

Investigation of Deformation Mechanism in Polypropylene/Glass Fiber Composite

S. M. Zebarjad,¹ R. Bagheri,² S. M. Seyed Reihani,² M. Forunchi²

¹Department of Materials Science and Engineering, Engineering Faculty, Ferdowsi University, Mashhad, Iran

²Department of Chemical Engineering, Sharif University of Technology, Tehran, Iran

Received 12 November 2001; accepted 12 March 2002

ABSTRACT: A great amount of work has been done over the past few years to understand the structure–properties relationship in polypropylene/glass fiber composites. This is because of the very fast-growing rate of polypropylene applications in the automotive and other industries. This work focused on the role of glass fibers and fiber–matrix adhesion on deformation mechanism. Composites with different fiber content, with and without adhesion promoter, were fabricated. Tensile tests and microscopy techniques were conducted. Based on the results, a physical model has been proposed that illustrates the initiation and growth of

the damage under static loading condition. According to this work, the introduction of glass fibers shifts the deformation mechanism from shear yielding to crazing. Although crazes propagate in a different manner in composites containing standard and treated fibers, no influence of interfacial adhesion was observed on craze thickening phenomenon. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 87: 2171–2176, 2003

Key words: polypropylene composite; deformation; mechanical properties

INTRODUCTION

Polypropylene (PP) is one of the fastest-growing classes of commodity thermoplastics, with a market share growth of 6%–10% per year.^{1,2} The volume of polypropylene produced is exceeded only by polyethylene and poly(vinyl chloride).² The moderate cost and favorable properties of polypropylene contribute to its strong growth rate. It is one of the lightest of all thermoplastics (density of 0.9 g/cm³), so that fewer grams are required for finished parts.³

To reduce cost or enhance physical and mechanical properties, fillers and reinforcements are added to polypropylene. Fillers and reinforcements used include talc, calcium carbonate, mica, wollastonite, glass fiber, carbon fiber, and jute.^{2,4–6}

Glass fiber (GF) is one of the most widely used reinforcements in thermoplastics because it is cost effective and yields a broad range of physical and mechanical properties suitable for a large number of applications.^{7,8} In application terms among the glass-filled thermoplastics, polypropylene has the second highest tonnage use following nylon 66. The rate of consumption of glass-reinforced PP, however, has been consistently higher in recent years, and it may eventually become the most important reinforced

thermoplastic.⁴ Glass-fiber-reinforced PP is used in the automotive, appliance, furniture, and electrical industries. Presently, a wide range of reinforced PP with glass fiber exists.²

There are many studies that have investigated the mechanical behavior of PP–GF composites.^{8–20} Two main approaches to investigating this behavior have been found in these works. The first approach is concerned with adhesion at the fiber–matrix interface, and thus the incorporation of the coupling agent. Promoting interfacial shear strength (IFSS) has been the main purpose of this group of investigators.^{8–14} For example, Folkes and Hardwick¹³ and Rijdsdijk et al.¹⁴ reported that the improvement of interfacial shear strength contributes to the improvement of the flexural properties of the PP–GF composite. The second approach pays attention to the morphology of the spherulitic structure and its interaction with glass fibers.^{15–20} Those using this approach have observed that the nucleation density of spherulites along the fibers is different from the density in other places.^{18–20}

Despite the importance of deformation mechanism on the mechanical behavior of materials, little attention has been paid to this subject in PP–GF composites. There are only a few works on deformation of pure polypropylene.^{1,21–23} These researchers have mostly focused on the macroscopically inhomogeneous deformation, that is, necking, of polypropylene under uniaxial tensile loading.^{21–23}

The goal of the current study, however, is to present a physical model for elucidating the role of short glass

Correspondence to: S. M. Zebarjad (zebarjad@mehr.sharif.ac.ir).

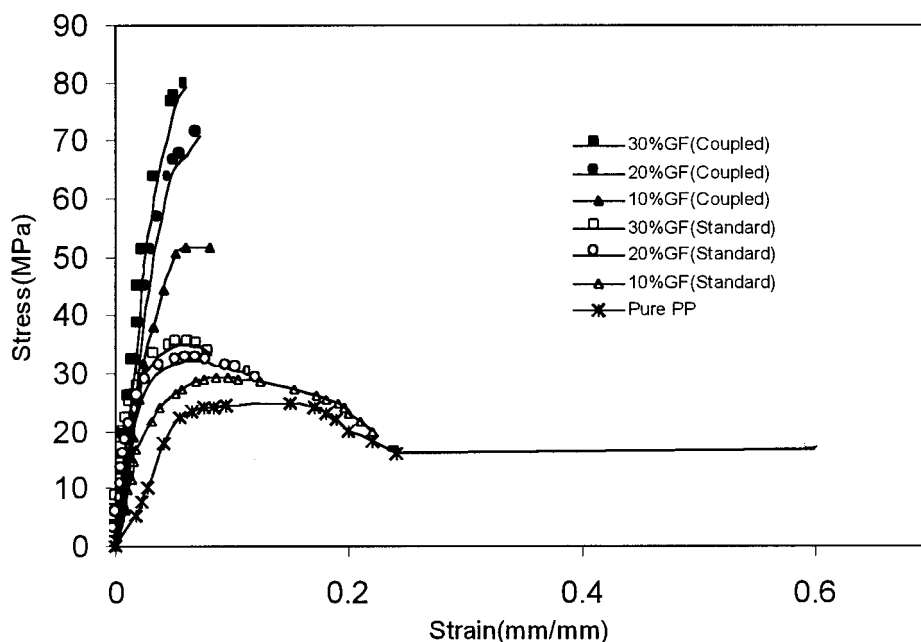


Figure 1 Dependence of the tensile behavior on glass fiber type and content.

fibers and their adhesion to the matrix on the deformation behavior of polypropylene. It is noteworthy that deformation mechanism contributes to the energy absorbed prior to fracture. Therefore, understanding this phenomenon is a great task in analyzing the mechanical performance of this composite system.

EXPERIMENTAL

Materials

To look at the role of interface, standard and silane-treated E-short-glass fibers were incorporated into this study. Composite pellets containing 30 wt % of both types of glass fibers with an average length and diameter of 6 mm and 13 μm , respectively, were obtained. The reinforcement contents of composite materials were then reduced to 20, 10, and 5 wt % by diluting them with neat polypropylene supplied by Imam Khomeini Petrochemical Complex (Bandar Imam, Iran). According to the manufacturer, that is an isotactic polypropylene (iPP) with a melt flow index (MFI) of 8. Standard tensile specimens were made through injection molding. The temperature in the four successive zones of the injection-molding apparatus was increased along its length from 200°C to 230°C.

Tensile test

The tensile test was done according to ASTM D 638 using an 1115 Instron tensile frame at a cross-head speed of 5 mm/min. Three samples per material were

tested at room temperature. To investigate the dominant deformation mechanism in the tensile test, the side surfaces of some samples were polished prior to the test to achieve a surface roughness of about 5 μm . After deformation occurred on some surfaces, those surfaces were studied via optical and scanning electron microscopes.

Microscopic evaluation

An Olympus PME3 optical microscope was employed to examine the damage in tensile specimens under a reflective light condition. A Jeol XJA-840 scanning electron microscope (SEM) was used to study the side surface of tensile bars and the fracture surface of selected samples. The specimens were coated with a thin layer of gold prior to microscopy to avoid charge buildup.

RESULTS AND DISCUSSION

Figure 1 shows the influence of glass fiber (with and without a coupling agent) and content on the stress-strain curve. As seen, increasing GF content reduced the tendency of the material to draw. As a matter of fact, the necking phenomenon was observed in pure PP while the neck extended throughout the gauge length. Figure 2 shows the steps of initiation and propagation of necking through the gauge length in pure PP. As seen in this figure, plastic deformation starts somewhere in the gauge length of the specimen, and further stretching causes the neck to grow along

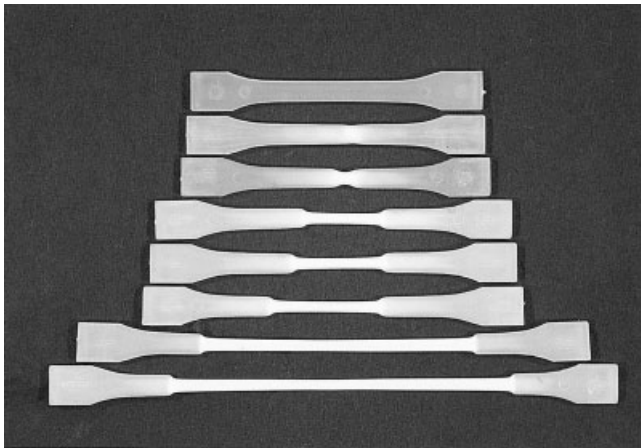


Figure 2 Initiation and propagation of the necking phenomenon in pure PP is seen from top to bottom.

the specimen. The results of this study illustrate that the addition of glass fibers to PP suppresses its tendency to neck. Also the results of the tensile test are summarized in Table I. As expected, yield stress and tensile modulus increased with increasing glass fiber content. This is because glass fibers have high tensile strength and modulus compared with neat polypropylene. Moreover, it can be seen that yield stress, modulus of elasticity, and elongation to failure were highly dependent on the surface treatment of glass fibers, that is, the fiber–matrix interface. It has been claimed that the primary function of the fiber–matrix interface is to transmit stress from the weak polymer matrix to the high-strength fibers.^{24,25} The absence of adhesion between the fibers and the matrix causes debonding and pullout of the fibers at low stress levels. Nevertheless, a high interfacial strength between glass fibers and polypropylene causes ease-of-stress transmission from the weak matrix to strong fibers before the fracture occurs²⁴.

The pictures in Figure 3 were taken from the tensile fracture surface of composites containing 20 wt% of standard [Fig. 3(a)] and silane-treated glass fibers [Fig. 3(b)]. The pulled-out fibers with a smooth and clean

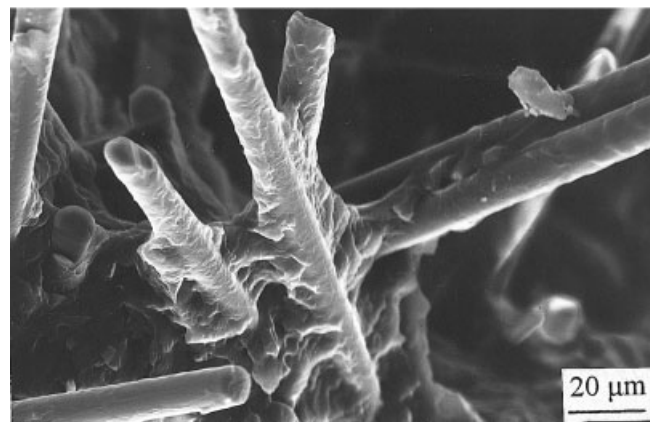


Figure 3 Fracture surface of PP reinforced with: (a) standard glass fibers, (b) treated glass fibers.

surface seen in Figure 3(a) indicate the weak interface in this system. The glass fibers seen in Figure 3(b), on the other hand, have a rough surface, indicating the strong adhesion between the fibers and the matrix. The ease of interfacial debonding for the standard glass fibers used enhanced void formation in this system [Fig. 3(a)]. Therefore, the lower modulus observed for the untreated fibers (Table I) may be attributed to the higher void content resulting from the lower wettability during impregnation of PP into the glass fibers.²⁴ Void content measurement in PP–GF composites done by Lee et al.²⁴ showed a counter correlation between the void content and the interfacial strength.

The pictures in Figure 4(a,b) show the results of the optical microscopy done on the polished surfaces of pure PP and PP–GF (20 wt %, treated GF), respectively. As seen in Figure 4(a), shear bands are evident as bright cross patterns in pure PP. The formation of shear bands at an angle of about 45° to the direction of the remote loading provides evidence that the dominant deformation mechanism in pure PP is shear yielding. This result coincides with the findings of previous investigators.^{1,21–23} The introduction of glass fibers, as seen in Figure 4(b), caused a totally different

TABLE I
Comparison of Tensile Properties of Polypropylene and Its Composites

Material	Yield stress (MPa)	Young's modulus (GPa)	Elongation (%)
Pure PP	24.9	1.3	No Break
PP/10 wt % GF (untreated)	29	2.68	23
PP/10 wt % GF (treated)	51.62	3.12	8
PP/20 wt % GF(untreated)	32	3.38	13
PP/20 wt % GF (treated)	70.9	4.68	7.8
PP/30 wt % GF (untreated)	35	3.9	9
PP/30 wt % GF (treated)	79.07	5.538	6

deformation behavior. Crazes seen in Figure 4(b) form perpendicular to the loading direction. Variation of the deformation mechanism from shear yielding to crazing was observed in this study for composites reinforced with both standard and surface-treated glass fibers. Therefore, it can be concluded that the interfacial bonding at the fiber–matrix interface has no influence on the variation in the deformation mechanism caused by the introduction of glass fibers.

For further elucidation of the crazing phenomenon in composite materials, electron microscopy was employed. Figure 5(a–c) shows the result of the scanning electron microscopy done on the polished surfaces of PP–GF composites. The results of this investigation and our previous work²⁶ illustrate that crazes in PP–GF composite form mostly at the fiber–matrix interface. It was also found that this is independent of the adhesion at the interface. This means that debonded areas at the fiber–matrix interface form at the low stress levels that ease initiation of crazes. In fact, fiber ends are good places for craze initiation

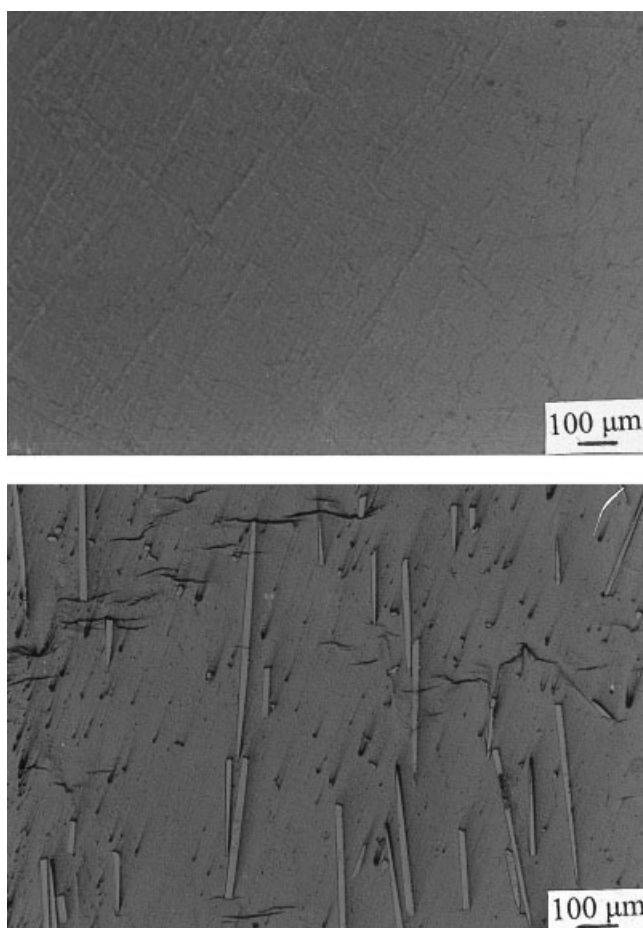


Figure 4 Optical microscopy of polished surfaces of tensile specimens after some deformation showing the damage in (a) pure PP and (b) 20 wt % PP of treated GF. The loading direction is vertical in both cases.

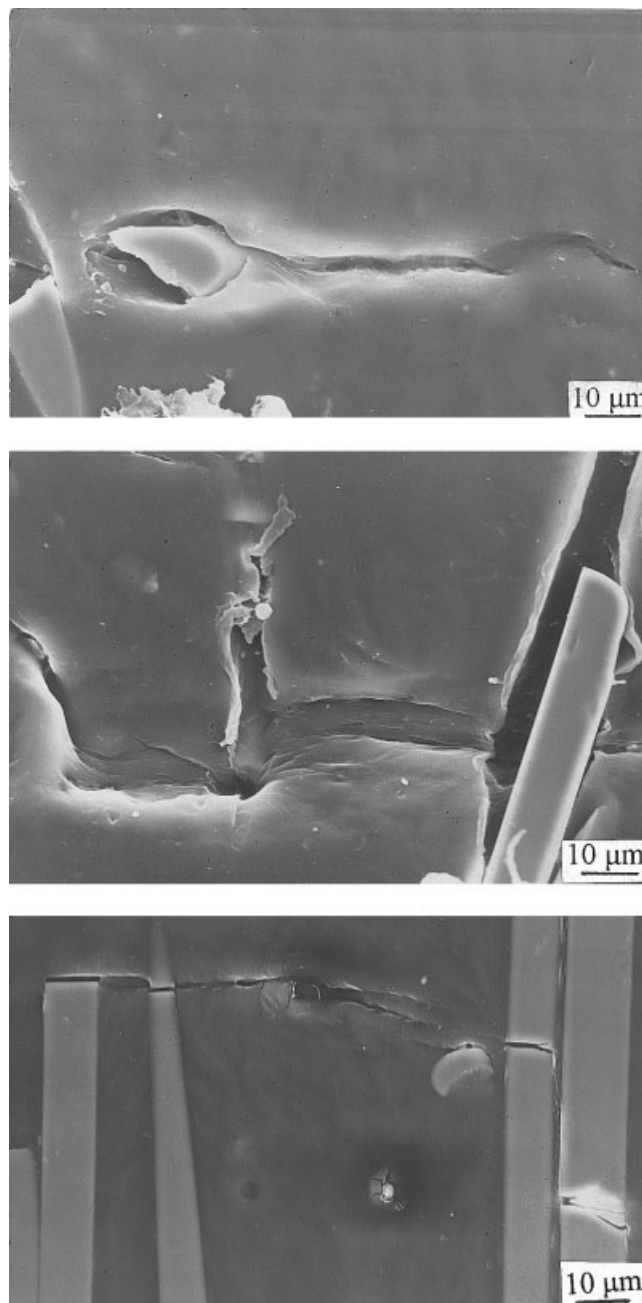


Figure 5 SEM micrographs taken from the polished surfaces of tensile specimens showing (a) initiation of a craze and, after some deformation, for: (b) standard GF, (c) treated GF. The loading direction is vertical.

because they act as stress concentrators as a result of their weak bonding to the matrix.^{8,26} Further stretching of the sample causes crazes to grow at the fiber–matrix interface and the pulling out of fibers in the absence of interfacial adhesion [Fig. 5(b)]. However, if there is strong adhesion at the interface, the crazes grow perpendicular to the direction of the applied load [Fig. 5(c)] and then try to break the fibers. In the latter case, glass fibers have a better chance to bridge

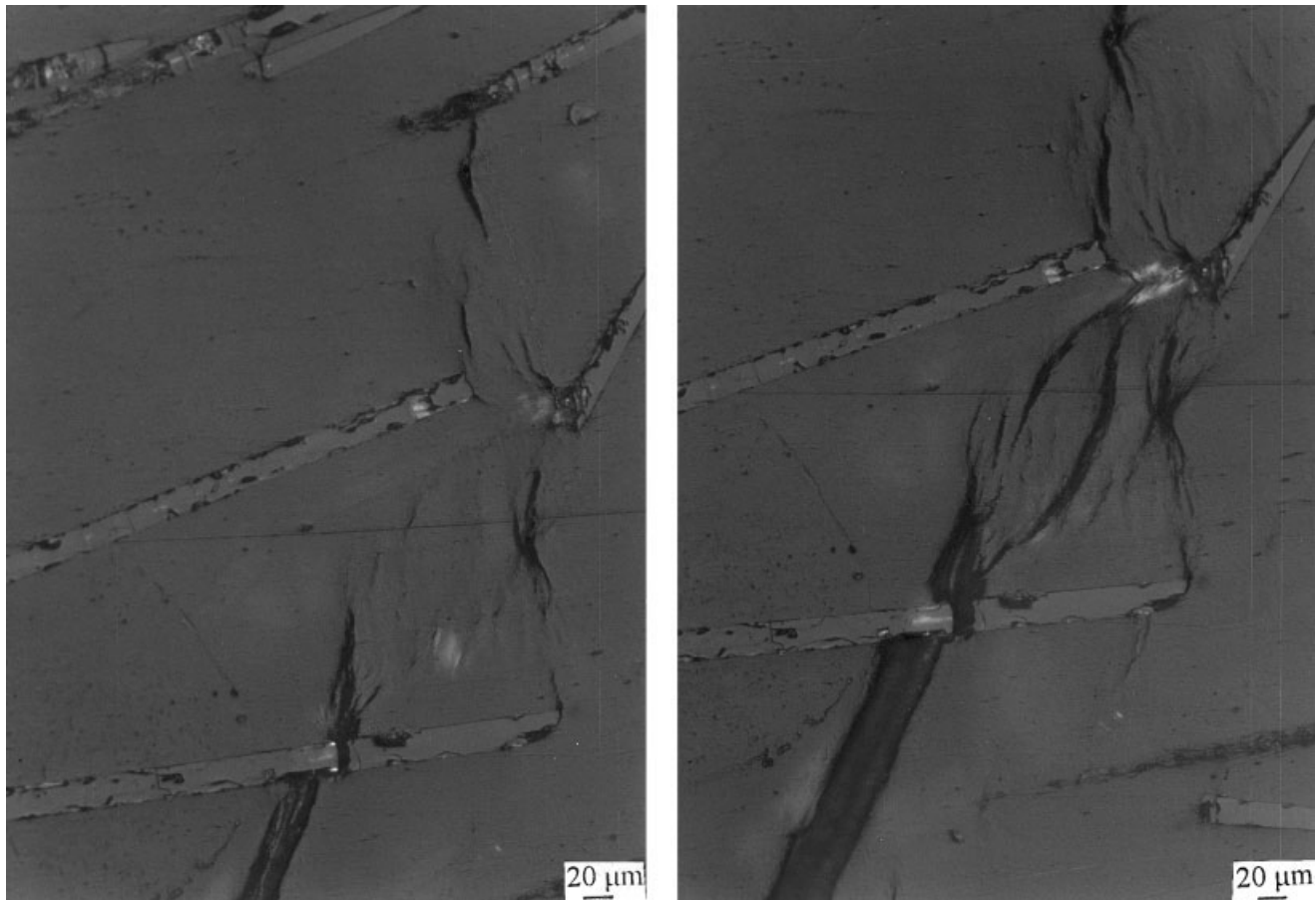


Figure 6 Steps of initiation and thickening of craze: (a) at beginning of deformation, (b) after further stretching of the specimen.

the craze wake and suppress crack formation and growth.

For further elucidation of craze growth in this system, additional optical microscopy was employed. Figure 6(a,b) shows the interaction between a growing craze and a glass fiber in the composite containing 20 wt % of treated fibers. As seen in Figure 6(a), the glass fiber stops the craze from growing. Further propagation of the craze needs additional energy to overcome the barrier. This energy provided by further stretching of the sample causes extra crazes to form in the vicinity of the original craze. As seen in Figure 6(b), further loading of the sample causes the new crazes touch each other and grow in the matrix. Figure 7 illustrates linkage of neighboring crazes in another position of the same specimen. Meantime, the original craze, which was stopped by the fiber, thickens with limited longitudinal growth [Fig. 6(a,b)].

The results of this study revealed no considerable influence of interfacial adhesion at the fiber–matrix interface on the craze thickening mechanism. This could be because despite the crazes growing into the interface in standard fibers [Fig. 6(b)], the growth of

the crazes in a direction different from the normal direction also needs extra energy. Thickening of the original crazes may be caused by this extra energy input, and so whether the interfacial adhesion at the fiber–matrix is strong or not, thickening occurs after some craze extension. According to Kramer,²⁷ the craze-thickening mechanism involves separating the undeformed matrix to generate fibrils and join the neighboring deformed zone. Therefore, it can be concluded that the influence of fibers on craze stoppage causes a thickening mechanism in this system similar to that proposed by Kramer.²⁷

CONCLUSIONS

To clarify the role of glass fiber on mechanical properties and mechanism of deformation in a polypropylene matrix, a set of tensile tests and microscopy work was performed. The results of this work showed the following:

1. Pure PP showed shear yielding under a static loading condition, and PP–GF composite crazed under similar conditions.

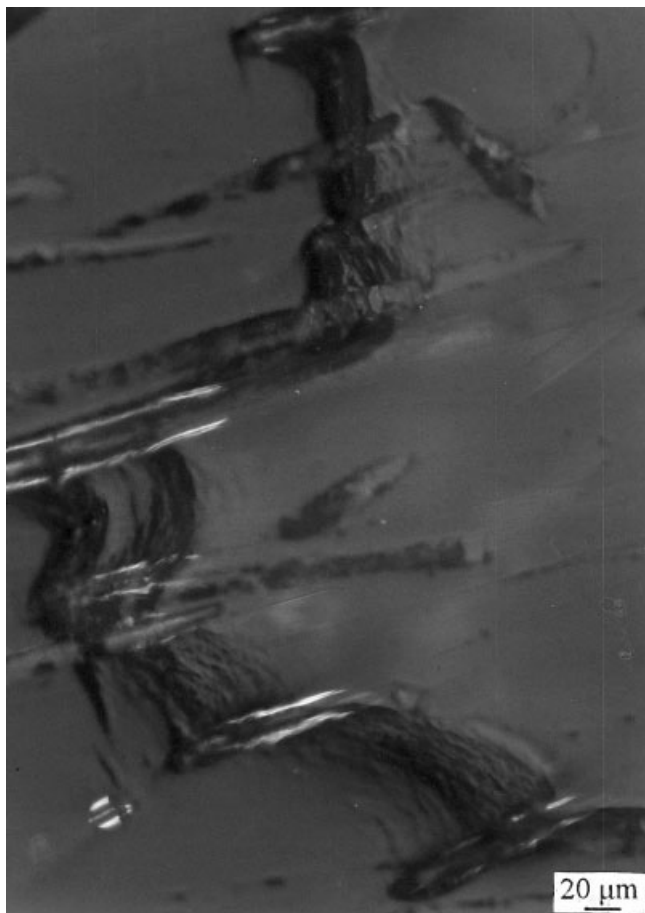


Figure 7 Linkage of crazes which is formed near the glass fibers.

2. At a weak interface the craze deviated toward the interface, and at a strong interface the craze grew in its original direction, causing breaking of fibers.
3. Craze thickening occurred as a result of craze-fiber interaction. Apparently, there was no influence of interfacial adhesion on this phenomenon.

References

1. McCrum, N. G.; Buckley, C. P.; Bucknall, C. B. *Principles of Polymer Engineering*, 2nd ed. Oxford University Press: New York, 1997.
2. Woishnis, W. *Polypropylene*; PDI Publisher: New York, 1998.
3. Ogoczkiewicz, R. *Engineering Properties of Thermoplastics*, 1st ed.; Wiley: New York, 1970.
4. Karger-Kocsis, J. *Polypropylene Structure, Blends and Composites*, 1st ed.; Chapman and Hall: London, UK, 1995.
5. Amash, A.; Zugenmaier, P. *J Appl Polym Sci* 1997, 63, 1143.
6. Qiu, W.; Mai, K.; Zeng, H. *J Appl Polym Sci* 1999, 71, 1537.
7. Clegg, D. W.; Collyer, A. A. *Mechanical Properties of Reinforced Thermoplastics*; Elsevier: London, UK, 1986.
8. Yue, C. Y.; Cheung, W. L. *J Mater Sci* 1992, 27, 3843.
9. Wulin, Q.; Kancheng, M.; Hanmin, Z. *J Appl Polym Sci* 1999, 71, 1537.
10. Xiaodong, Z.; Gance, D.; Wenjun, G.; Qunfang, L. *J Appl Polym Sci* 2000, 76, 1359.
11. Lee, N. J.; Jang, J. *Comp Sci Tech* 1997, 57, 1559.
12. Wu, H. F.; Dwright, D. W.; Huff, N. T. *Comp Sci Tech* 1997, 57, 975.
13. Folkes, M. J.; Hardwick, S. T. *J Mater Sci Lett* 1990, 25, 2598.
14. Rijdsdijk, H. A.; Contant, M.; Drazal, L. T. *Comp Sci Tech* 1993, 48, 161.
15. Nagae, S.; Otsuka, Y.; Nishida, M.; Shimizu, T.; Takeda, T.; Yumitori, S. *J Mater Sci Lett* 1995, 14, 1234.
16. Cai, Y.; Petermann, J.; Wittich, H. *J Appl Polym Sci* 1997, 65, 8995.
17. Moon, C. K. *J Appl Polym Sci* 1998, 67, 1191.
18. Lee, Y. C.; Porter, R. S. *Polym Eng Sci* 1986, 26, 633.
19. Bessell, T.; Shortall, J. B. *J Mater Sci* 1975, 10, 2035.
20. Moon, C. K.; Um, Y. S.; Cho, H. H.; Lee, J. O.; Park T. W. *Polymer* 1990, 14, 630.
21. Li, J. *Polym Eng Sci* 1984, 24, 895.
22. Peterlin, A. In *Ultra-high Modulus Polymers*; Ciferri, A, Ward, I.W., Eds.; Appl Sci Pub: London, 1979; p 279.
23. Lindhagen, J.; Berglund, L. *J Appl Polym Sci* 1998, 69, 1319.
24. Lee, N.; Jang, J. *J Comp Sci Tech* 1997, 57, 1559.
25. Zhang, S. Y. *J Comp Sci Tech* 1998, 58, 163.
26. Zebarjad, S. M.; Bagheri, R.; Lazzeri, A. *Plast, Rubber Compos Process Appl* 2001, 30, 370.
27. Lautterwasser, B. D.; Kramer, E. J. *Philos Mag A* 1979, 39, 469.