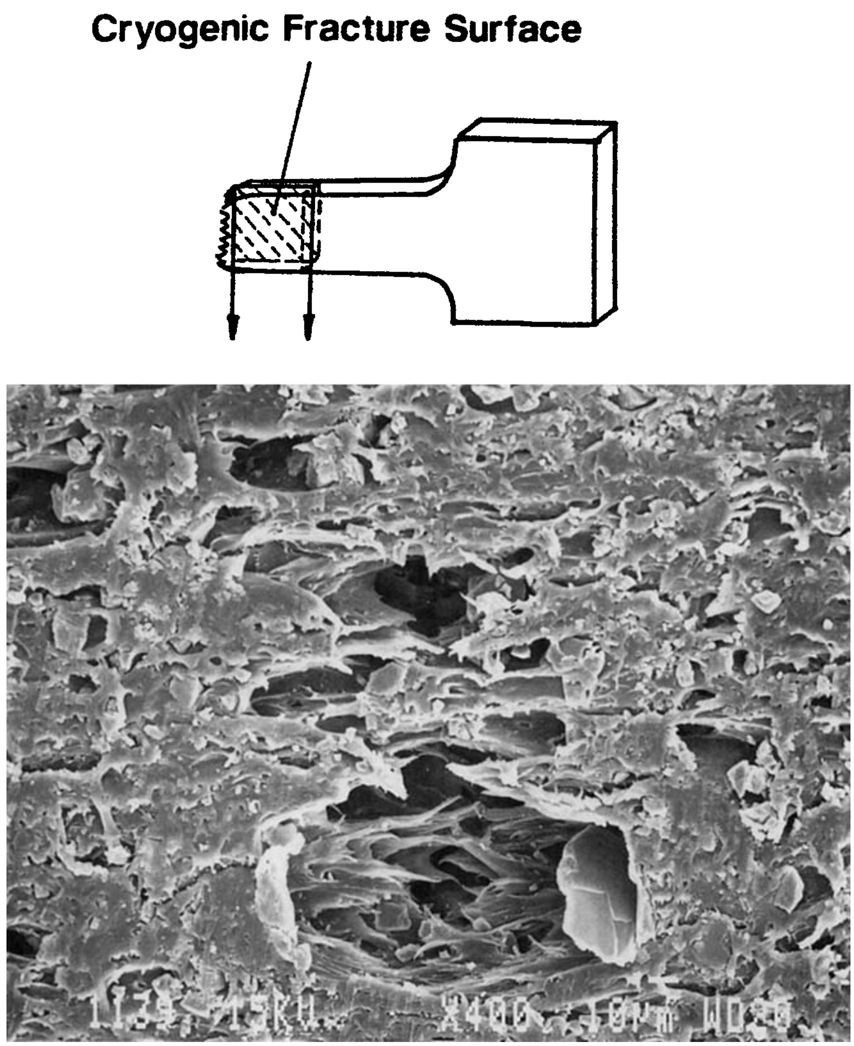


Figure 10 Necked region of PETG with 14 vol % $CaCO_3$-2 close to the fracture site. The specimen was cryogenically fractured parallel to the loading direction.

center line, and as a result, there was a well-defined directionality to crack growth.

Regardless of the macroscopic features, all Mode C fracture surfaces exhibited profuse cavitation with filler particles enmeshed in a network of small fibrils or fibril bundles. This is seen in Figure 9(c) and (d), which are higher magnifications of the fracture surfaces in Figure 9(a) and (b). Comparing the two examples, the surface of PETG with 14 vol % $CaCO_3$-2 contained pulled-out fibril bundles [Fig. 9(e)] that were longer and thicker than the small fibril bundles and single fibrils on the surface of PETG with 27 vol % CaT-1 [Fig. 9(f)]. This difference reflected the lower filler content of the specimen in Figure 9(a) (14 vol %) compared to the specimen in Figure 9(b) (27 vol %).

Cryogenic fracture of the deformed region adjacent to the fracture surface of PETG with 14 vol % $CaCO_3$-2 showed voiding around the filler particles (Fig. 10). In some regions, the thin ligaments between filler particles were highly drawn out. Broken fibrils and fibril bundles revealed that many of the pulled-out ligaments had fractured. Figure 10 shows how fibril fracture and lateral coalescence of the voids created a large flaw. When flaws such as the one in Figure 10 grew to critical size, they became the sites for primary and secondary crack initiation.

The ductile-to-quasi-brittle transition in filled PETG has been described by a microscopic failure condition that is able to predict macroscopic fracture behavior.[1] The condition for quasi-brittle fracture is attained at a critical filler volume fraction when the local stress on the ligaments between debonded filler particles reaches the tensile strength of the matrix and the ligaments are no longer able to support the engineering draw stress. Examination of the fracture surfaces, particularly, the Mode C fracture surfaces, provided experimental confirmation of this microscopic model for the ductile-to-quasi-brittle transition. The Mode C fracture mechanism is shown schematically in Figure 11. The filler particles were close enough to each other that when global or macroscopic necking started the high local stress on the thin ligaments between filler particles produced local necking with large extensions. When the thin ligaments were not able to support the load, they fractured and the voids coalesced laterally.

Fracture of drawn ligaments and coalescence into critical-sized voids was inherent to Mode C compositions and could occur at any location along the gauge section where necking started. For the same reason, Mode C fracture surfaces featured multiple crack initiation in the center of the cross section where triaxiality was highest. Whereas Mode B

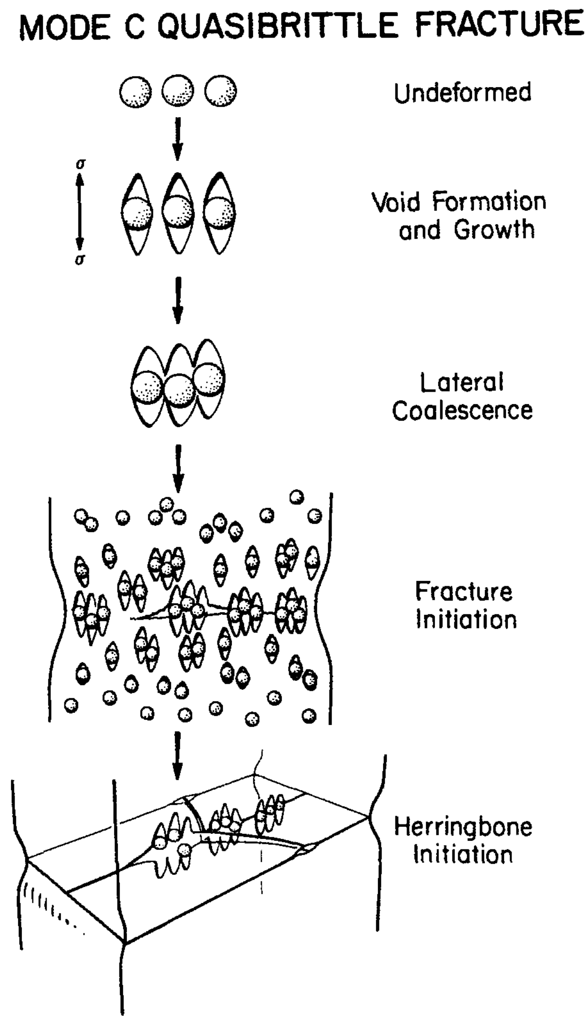


Figure 11 Schematic representation of Mode C fracture.

fracture also initiated from critical size voids that formed by ligament fracture, there was an important difference from Mode C fracture. Typically, the ligaments in Mode B compositions were able to support the load, and formation of the critical-size void was a singularity that led to fracture during propagation of an otherwise stable neck.

Mode D Quasi-brittle Fracture

Compositions that exhibited Mode D behavior fractured almost immediately after macroscopic yielding. Fracture initiated at the stress concentration at one end of the macro-shearband and initially propagated through the shearband at an angle to the loading direction. As the crack speed increased, the fracture path deviated from the shearband and proceeded through unyielded material in a direction that was nearly perpendicular to the loading direction. The fracture surface of PETG with 27 vol % CaT-2 in Figure 12(a) was typical of Mode D quasi-brittle fracture. Fracture initiated at the left edge and propagated through the macro-shearband to produce the stress-whitened region. About halfway across the fracture surface, a marked change in appearance indicated where the crack path deviated from the macro-shearband. The rough texture and absence of stress-whitening on the remainder of the fracture surface were indicative of brittle fracture.

Near the fracture initiation site, pulled-out fibrils around the filler particles created a dimple pattern (Fig. 12(b) and (e)]. The dimples became increasingly shallow as the crack speed increased away from the initiation site until close to the transition to brittle fracture they were only circular cavities that contained debonded filler particles (Fig. 12(c) and (f)]. In the brittle region, the polymer was not debonded from the particles, and the particles were flush with the fracture surface, suggesting that the

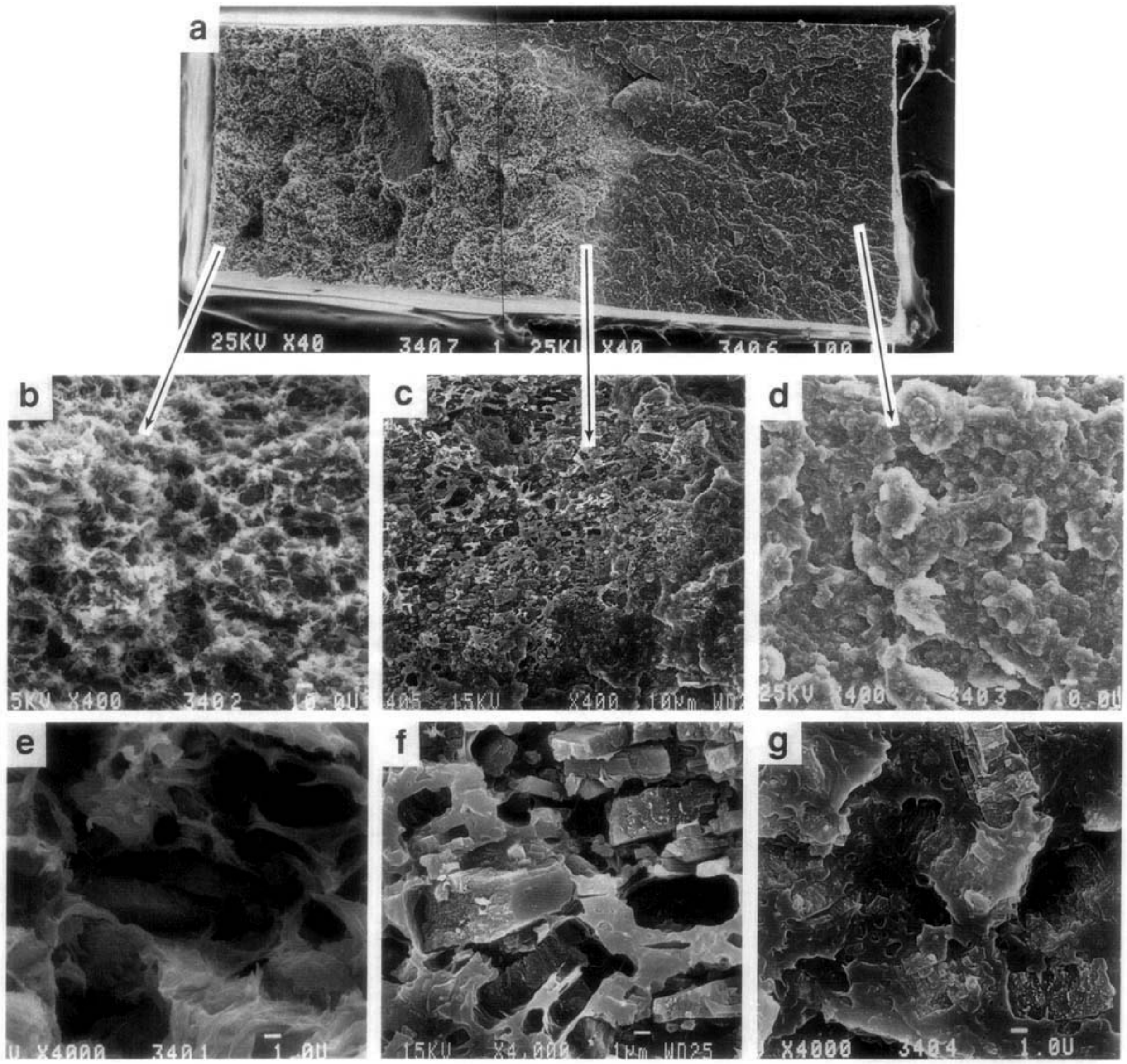


Figure 12 Mode D fracture surface of PETG with 27 vol % CaT-2: (a) the entire fracture surface; (b, e) higher magnifications of the region near the fracture initiation site; (c, f) higher magnifications of the region close to the transition to brittle fracture; (d, g) higher magnifications of the brittle fracture region.

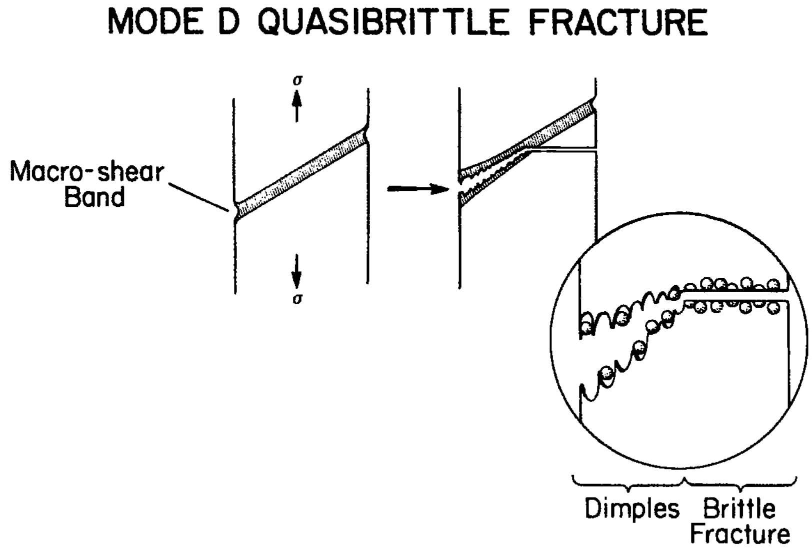


Figure 13 Schematic representation of Mode D fracture.

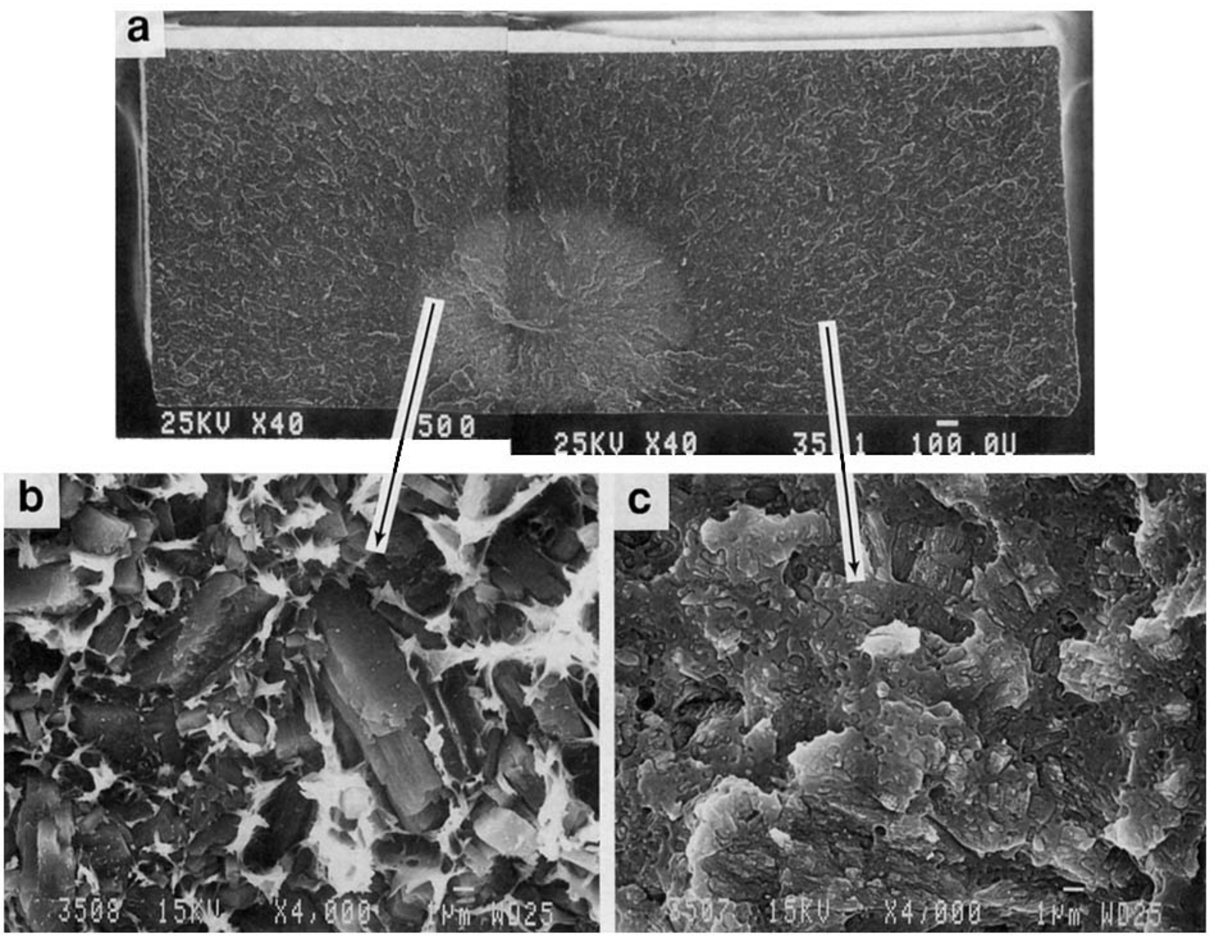


Figure 14 Mode E fracture surface of PETG with 45 vol % CaT-2: (a) the entire fracture surface; (b) higher magnification of the slightly stress-whitened region; (c) higher magnification of the brittle fracture region.

particles in the crack path had fractured [Fig. 12(d) and (g)].

Mode D fracture is shown schematically in Figure 13. Yielding occurred with formation of a macro-shearband. Within the macro-shearband, the thin ligaments between debonded filler particles were highly drawn. Tearing fracture of the ligaments as the crack propagated through the macro-shearband left a dimple pattern on the surface. The size and shape of the dimples were determined by the filler, while the depth of the dimples reflected the amount of plastic deformation in the ligaments before they fractured. The dimples became increasingly shallow as the crack accelerated until there was a change to brittle fracture.

Mode E Brittle Fracture

An example of a brittle fracture surface is shown in Figure 14. Fracture of this specimen of PETG filled with 45 vol % CaT-2 initiated at a flaw near the center. A region of slight stress-whitening surrounded the initiation site on the otherwise brittle fracture surface [Fig. 14(a)]. The stress-whitening was caused by numerous very short fibrils that remained on the fracture surface where the thin polymer ligaments had pulled out from the filler particles and fractured [Fig. 14(b)]. In the brittle region of the fracture surface [Fig. 14(c)], it was difficult to distinguish the filler particles from the polymer matrix. It appeared that the particles had not debonded, and in the crack plane, particles had fractured together with the undeformed matrix.

CONCLUSIONS

Five fracture modes were previously described in filled PETG on the basis of the macroscopic stress–strain behavior. The fracture mode changed from ductile (Modes A and B) to quasi-brittle (Modes C and D) to brittle (Mode E) with increasing filler content. Examination of the fracture surfaces for the purpose of characterizing the failure mechanisms associated with each of the modes led to the following conclusions:

1. Each fracture mode was identified with certain features of the fracture surface; these features were characteristic of the fracture mode and did not depend on filler type or filler content.
2. The Mode A fracture surface consisted of two regions: a pullout region of slower crack growth and a rosette region of faster crack growth; the Mode B fracture surface contained only a ductile pullout texture; the Mode C fracture surface exhibited secondary fracture features that sometimes included the herringbone pattern; the Mode D fracture surface consisted of a stress-whitened dimple region and a brittle fracture region; and the Mode E fracture surface exhibited primarily the rough texture characteristic of brittle fracture.
3. The failure mechanisms inferred from analysis of the fracture surfaces confirmed a microscopic failure model of the ductile-to-quasi-brittle transition in filled PETG; ligaments between filler particles were able to support the engineering draw stress during Modes A and B ductile fracture, whereas the transition to Mode C quasi-brittle fracture was accompanied by fracture of drawn ligaments and coalescence into critical-sized voids.

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