Notch Tip Damage Zone in Biaxially Oriented Polypropylene at Low Temperature

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SYNOPSIS

It is well established that biaxial orientation produces large improvements in the mechanical properties of polypropylene; this study further shows that the large improvements in mechanical behavior are magnified especially below the glass transition temperature. In this study, the irreversible deformation behavior of polypropylene during sharp single-edge notch tension testing has been studied at two levels of biaxial orientation at −40°C. Unoriented polypropylene formed a narrow wedge-shaped damage zone that grows with increasing stress until catastrophic fracture occurs in a brittle manor. The damage zone consisted of many crazes that mainly grew perpendicular to the loading direction. The 50% oriented material initially developed a wedge-shaped damage zone that grew wider as loading increased. This resulted in a drop of the length-to-width ratio at high sample extensions. The specimen fractured with stable crack growth in a ductile manner, showing a large resistance to crack growth. The 80% oriented material had a circular damage zone that consisted of many delamination crazes. These crazes grew by splitting the specimen in the thickness direction. Stable crack growth dominated the final failure process with the 80% oriented material showing nearly three times the toughness of the unoriented material.

INTRODUCTION

Recent studies of prefracture damage of polypropylene have revealed diverse mechanical and fracture behaviors depending on variables such as temperature, strain rate, and processing history.[1-6] At temperatures above the glass transition (T_g) of about 0°C and low strain rates, polypropylene exhibits ductile behavior and is insensitive to cracks, flaws, or notches. Under these conditions, polypropylene is capable of sustaining large amounts of prefracture damage at a notch tip prior to crack propagation.[2,3] However, below T_g or at high test speeds, the behavior of polypropylene shows a transition to brittle behavior, and, likewise, the prefracture damage also changes. Under these severe conditions, polypropylene is very sensitive to notches, and a prefracture craze damage zone grows from a sharp notch tip in a wedge shape. This wedge-shaped damage zone has been analyzed by the authors in detail.[1]

Several studies have shown that certain properties of polypropylene sheet can be substantially improved through processing techniques that impart biaxial orientation to the sheet.[7-10] Compressive biaxial orientation of polypropylene sheet by hydrostatic extrusion or by cross-rolling radically changes the properties of the starting material. In a recent study of hydrostatically extruded polypropylene sheet, it was shown that the oriented polypropylene had considerably more ductility and impact strength than that of the unoriented material at low temperatures ($< T_g$) and high strain rates.[7] This study also revealed major anisotropy between the in-plane properties and the properties in the thickness direction. This anisotropic behavior, which is also present in uniaxially oriented polypropylene, results in delamination-type failures during normal tension testing.[7,9] The goal of this article was to describe the behavior of biaxially ori-

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ented polypropylene at −40°C in the severe stress state produced by sharp-notched tension testing. This temperature was chosen because the effect of biaxial orientation on the mechanical properties is most striking below T_g.

EXPERIMENTAL

Polypropylene used in this investigation was commercially extruded sheet from Cadillac Plastics, Cleveland, OH. The sheets were prepared for cross-rolling by machining to the proper thickness and planar dimensions so that after cross-rolling all specimens would have the same final dimensions of 9 × 9 in. (23 × 23 cm) by 1.3 mm thick. Prerolled specimen dimensions were calculated assuming that the volume of the specimen remained constant. The measured specimen dimensions before and after rolling proved this to be a good assumption. Cross-rolling produced specimens with final levels of orientation of 30, 50, 70, and 80%, defined as the percentage reduction in thickness (t), and calculated using the equation $100 \times (t_i - t_f)/t_i$. Cross-rolling was conducted on a Fenn Manufacturing Co. 12 in.-diameter by 12 in.-wide rolling mill. The distance between the rolls was set to produce a net reduction in thickness of the sheet of 10% per pass. The sheets were rotated 90° between each pass, resulting in a balanced biaxial orientation. Each specimen was continually rolled in successive passes until the desired thickness reduction was achieved. After rolling, the sheets had a distinct bow that was more pronounced with increased orientation. The bowing was removed by flattening the specimens in a press at 110°C and a pressure of 100 psi for 15 min, then cooling to room temperature while under pressure. There was no change in the thickness of the sheet during the flattening process.

ASTM D638, type D, dog-bone tension specimens with dimensions of 60 mm gauge length, 13 mm width, and 1.3 mm final thickness were cut from the sheets. Specimens were polished using 800, 1200, and 2400 grit wet sandpaper to remove surface damage from the rolling process. For notched testing, a single 1 mm edge notch was slowly cut at room temperature using a fresh razor blade.

Tension testing was conducted on an Instron 1123 testing instrument equipped with a liquid nitrogen-cooled environmental chamber. Unnotched testing was carried out at a strain rate of 0.2% per min and the notched testing at a crosshead speed of 0.1 mm per min. Although test results for only one direction are presented in this text, there was no significant difference in stress–strain behavior in specimens taken in the 45° and 90° sheet directions. The prefracture damage ahead of the notch during loading was recorded using a 35 mm camera with a telephoto lens.

Several specimens were loaded to predetermined stress levels and unloaded for microtome sectioning. Microtoming was conducted at room temperature using glass knives. The specimens were sectioned to view the damage zone from the three orthogonal directions. Observations and micrographs were made using an Olympus optical microscope.

A JEOL 840A, low-voltage, scanning electron microscope was used to observe cross sections cut through several of the damage zones. Microtoming was used to prepare a smooth section through the damage zone; to help reveal the crazes, the microtomed specimens were immersed in benzene for several hours following a technique similar to that described previously.[6]

RESULTS AND DISCUSSION

Uniaxial Tensile Properties

Compressive biaxial orientation of polypropylene primarily affected yielding and elongation to failure in uniaxial tension testing at −40°C, with no change in the elastic modulus (Table I). Figure 1 shows the changes in tensile stress–strain behavior of polypropylene as a function of the level of orientation. The unoriented polypropylene control exhibited a relatively brittle response to loading with fracture occurring shortly after yielding. Many crazes appeared along the gauge length of the specimen as it yielded. The 30 and 50% oriented specimens showed progressive increases in ductility with increasing

Table I Mechanical Properties of Cross-rolled Polypropylene at −40°C

	Yield Stress (MPa)	Elastic Modulus (GPa)	Fracture Strain (%)
Unoriented	64.5	4.1	5
30%	65.5	3.9	12
50%	64.3	3.8	35
70%	63.9	4.1	156
80%	69.3	4.2	92

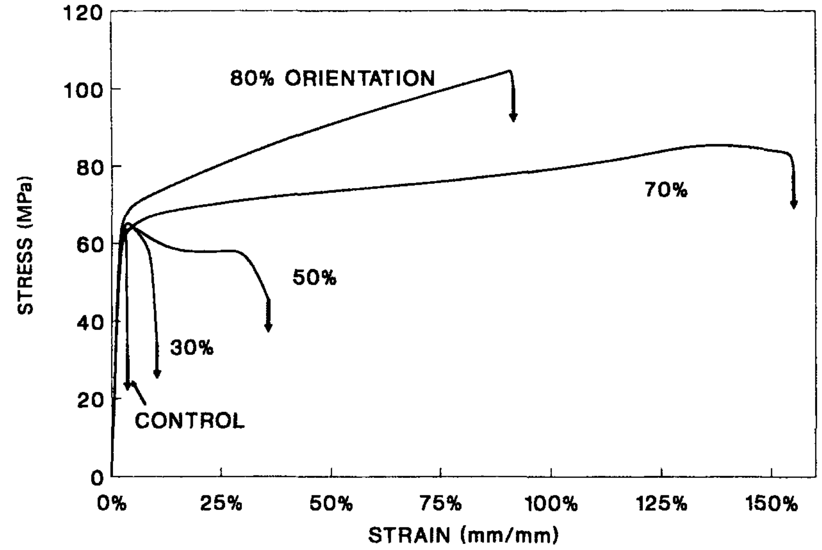

Figure 1 Engineering stress–strain curves of polypropylene at −40°C showing the effect of biaxial orientation.

biaxial orientation. Yielding in these cases occurred at similar stress levels with the formation of a stress-whitened neck that grew during subsequent drawing. Fracture occurred in the most intensely whitened region of the neck before the neck propagated the entire gauge length. In the 50% oriented specimen, there was substantially more drawing of the neck prior to failure than in the 30% specimen.

The 70% oriented specimen showed a substantial increase in ductility compared to the 50% oriented material; moreover, deformation was uniform along the entire gauge length. In contrast to lower orientations, no stress whitening was observed during yielding and drawing. Fracture also occurred in quite a different manner. Large amounts of delamination occurred when the specimen failed, with the specimen fragmenting into several pieces as it fractured. Delamination failures have been reported in both

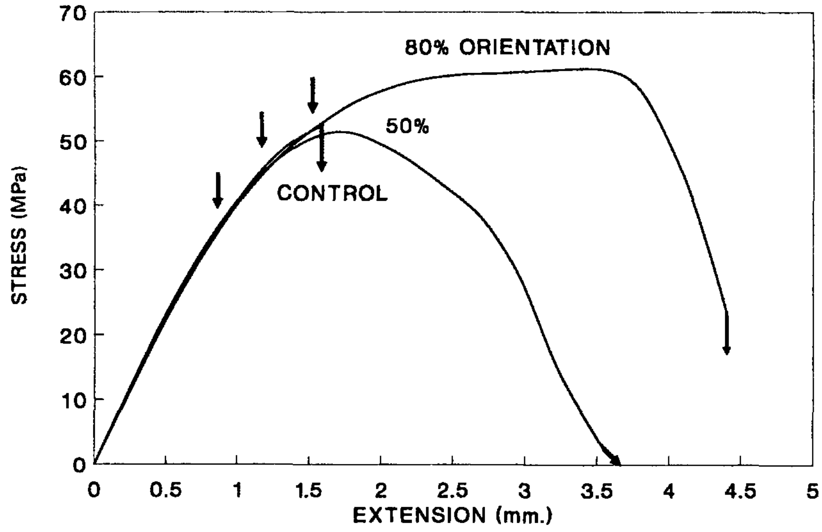

Figure 2 Notched stress-extension curves of polypropylene at −40°C showing the effect of biaxial orientation. The arrows indicate when photographs were taken.

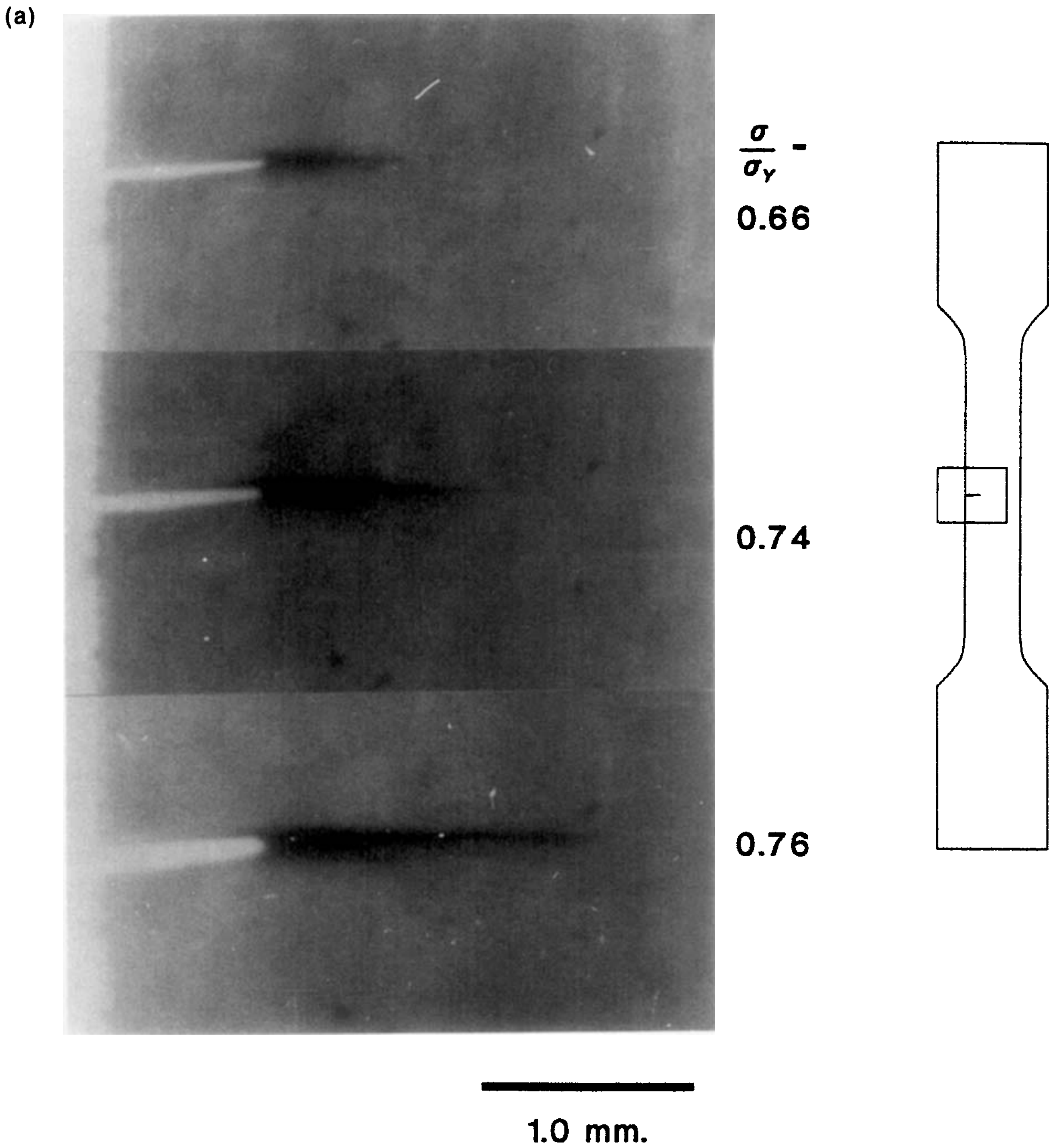

Figure 3 Photographs of the notch tip area during tension testing: (a) unoriented; (b) 50%; (c) 80% biaxial orientation.

biaxially and uniaxially oriented polypropylene produced by compressive processes.[7,9] This fracture mode reflected the imbalance in properties between the very tough rolled directions and the comparatively weak thickness direction. Material with the highest level of orientation, 80%, showed a slightly higher yield strength than that of the 30, 50, or 70% oriented materials. The macroscopic yielding and drawing characteristics were similar to 70% orientation, but these occurred at higher stresses and failure occurred at a lower strain. Fracture occurred in a similar delamination mode.

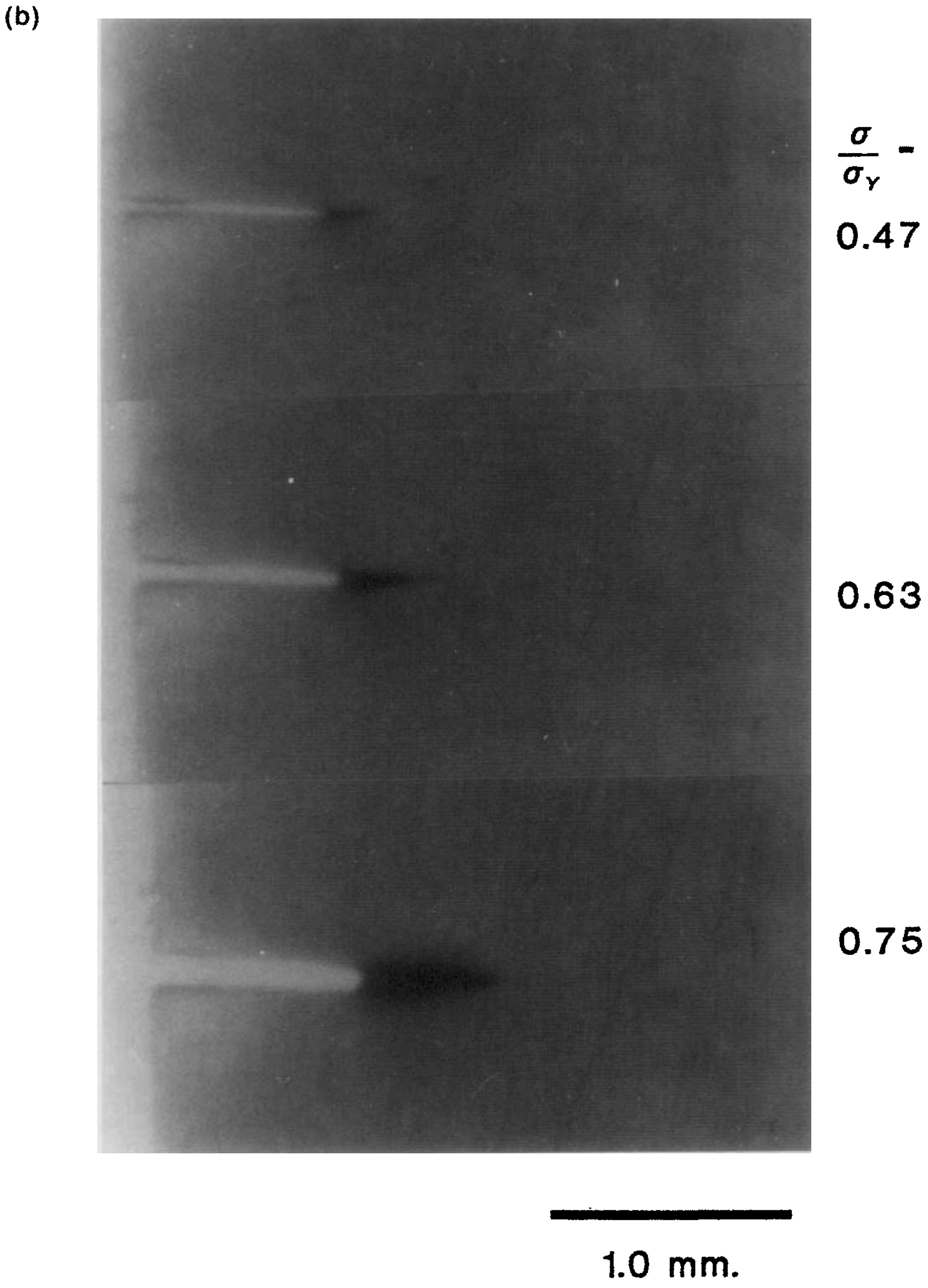

Figure 3 (*Continued from the previous page.*)

The Macroscopic Damage Zone at a Sharp Single-edge Notch

Figure 2 shows the results of notched tension testing on the control and 50% and 80% oriented materials at −40°C. The three stress-extension curves were very similar in the initial linear portion, with differences occurring after an extension of 1.5 mm. The control fractured in a brittle manner with very rapid crack growth. In contrast, the 50% oriented material continued to extend after 1.5 mm, with the stress continually dropping from a maximum at 51 MPa at 1.5 mm extension. The drop in stress accompanied slow growth of a tearing-type crack from the initial

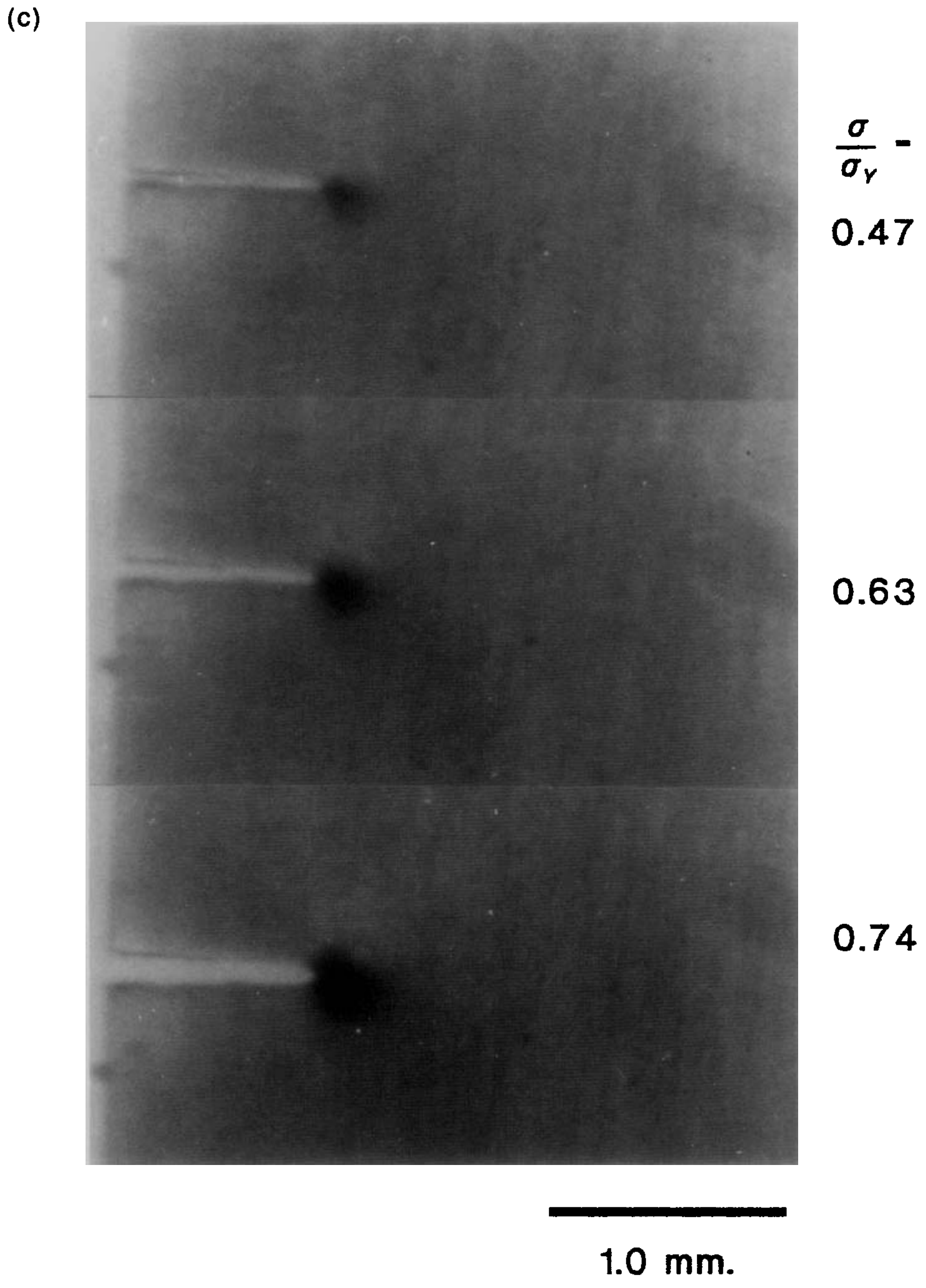

Figure 3 (*Continued from the previous page.*)

notch across the specimen. In the 80% material, the stress continued to increase from a value of 52 MPa at 1.5 mm extension, leveling off at 61 MPa at high extension. As in the 50% case, a slow stable crack grew from the notch tip starting at 1.5 mm extension. However, in contrast to the 50% case, crack growth in the 80% material occurred with an increase in the stress between 1.5 mm extension, when crack growth began, and an extension of 3.6 mm. These results showed that there was a great improvement in resistance to crack propagation as the level of orientation increased. The toughness, as indicated by the area under the curve, revealed qualitatively that the 50% oriented material was roughly twice as tough as the control, and the 80% material, approximately three times as tough as the control.

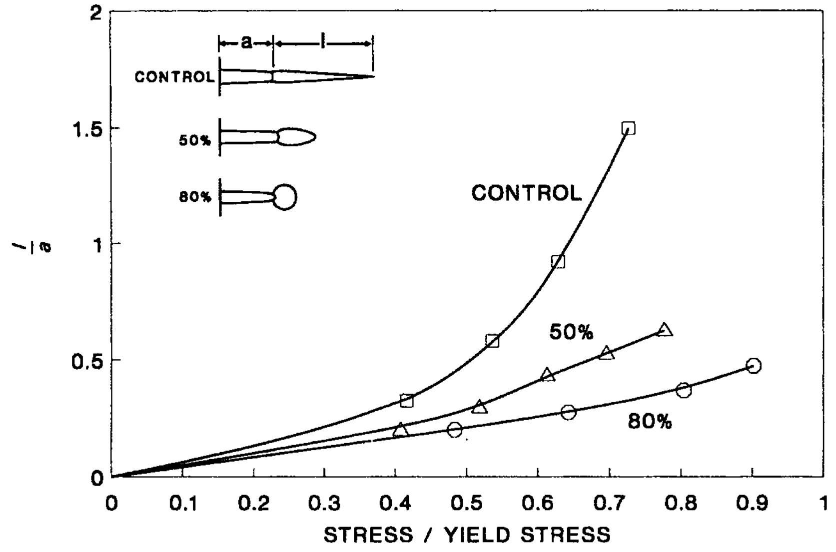

Figure 4 Plot of the normalized damage zone length (l/a) vs. normalized stress.

These results paralleled the unnotched testing showing the large improvements in properties due to biaxial orientation.

Prior to the crack growth stage, prefracture damage was observed at the notch root in all three cases between 35 and 52 MPa. The arrows in Figure 2 indicate points at which corresponding photographs in Figure 3(a)–(c) were taken of the crack tip damage zone. The wedge-shaped damage zone of the unoriented polypropylene [Fig. 3(a)] has been described in detail in a previous publication, where damage zone growth fit the Dugdale model.[1] In the 50% material [Fig. 3(b)], the macroscopic damage zone also had a wedge shape but was shorter in length than the damage zone of the unoriented material. In contrast, the highly oriented 80% material [Fig. 3(c)] developed a damage zone that was circular in shape.

The length of the damage zone for each of the three levels of orientation is plotted in Figure 4 as a function of stress normalized with respect to the yield stress. The plotted points, which are the average of photograph measurements from at least two specimens, show that at a given normalized stress

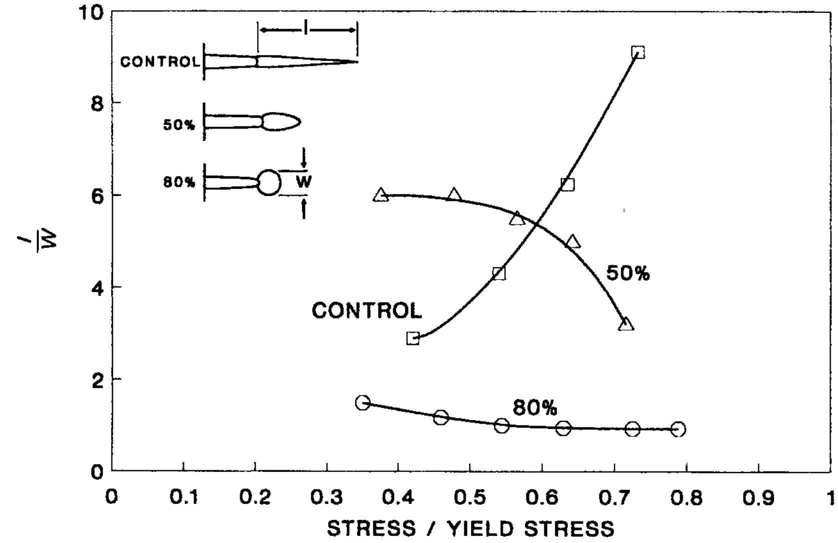

Figure 5 Plot of the damage zone length-to-width ratio as a function of normalized stress, comparing unoriented polypropylene with the 50 and 80% material.

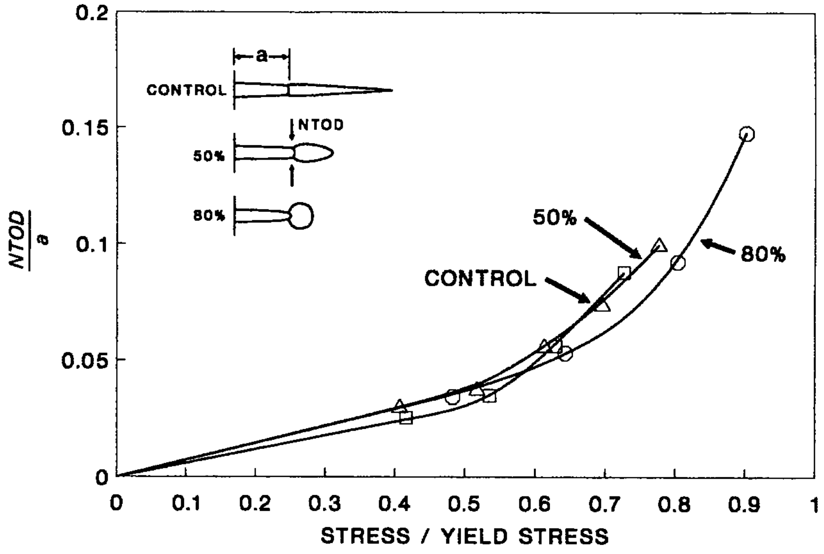

Figure 6 Plot of the normalized notch tip opening displacement (NTOD/a) vs. normalized stress showing the similarity between unoriented and oriented polypropylene.

the length of the damage zone decreased as the level of orientation increased. This reflected changes in damage mechanisms within the zone as a result of biaxial orientation. Previously, the length of the narrow wedge-shaped zone in unoriented polypropylene was adequately described by the one-dimensional Dugdale model that relates the damage zone length to a function of the applied stress normalized by the unnotched yield stress.[1] In the case of the oriented materials, it was obvious that a different approach was needed.

Although the damage zone of the control was longer at any relative stress level than that of the oriented materials, at high stresses, it was much narrower and resembled a crack. Figure 5 shows the damage zone length-to-width ratio (l/w) as a function of normalized stress. The unoriented material showed an increase in this ratio with increased applied stress until the crazed damage zone actually became a crack and catastrophic fracture occurred. The damage zone of the 50% oriented specimens was initially sharper than that of the control with

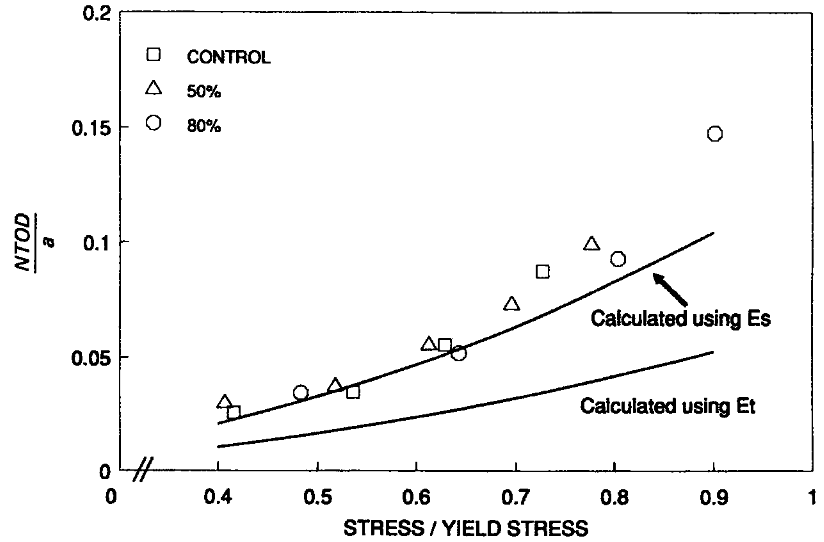

Figure 7 Comparison of the measured notch tip opening displacement with the prediction using the tangent modulus (E_t) and using a secant modulus (E_s).

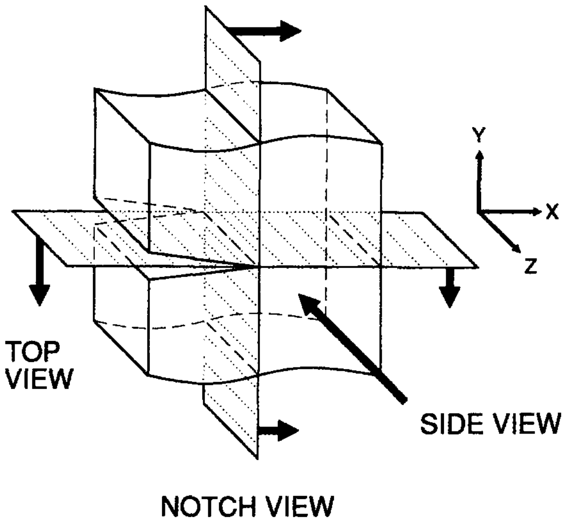

Figure 8 Schematic drawing of the sectioning used to view the damage zones in three different planes.

a length-to-width ratio that remained fairly constant at 6 : 1 up to a normalized stress of 0.55 σ_y. Above this stress level, the damage zone width grew faster than the length, and the ratio dropped approaching 3 : 1 when crack propagation began at a normalized stress of 0.75 σ_y.

In marked contrast, the 80% oriented material showed a damage zone with a nearly constant length-to-width ratio of about 1 at all stress levels, reflecting its nearly constant circular shape. This ratio persisted to a stress level of 0.80 σ_y when crack propagation through the damage zone began. Since the fully developed damage zone had relieved the triaxial stress state, crack propagation occurred in a plane stress mode with no further development of the macroscopic damage zone.

The notch tip opening displacement measured for each of the tested materials is reported in Figure 6. The figure shows that these data fell together for

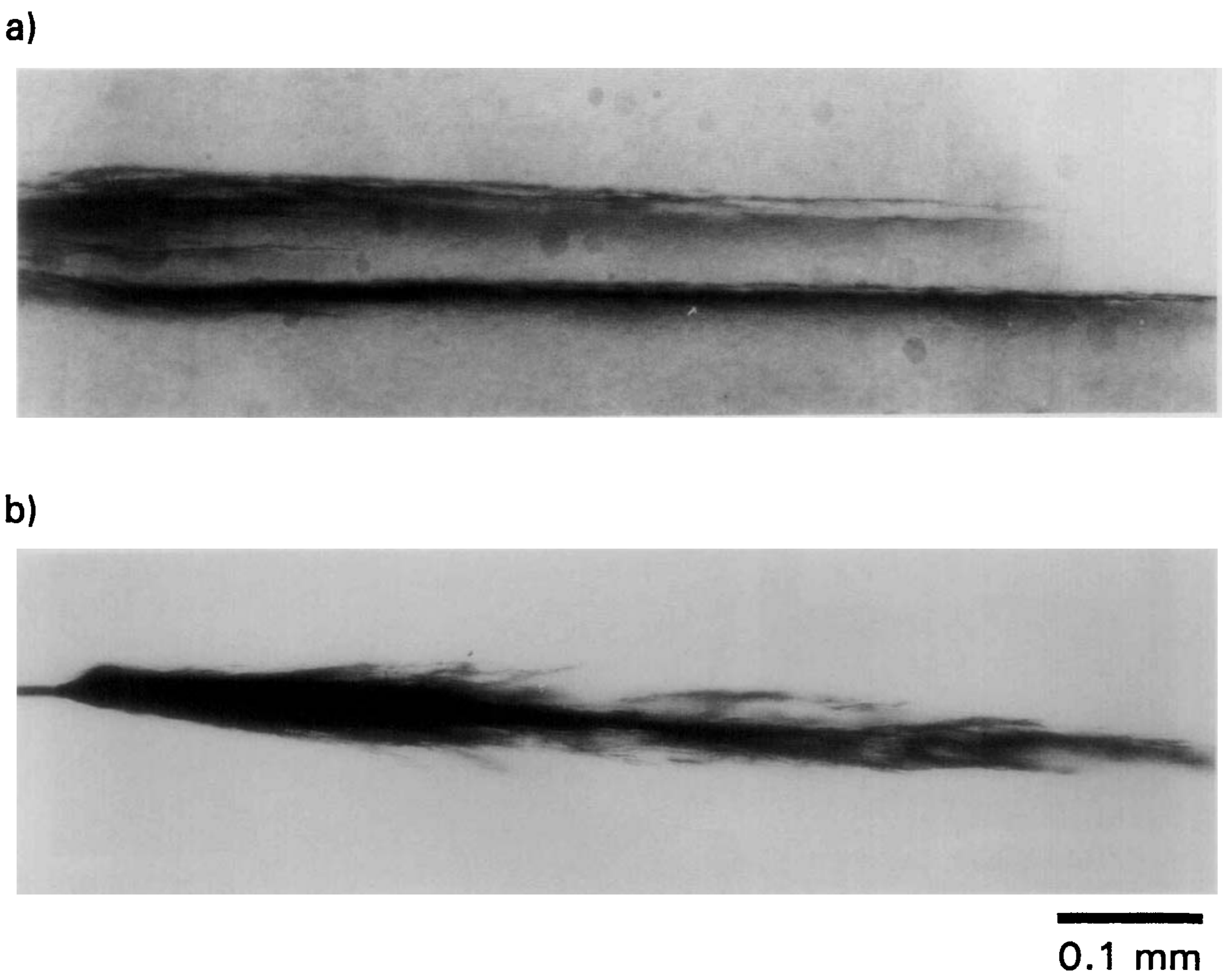

Figure 9 Optical micrograph of a cross section of (a) unoriented polypropylene and (b) 50% cross-rolled polypropylene (side views).

all levels of orientation within the range of experimental error. This result suggested that the strain at the notch tip was not significantly influenced by the damage zone immediately ahead of the notch tip, but was controlled by the large volume of material outside of the damage zone. At a given stress level, the elastically strained material outside the damage zone was displaced the same amount regardless of level of orientation since the modulus of elasticity had not changed at these levels of orientation.

Prediction of the notch tip opening displacement (NTOD) for the case of an elastic stress field ahead of a sharp single-edge notch has been published by several authors.[11,12] The NTOD can be described by

$$\mathrm{NTOD} = \frac{K_I^2}{\sigma_y E} = \frac{\sigma_0^2 a Y^2}{\sigma_y E} \qquad (1)$$

which can be rewritten as

$$\frac{\mathrm{NTOD}}{a} = \left(\frac{\sigma_0}{\sigma_y}\right)^2 \left(\frac{\sigma_y}{E}\right) Y^2 \qquad (2)$$

where $K_I = \sigma_0 Y \sqrt{a}$, $Y = 1.99 - 0.41(a/W) + 18.7(a/W)^2 - 38.48(a/W)^3 + 53.85(a/W)^4$, σ_0 is the externally applied stress; a, the notch length; σ_y, the yield stress; and W, the specimen width. This prediction is plotted in Figure 7 using both the tangent modulus (E_t = 4.11 GPa) and the secant modulus (E_s = 2.07 GPa), determined in previous work with unoriented polypropylene.[1] This figure showed that the notch opening displacement data fit one curve, independent of level of orientation, which was described by the secant modulus.

Microscopic Damage Zone Morphology

Notched specimens were loaded to several stress levels and then unloaded for sectioning as shown in Figure 8. In Figure 9, optical micrographs of the side view of the damage zone of unoriented and 50% oriented materials show the gradual change in damage zone morphology that occurred with biaxial orientation. The damage zone of the unoriented polypropylene consisted of crazes that grew on through-

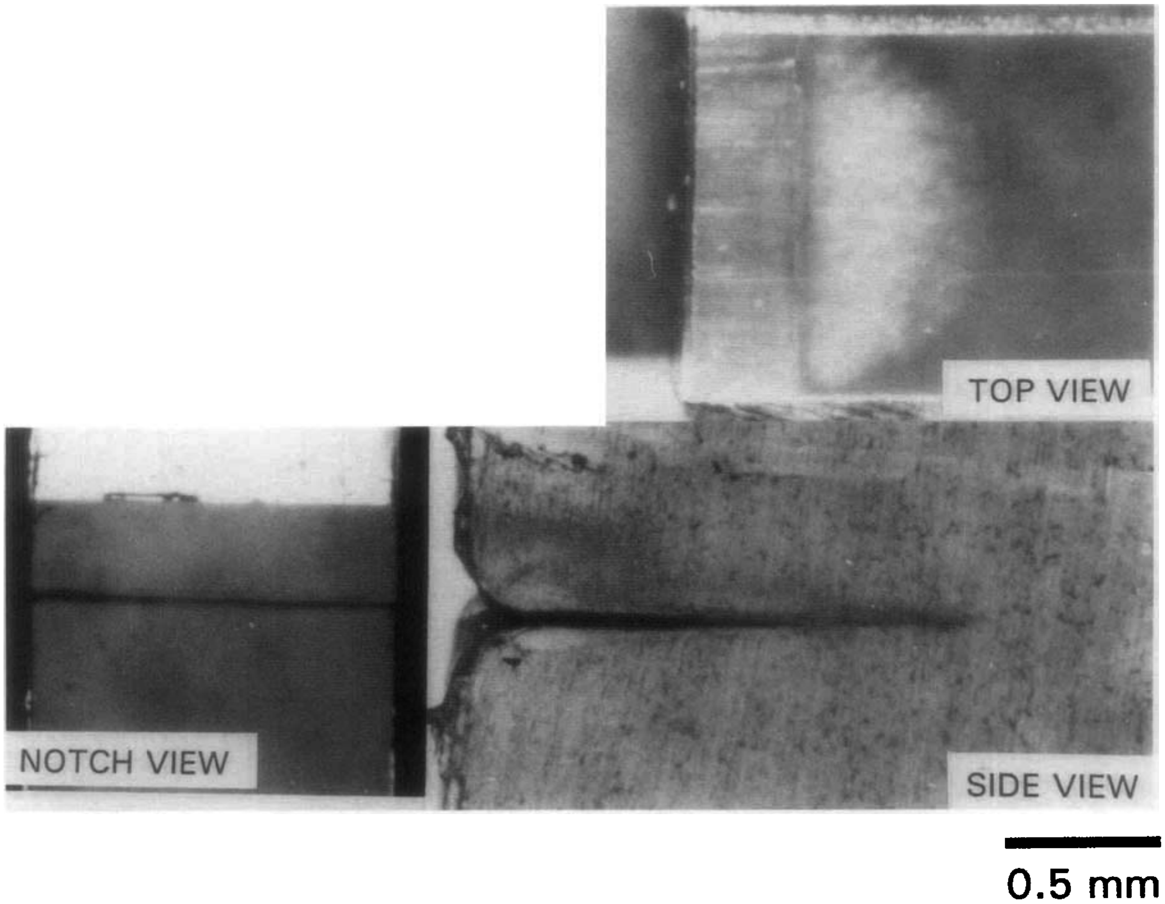

Figure 10 Three orthogonal views of the damage zone in an 50% oriented specimen loaded to 0.7 σ_y at −40°C.

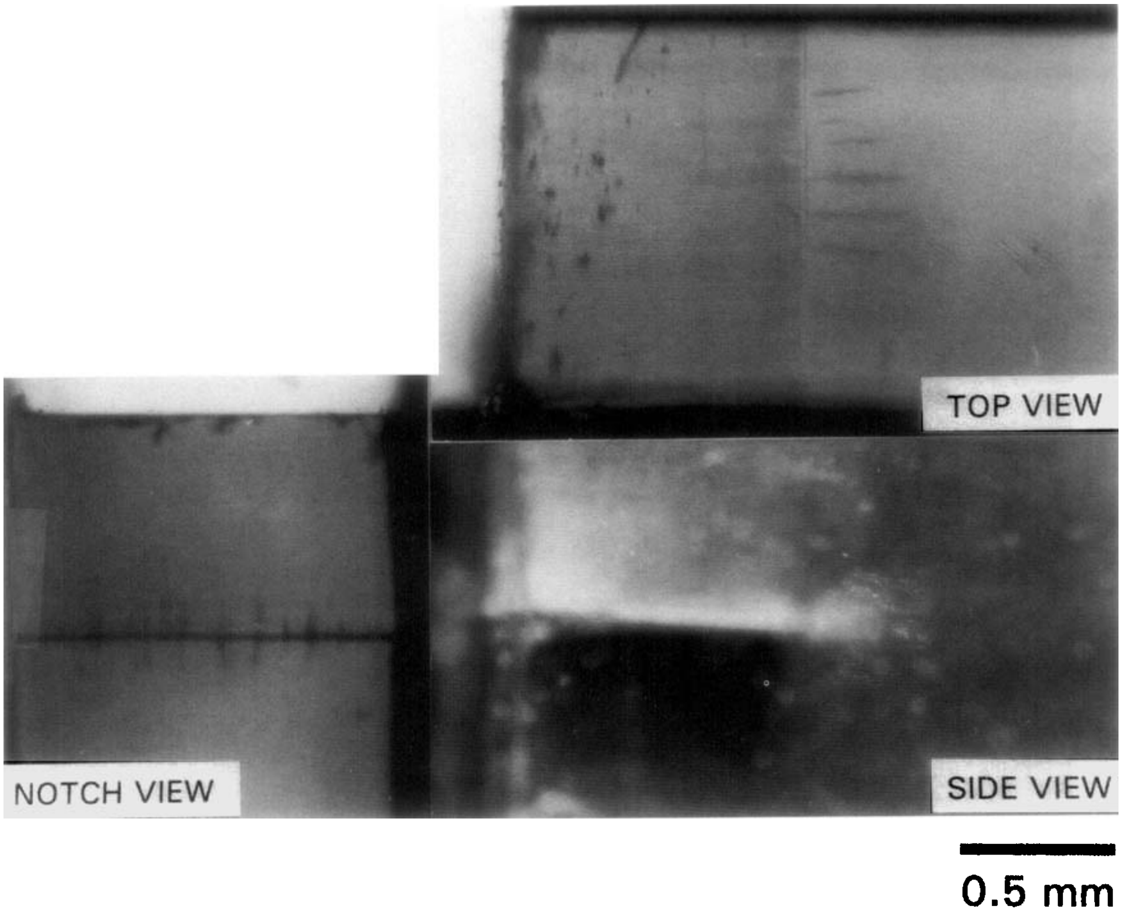

Figure 11 Three orthogonal views of the damage zone in an 80% oriented specimen loaded to 0.7 σ_y at −40°C.

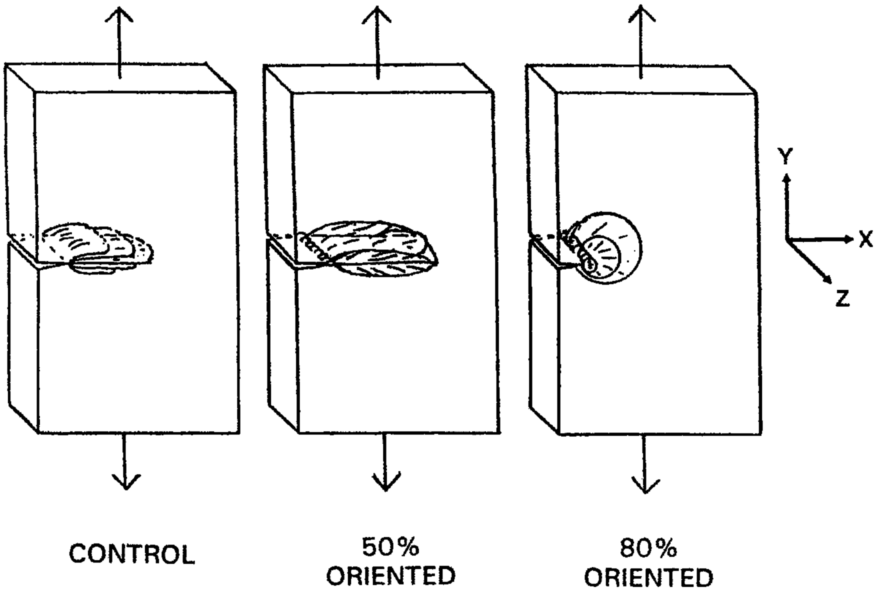

Figure 12 Schematic drawings comparing the damage zone morphology as a function of biaxial orientation.

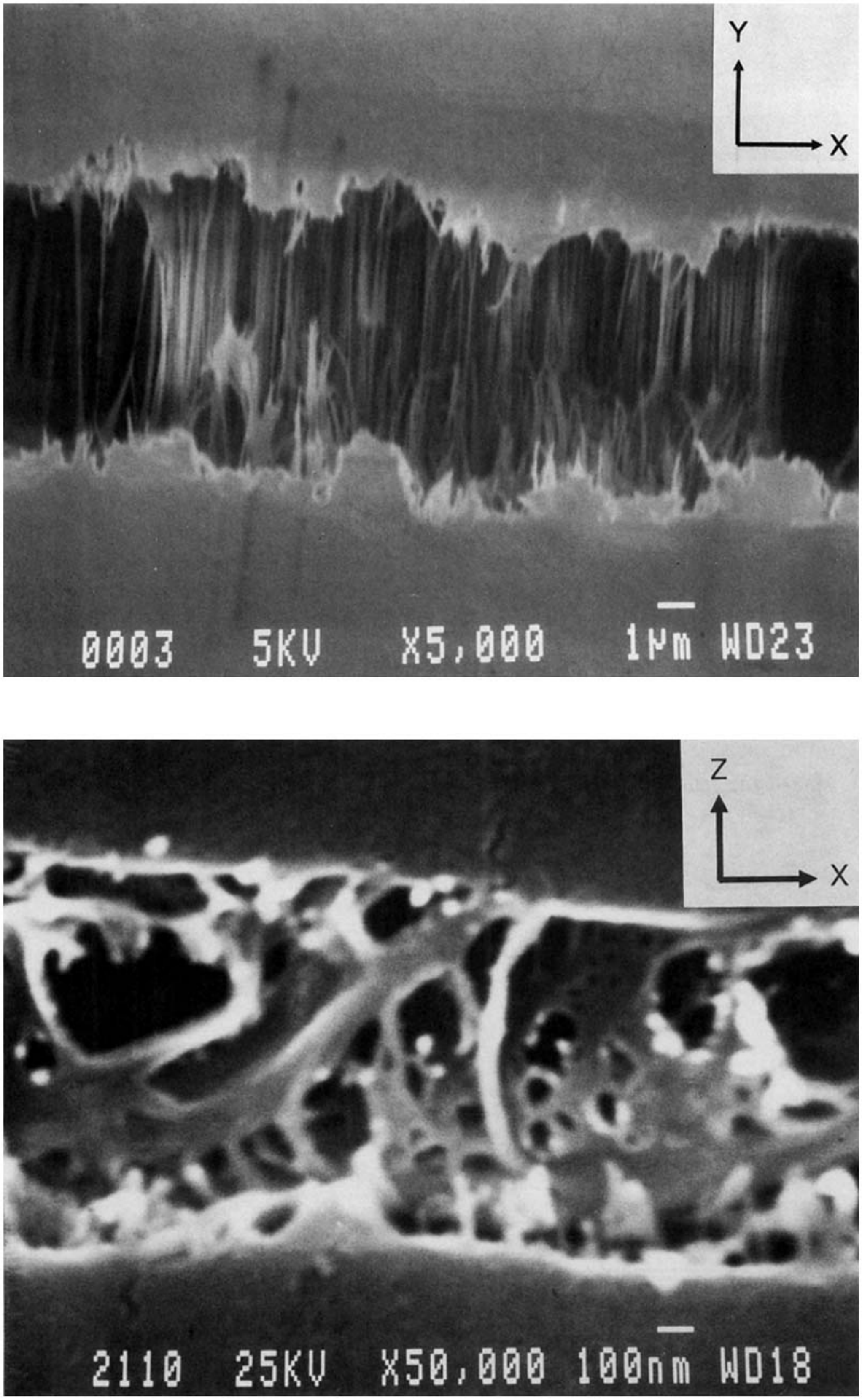

Figure 13 SEM micrographs showing high magnification of crazed material in (a) unoriented polypropylene and (b) 80% oriented polypropylene damage zone.

thickness planes primarily in the x-direction following the principal stress trajectories.[1] In contrast, the damage zone of the 50% oriented material had a feathery appearance, indicating that the crazes meandered in both the y- and z-directions during x-direction growth. The growth in the y-direction was reflected in the changing l/w ratio, which was initially high and subsequently dropped at higher stress levels.

The three orthogonal views of the damage zone of the 50% oriented specimen in Figure 10 show a stress-whitened morphology that extended through the thickness (top view), but was confined to a very narrow band barely visible in the side view. This planar damage zone growing primarily in the x-direction was similar to the unoriented material. Both these cases were, however, very different from the 80% oriented material.

Figure 11 shows micrographs of the three orthogonal views of the damage zone of 80% biaxially oriented material after being stressed to 0.7 σ_y. The top and notch views revealed that the circular macroscopic damage zone observed in the side view consisted of many penny-shaped delamination crazes growing perpendicular to the thickness direction. This unique form of crazing demonstrated the weakness of the thickness direction compared to the much stronger rolled directions. Figure 12 shows schematically the three-dimensional character of the three types of damage zones, illustrating the change from planar crazing in the unoriented material to penny-shaped delamination crazing in the highly oriented material.

The craze morphologies were compared by observing sections through the damage zones of unoriented and highly oriented polypropylene in the scanning electron microscope. The craze of the unoriented material was observed in the x–y plane [Fig. 13(a)]. It had the typical craze morphology with numerous fibrils spanning the craze in the direction of applied stress. A similar craze morphology was reported by Jang et al.[6] for polypropylene fractured below the T_g. In contrast, the crazed material of the 80% oriented material, which was viewed in the x–z plane, was not fibrillar [Fig. 13(b)]. The material bridging the craze was sheetlike with many voids and resembled the appearance of wood split along the grain. Propagation of delamination crazes parallel to the stress direction was a direct result of the high anisotropy of the strength between the plane of the rolled directions and the thickness direction.

CONCLUSIONS

The results of this investigation showed that the compressive biaxial orientation of polypropylene produced a major change in the mechanisms of irreversible deformation and damage. As a result, this produced a material that was very tough at −40°C, specifically:

1. Biaxial orientation produced large improvements in the mechanical properties of polypropylene at −40°C. The most significant change was in the ductility, which increased from a strain to fracture of 5% in the unoriented case to over 150% in the highly oriented case.
2. The shape of the damage ahead of a sharp-edge notch changed with the amount of biaxial orientation from a narrow wedge in the unoriented case to a circular zone in the highly orientated case.
3. On the microscopic scale, the circular damage zone of the highly oriented material consisted of many delamination crazes that grew in a splitting mode parallel to the weak thickness direction.

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