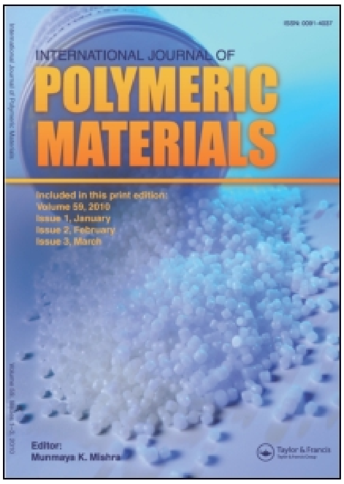

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

Influence of Epolene G-3003 as a Coupling Agent on the Mechanical Behavior of Palm Fiber-Polypropylene Composites

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To cite this Article Kahraman, R. , Abbasi, S. and Abu-Sharkh, B.(2005) 'Influence of Epolene G-3003 as a Coupling Agent on the Mechanical Behavior of Palm Fiber-Polypropylene Composites', International Journal of Polymeric Materials, 54: 6, 483 — 503

To link to this Article: DOI: 10.1080/00914030390278293

URL: http://dx.doi.org/10.1080/00914030390278293

International Journal of Polymeric Materials, 54:483–503, 2005
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ISSN: 0091-4037 print/1543-5253 online
DOI: 10.1080/00914030390278293

Influence of Epolene G-3003 as a Coupling Agent on the Mechanical Behavior of Palm Fiber-Polypropylene Composites

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Composites of palm fiber and polypropylene were compounded using a mixing equipment connected to an extruder. The composites were then injection molded into standard tensile specimens for mechanical characterization. The fracture morphology of the specimens was also analyzed by Scanning electron microscopy. It was observed that as the fiber content increases the composite modulus also increases, which is an indication for the existence of adhesion to some degree between polypropylene and the much stiffer palm fiber. However, the adhesion is not satisfactory, resulting in decrease in composite tensile strength with fiber addition. The compatibilizer Epolene G-3003 was used to minimize this incompatibility between the wood fibers and the polypropylene matrix. Utilizing Epolene G-3003 improved the fiber-matrix adhesion, resulting in a significant improvement in composite performance. The composite strength with 40 wt% fiber content and 6 wt% compatibilizer almost reached the strength of pure polypropylene.

Keywords: cellulose, palm, polypropylene, composite, compatibilizer, maleated polypropylene

Received 20 September 2003; in final form 27 September 2003.

This project has been funded by King Abdulaziz City for Science and Technology under Project # ARP 18-14. The authors also gratefully acknowledge the assistance of King Fahd University of Petroleum and Minerals, in particular the Research Institute, in the performance of this work.

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INTRODUCTION

Composite materials based on fibers of natural polymers, such as wood cellulose fibers and thermoplastics recently attracted much attention due to the remarkable environmental and economical advantages. The primary advantages of using cellulosic fibers as reinforcements in thermoplastics can be listed as low densities, low cost, nonabrasive nature, possibility of high filling levels, low energy consumption, high specific properties, biodegradability, availability of a wide variety of fibers throughout the world, and generation of a rural/agricultural-based economy [1–10]. It has been reported that, in the building community, there is a growing demand for high-performance, low-maintenance, and low-cost building components [11]. To meet this demand, natural fiber–thermoplastic composites are being used to produce such products as decking, window and door elements, panels, roofing, and siding. There has also been a rapid growth in the extrusion business with the use of wood-filled compounds for buildings and construction [12]. The better provision for longevity, appearance, life-cycle cost, and value makes wood–plastics composites popular in use in the markets. Wood-plastics are resistant to insect, rotting, slip, and are attractive and paintable and are stiffer than plastics and used like wood. The extruded composites can eliminate sizing and calibration. This is because the stiff extrudate can hold its shape much better than unfilled plastics. Engineered wood materials, that is, wood–plastic composites (WPCs), are also being investigated for waterfront construction applications primarily because of their superior durability characteristics compared to wood [13].

Polypropylene/wood fiber composites have attracted special attention because of their wide applicability in automobile and panel manufacturing applications. These composites take advantage of the superior properties of polypropylene in comparison to other thermoplastics, including easy processibility by all processing methods (molding, extrusion, film and fiber manufacturing). In addition, polypropylene is far superior to polyethylene in terms of heat resistance and mechanical properties. Its low density makes it especially attractive in lightweight applications that require strength. Furthermore, PP composites can be used in electrical applications because of their excellent electrical properties.

There are, however, a few disadvantages of using cellulosic fibers in thermoplastics such as the high moisture absorption of the fibers [14–15], the low processing temperatures permissible [2, 16–17], and the incompatibility of hydrophilic cellulose fibers and typical hydrophobic commodity thermoplastics, such as polyolefins [2, 15–19].

The moisture absorbed by the composite and the corresponding dimensional changes can be reduced dramatically if the fibers are thoroughly encapsulated in the plastic and the adhesion between the fiber and the matrix is strong [15, 17]. If necessary, moisture absorption of the fibers can be significantly reduced by the acetylation of the hydroxyl groups present in the fiber [20]. The shortcomings of the moisture absorption of the composite can also be minimized by selecting applications where the high moisture absorption is not a major drawback. For example, polyamide and its composites absorb water, but applications are such that this deficiency is not of prime importance [17].

The processing temperature of the cellulosic fibers in thermoplastics is limited due to the potential fiber degradation at higher temperatures. This just limits their application to plastics with low melting temperatures. However, it has been reported that no deterioration of properties are observed when processing temperatures are maintained below about 200°C [17]. It has also been noted that if the composite compositions are treated with sodium borate, boric acid, or phenolic resin, the chance of burning of the compositions during processing can be decreased [21–22].

The inherent polar and hydrophilic nature of the cellulosic fibers and the non-polar characteristics of polyolefins create difficulties in compounding and result in inefficient composites. However, it has been shown that the use of compatibilizing and coupling agents for treating fibers prior to, or as an addition in, the compounding step enhances the compatibility and adhesion between the fibers and the matrix and the fiber dispersion in the matrix, thus improving the mechanical properties [23–33].

Various chemical reagents have been employed to enhance the compatibility between the constituent materials. These include Epolene G-3002 [34], Epolene E-43 (Maelic anhydride modified polypropylene) [18], poly[methylene(polyphenyl isocyanate)] (PMPPIC), γ-methacryloxypropyltrimethoxysilane, poly(propylene acrylic acid), and poly (propylene-ethylene acrylic acid) [35].

Maleated (maleic-anhydride-modified) polypropylene has been particularly successful as a coupling agent in cellulose-polypropylene composites, improving mechanical properties as a consequence of enhanced interfacial adhesion [2, 17, 18, 21, 27, 36]. It has been reported that cellulose fibers treated with the copolymer turned totally hydrophobic due to the concentration of a considerable amount of copolymer on the fiber surfaces [2]. The copolymer was bonded to the fibers by ester linkages and hydrogen bonds accounting for the adhesion in the system.

Some other interface modifiers used as effective compatibilizers for polypropylene-wood fiber composites include bismaleimide- and itaconic anhydride–modified polypropylene [21], *m*-phenylene bismaleimide [16], and silane coupling agents, such as vinyl-tris(2-methoxy ethoxy) [31], with or without maleated polypropylene coating. Corona treatment of the cellulose fibers might also substantially improve the composite properties [28, 37]. Dynamic crosslinking of a maleated polypropylene compatibilized and thermosetting resin-coated cellulose-filled polypropylene composition has also been reported to produce a thermoplastic composite with tensile strength higher than that of unfilled polypropylene [38].

Isocyanates, such as poly[methylene(polyphenyl isocyanate)] (PMPIC), have been used successfully as compatibilizing agents in wood fiber–polystyrene and wood fiber–polyvinyl chloride (PVC) composites [39–40]. They have been observed to be more effective when used together with maleic anhydride in the presence of benzoyl peroxide for the case of wood fiber–polystyrene composites [39]. Pretreating the fibers with a silane agent and a polyisocyanate has been reported to improve the interfacial adhesion in wood fiber–polyethylene composites [41]. It has also been reported that polymethyl methacrylate (PMMA)–grafted cellulose fibers improved the mechanical properties of plasticized PVC composites [42].

Oxidation and darkening occur during the processing of composites made from thermoplastic matrices and cellulosic reinforcements. Urreaga et al. [43] investigated the effects of coupling agents on the oxidation and darkening of cellulosic materials used as reinforcements for thermoplastic matrices in composites. A maleated polypropylene wax (Epolene E-43) and two silanes (N-2-aminoethyl-3-aminopropyltrimethoxysilane and methyltrimethoxysilane) were used as coupling agents. Epolene wax E-43 produced scarce effects on both the oxidation and darkening of cellulosic materials at 200°C. Only for longer oxidation times was an increase in oxidation and darkening observed in E-43-treated samples. Silane coupling agents inhibited the formation of carbonyl and carboxyl groups for shorter oxidation times.

It was the objective of this study to investigate the feasibility in terms of mechanical properties for utilizing wood fibers obtained from waste palm tree branches in reinforcing polypropylene with Epolene G-3003 (a maleic-grafted polypropylene) as the compatibilizer. Palm trees are abundant in the Kingdom of Saudi Arabia and polypropylene is locally produced by Saudi Basic Industries Corporation (SABIC). If shown to be feasible, the utilization of waste palm tree branches in reinforcing thermoplastics will be an economical and useful service to the environment and society.

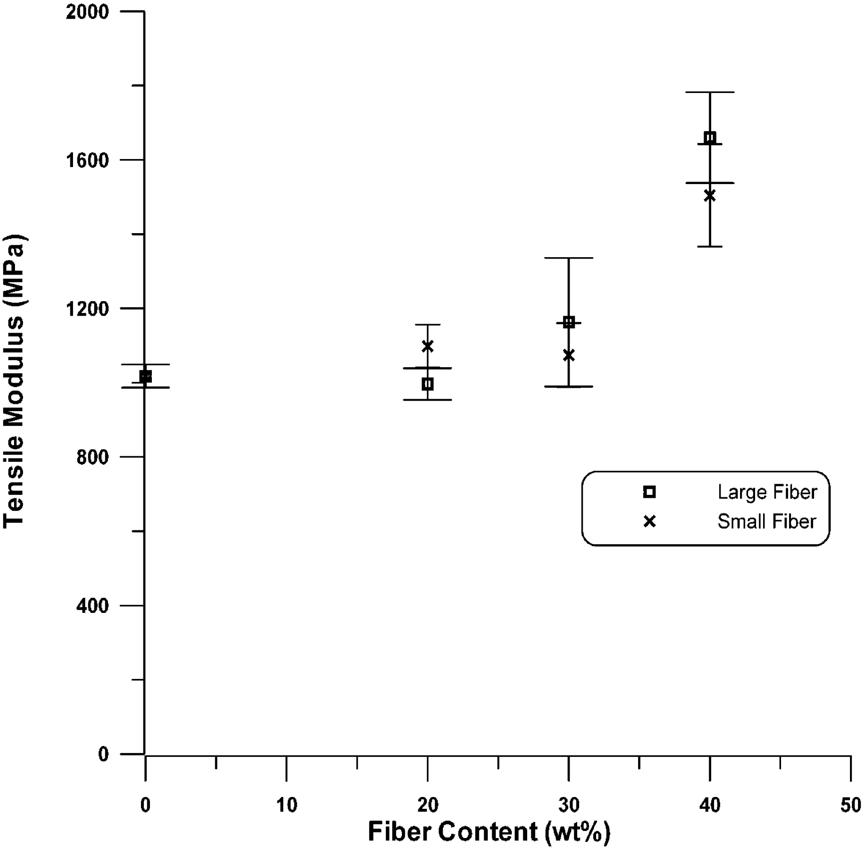

FIGURE 1 Tensile modulus vs. fiber content for polypropylene reinforced with small and large palm fiber.

EXPERIMENTAL

Materials

Polypropylene used in the study was supplied by Saudi Basic Industries Corporation. The brand name for it is Ladene PP570P. It is a homopolymer for producing rigid injection-molded articles. It is particularly suited for molding houseware items.

The compatibilizer used in the study is Epolene G-3003 supplied by Eastman Chemicals. Epolene G-3003 is a maleic-grafted polypropylene having optimum functionality and molecular weight to minimize phase separation between polar reinforcement/fillers and non-polar polymers in filled composites.

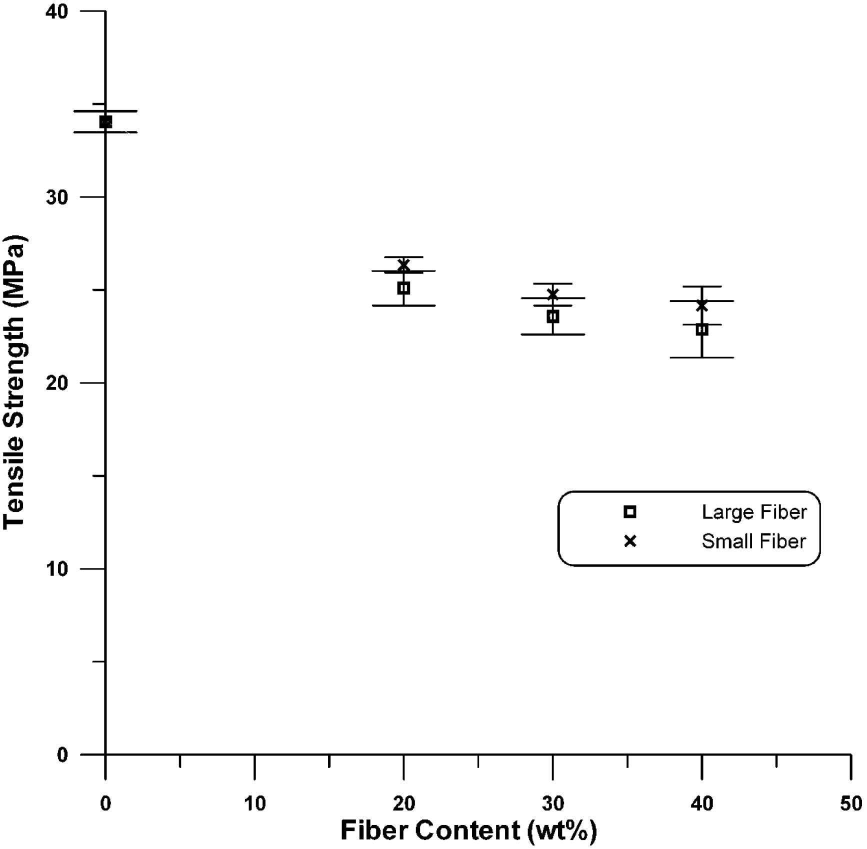

FIGURE 2 Tensile strength vs. fiber content for polypropylene reinforced with small and large palm fiber.

Fiber Preparation

Branches obtained from palm trees were cut into pieces about 6 in long. Thereafter they were dried in the sun for a few days so that most moisture was removed. The pieces of branches were then granulated to a small size using a granulator. The fibers were then size separated by using a sieving machine. Two different size distributions of fibers (called large and small fibers) were used for study. The fiber size distributions were characterized using a digital vernier caliper. The lengths of the fibers were determined to be 4.77 ± 1.75 mm and 2.96 ± 1.19 mm for large and small fibers, respectively. They both had aspect ratio of about 11.

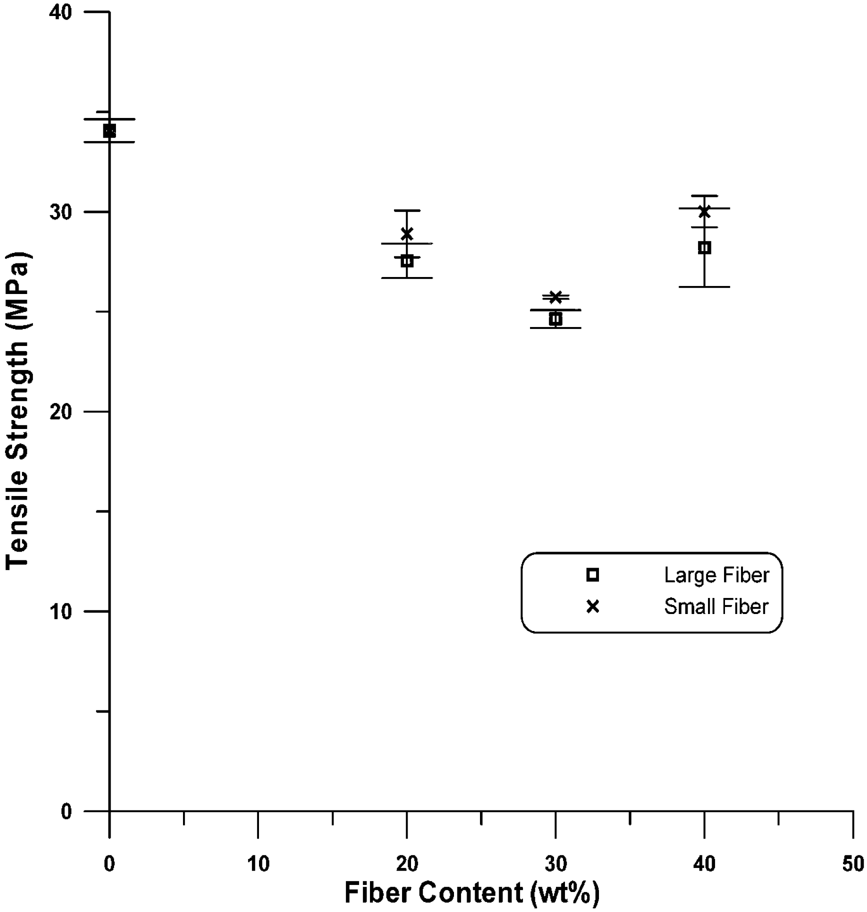

FIGURE 3 Tensile strength vs. fiber content for palm fiber-polypropylene matrix composites compatibilized by 2 wt% Epolene G-3003.

The fibers obtained were then cleaned to remove low molecular weight, soluble organic compounds. The chemicals used for cleaning were ethanol and toluene in the ratio of (1:2) (V/V). The mixture of these two chemicals was prepared in a large container. The fibers were then added to the mixture and kept in it for about two days. The fibers were then washed with water and put in an oven at 80°C to remove the moisture.

Composite Processing

Mixing of the composite components was done by a mixing device (Brabender Measuring Head, Model S-650, No:G126). This equipment

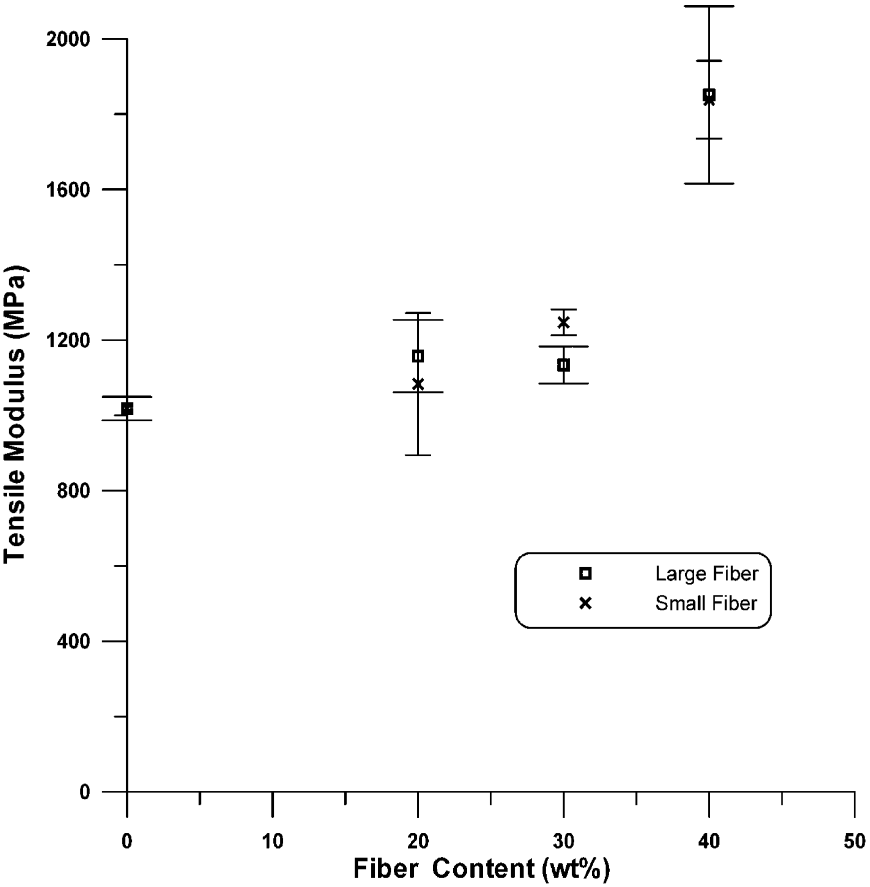

FIGURE 4 Tensile modulus vs. fiber content for palm fiber-polypropylene matrix composites compatibilized by 2 wt% Epolene G-3003.

was mounted with a single screw extruder. The fibers and the resins were first hand mixed in a bowl and then transferred to the extruder. Through preliminary tests, optimum processing conditions that will produce samples with maximum dispersion of fibers, good mechanical properties, good color and smell were determined. The mixed composite thus obtained from the extruder had a lumpy shape. It was granulated and transferred into a molding machine (Engel Electronics Model ES 80/25 ST pressure 160 bar, T = 200°C) to obtain tensile specimens of ASTM test standard No. D-638–94B.

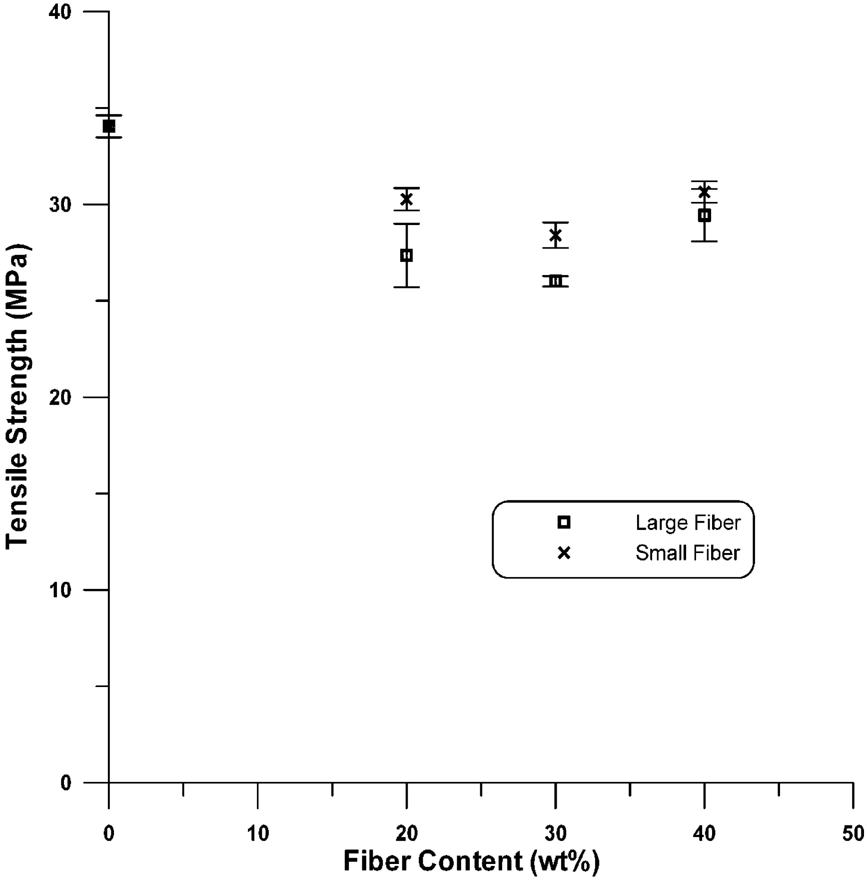

FIGURE 5 Tensile strength vs. fiber content for palm fiber-polypropylene matrix composites compatibilized by 4 wt% Epolene G-3003.

Material Characterization

The samples were mechanically characterized using an Instron 5560 Mechanical Testing Machine according to the ASTM test standard D-638. The tests were conducted at constant strain rate of 2 mm/min.

Scanning electron microscope (JEOL-JSM-T-300) was used to analyze the fracture surface of the composites from the tensile tests. The objective was to get information regarding the effect of the compatibilizer on fiber dispersion and adhesion quality between fibers and the matrix. JEOL-Fine Coat Ion Sputter was used to coat a thin layer of gold on the specimen to avoid electrostatic charging during

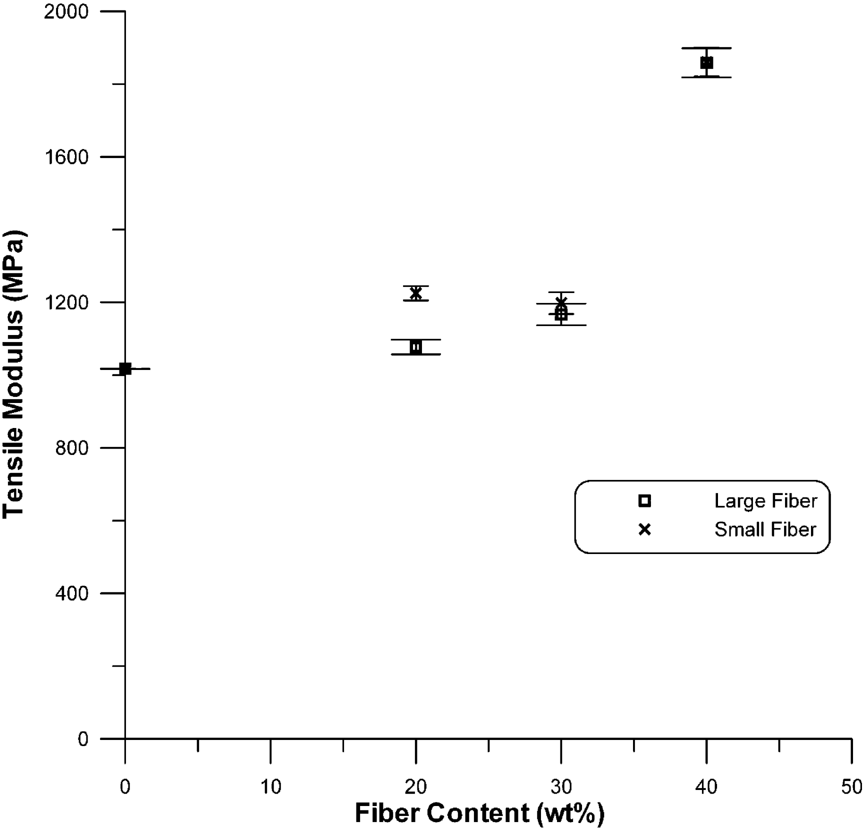

FIGURE 6 Tensile modulus vs. fiber content for palm fiber-polypropylene matrix composites compatibilized by 4 wt% Epolene G-3003.

examination. The fracture ends of the specimens were thereafter mounted on aluminum stub for analysis.

RESULTS AND DISCUSSION

Figure 1 shows the effect of fiber size and content on the tensile modulus of palm–polypropylene composite. As seen, the increase in fiber content increases the composite modulus for both small and large fiber, which is an indication for the existence of adhesion to some extent between polypropylene and the much stiffer palm fiber.

Figure 2 shows the effect of fiber size and content on the tensile strength of palm fiber–polypropylene composite. As the fiber content

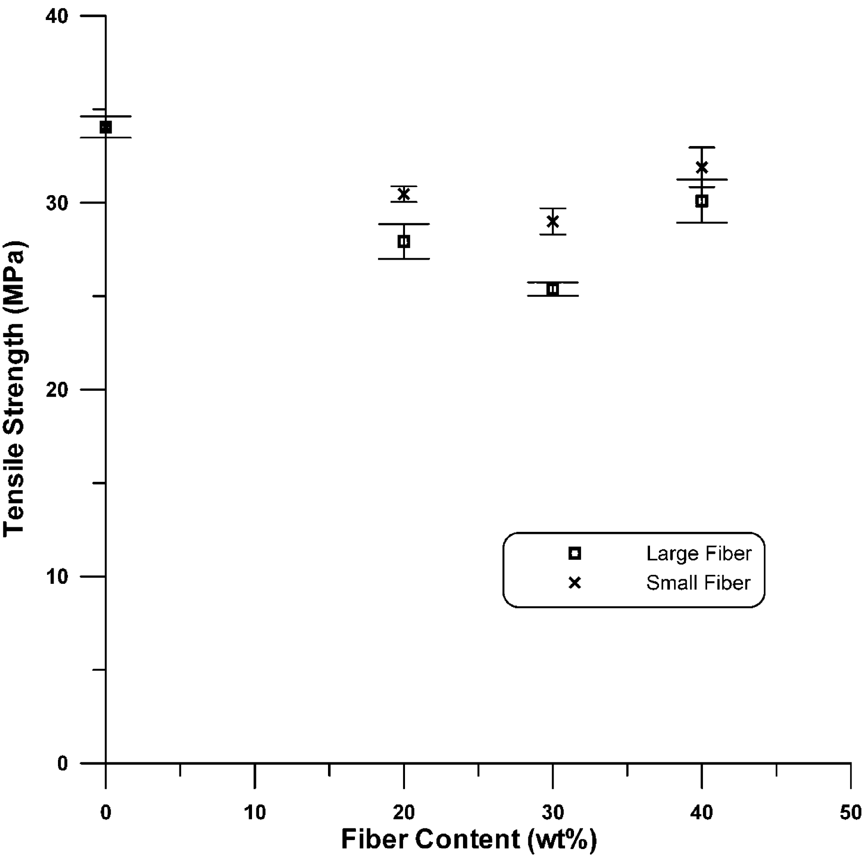

FIGURE 7 Tensile strength vs. fiber content for palm fiber-polypropylene matrix composites compatibilized by 6 wt% Epolene G-3003.

is increased the tensile strength decreases. This might be attributed to the poor adhesion between the fiber and polypropylene. This result does not contradict the improvement in composite stiffness with fiber addition shown in Figure 1. The modulus is related to the stiffness of the material at small deformations (before fracture) and it is obtained from the slope of the straight portion of the stress-strain curve. Existence of adhesion (even weak) between fibers and the matrix would then improve composite modulus. However, composite tensile strength is a result of material fracture at the weakest point of the material, which might occur below the matrix (polypropylene) strength if the adhesion between the fibers and the matrix fails at a lower stress.

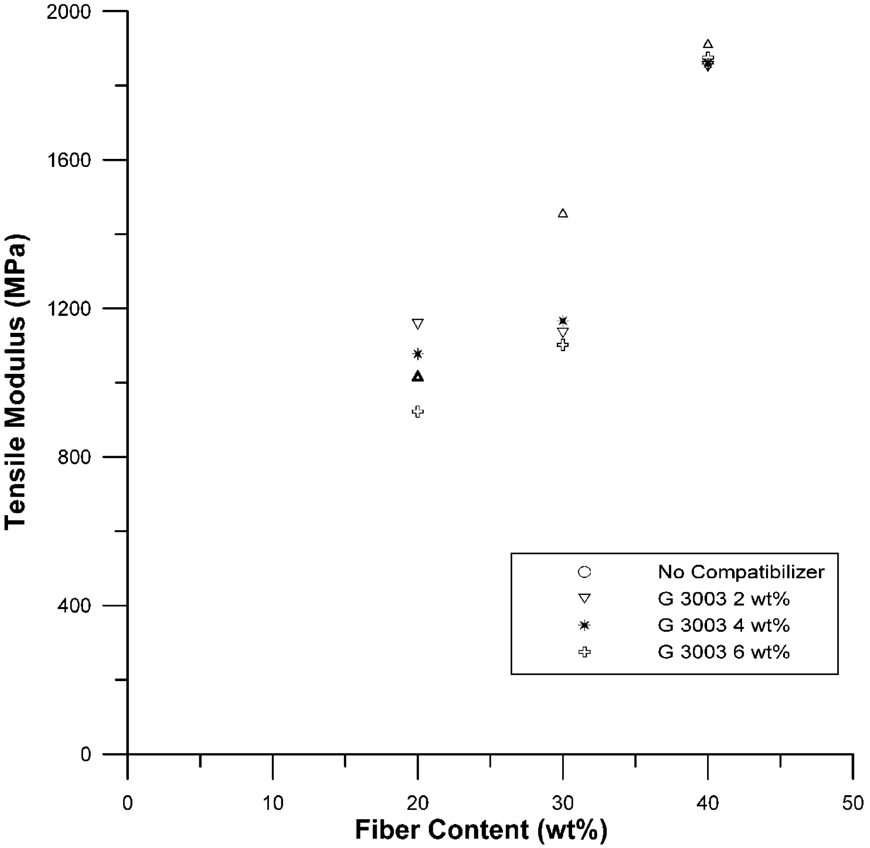

FIGURE 8 Tensile modulus vs. fiber content for polypropylene reinforced with large palm fibers without any treatment and with utilizing compatibilizer Epolene G-3003 at various amounts.

As also shown in Figures 1 and 2, the difference in the results obtained with small or large fibers was not significant considering also the scatter in data. But still the composite strength with small fiber was observed to be consistently higher than that with large fibers although the same effect was not observed in the case of composite modulus. This is probably because of better mixing and more uniform distribution of small fibers in the polypropylene matrix as compared to large fibers. Accumulation or non-uniform orientation of fibers at some parts of the composite can result in fracture at these weak points giving lower strength in the case of larger fibers. Non-uniformities in fiber distribution here and there (if not so extensive) would not affect

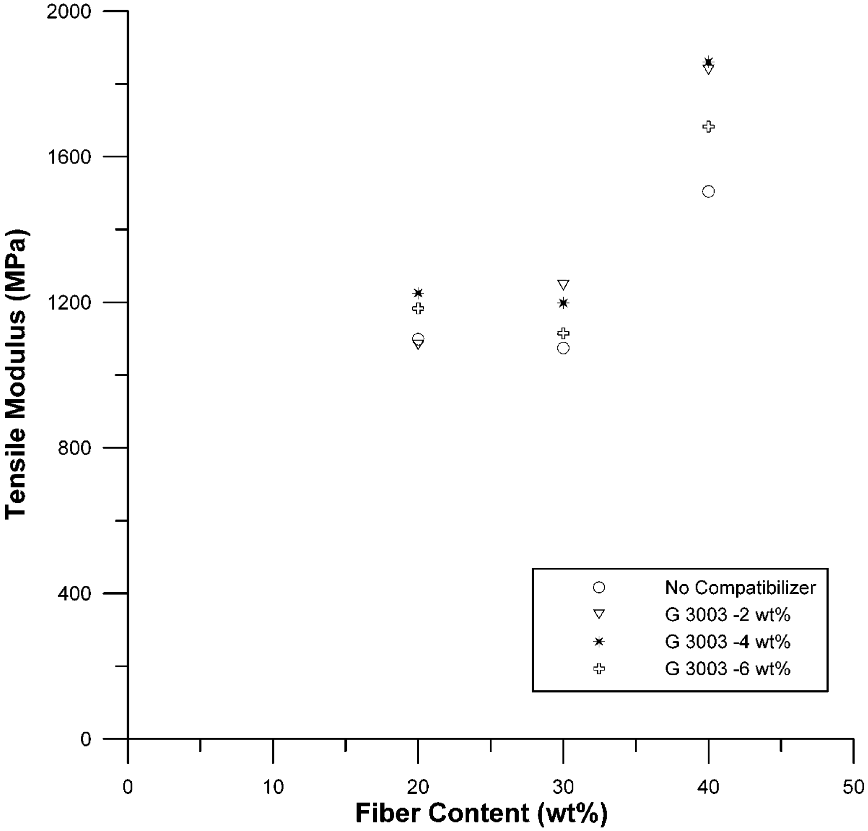

FIGURE 9 Tensile modulus vs. fiber content for polypropylene reinforced with small palm fibers without any treatment and with utilizing compatibilizer Epolene G-3003 at various amounts.

the composite modulus to the same extent because the modulus is the bulk material property before fracture.

The next step was to utilize the compatibilizer to see its effect on composite performance. Figures 3 and 4 show the effect of adding 2 wt% Epolene G-3003 on the tensile strength and modulus of the composite, respectively, with varying fiber loading and fiber length distribution. The composite behavior is similar to that of the uncompatibilized one. Again the tensile strength drops as fiber loading is increased. Small fibers result in somewhat better composite strength than large fibers, and the modulus increases with the increasing fiber loading. However, the noticeably higher composite strength values relative to that of uncompatibilized ones is an

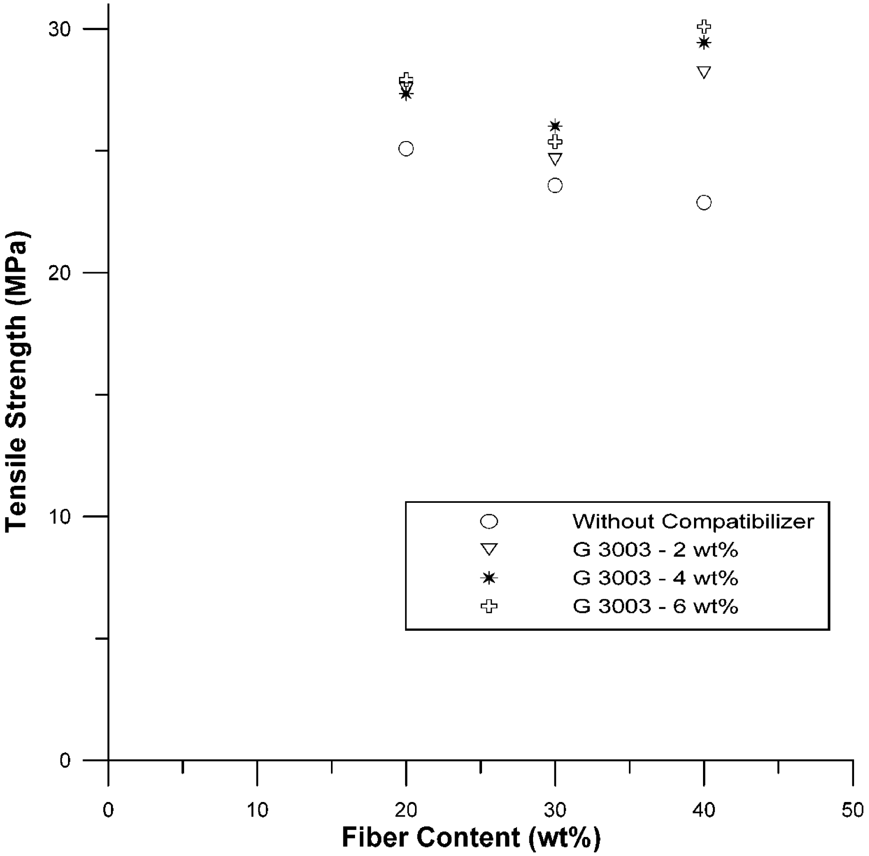

FIGURE 10 Tensile strength vs. fiber content for polypropylene reinforced with large palm fibers without any treatment and with utilizing compatibilizer Epolene G-3003 at various amounts.

indication of improvement in fiber-matrix adhesion with 2 wt% Epolene G-3003.

Figures 5 and 6 show the effect of 4 wt% Epolene G-3003 on composite strength and modulus with varying fiber loading and fiber length distribution. The composite behavior is similar to that of the composite compatibilized with 2 wt% Epolene G-3003 but with higher strength values obtained at each fiber content.

Mechanical performance of the composite improved further with increasing the compatibilizer content to 6 wt% (Figure 7) and the composite strength at 40 wt% fiber content (about 32 MPa for small fibers

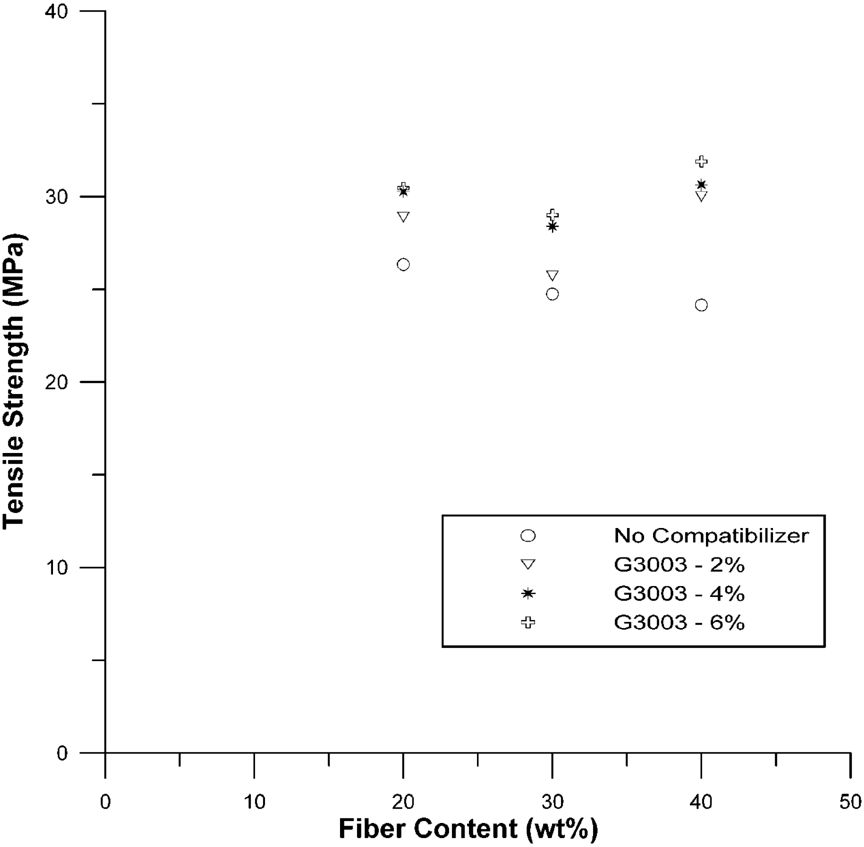

FIGURE 11 Tensile strength vs. fiber content for polypropylene reinforced with small palm fibers without any treatment and with utilizing compatibilizer Epolene G-3003 at various amounts.

and 30 MPa for large fibers) almost reached that of the unreinforced polypropylene (about 34 MPa).

The results discussed earlier were combined together in Figures 8–11 to show the effect of compatibilizer amount on the composite properties in a more convenient way for polypropylene reinforced with small and large palm fibers. As seen in Figures 8 and 9 composite modulus increases with fiber content for composites with the compatibilizer added at various amounts (2, 4, and 6 wt%) as well as for the ones with no compatibilizer. As also already discussed this is an indication for the existence of adhesion between palm fibers and polypropylene matrix even in the composites without any compatibilizer.

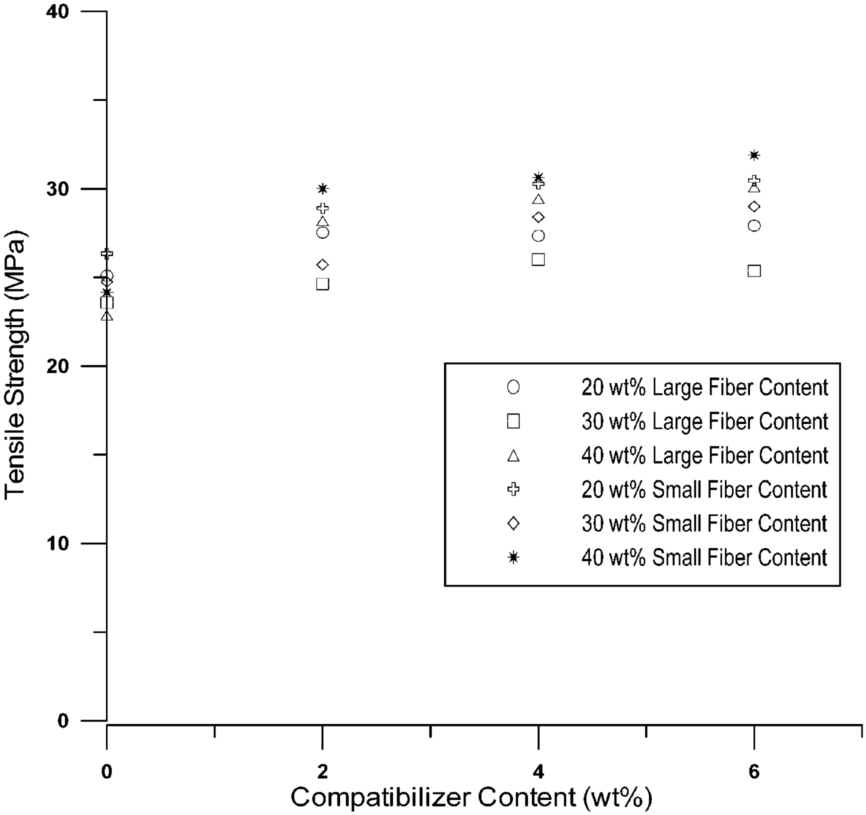

FIGURE 12 Tensile strength vs. compatibilizer content for polypropylene reinforced with palm fibers without any treatment and with utilizing compatibilizer Epolene G-3003 at various amounts.

However, the use of compatibilizer might have increased the adhesion strength, which is not clear in the plots of modulus versus fiber content but is quite clear in Figures 10 and 11, which present plots of tensile strength versus fiber content for uncompatibilized and compatibilized palm–polypropylene composite with small and large fibers. As seen, there is a significant improvement in tensile strength with addition of the compatibilizer.

The data presented earlier can be rearranged to be able to discuss the effect of varying compatibilizer content on the mechanical properties of the polypropylene composite. Figure 12 shows the tensile strength versus compatibilizer content for Epolene G-3003 with varying fiber loading. It is seen as a general trend for the two fiber length

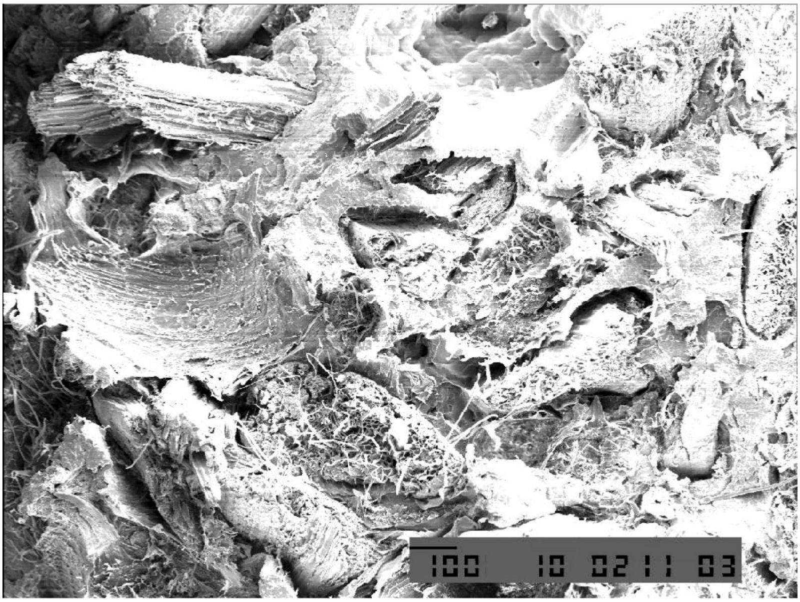

FIGURE 13 SEM micrograph of a fractured surface of a 30 wt% palm fiber-polypropylene composite with no compatibilizer.

FIGURE 14 SEM micrograph of a fractured surface of a 30 wt% palm fiber-polypropylene composite with no compatibilizer.

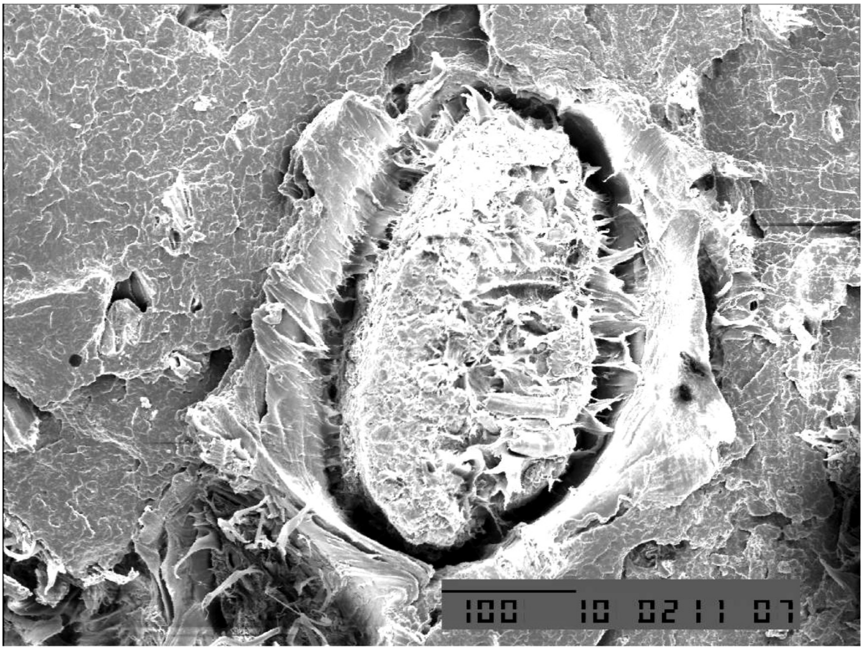

FIGURE 15 SEM micrograph of a fractured surface of a 30 wt% fiber palm-polypropylene composite compatibilized with 6 wt% Epolene G-3003.

distributions that increasing amount of compatibilizer content increases the tensile strength, which is an indication of a probable increase in fiber-matrix adhesion strength.

Fracture surfaces of the mechanically tested composite specimens were analyzed by scanning electron microscopy (SEM). It was seen that polypropylene composites containing no compatibilizer resulted in rough fracture surfaces with extensive fiber pullout (Figures 13 and 14). This is an indication of poor bonding between the fiber and the polypropylene matrix (as compared to that of the composites with compatibilizer treated fibers, discussion of which will follow). This explains why composites with untreated fibers perform worse than those with compatibilizer treated fibers.

Once the compatibilizer Epolene G-3003 was incorporated it was seen that instead of fiber pullout, a much smoother fracture surface was observed (Figures 15 and 16) as compared to that of the composite without the compatibilizer (Figures 13 and 14). The reason for this observation is probably that the incorporation of the compatibilizer G-3003 significantly enhanced the interfacial adhesion between the

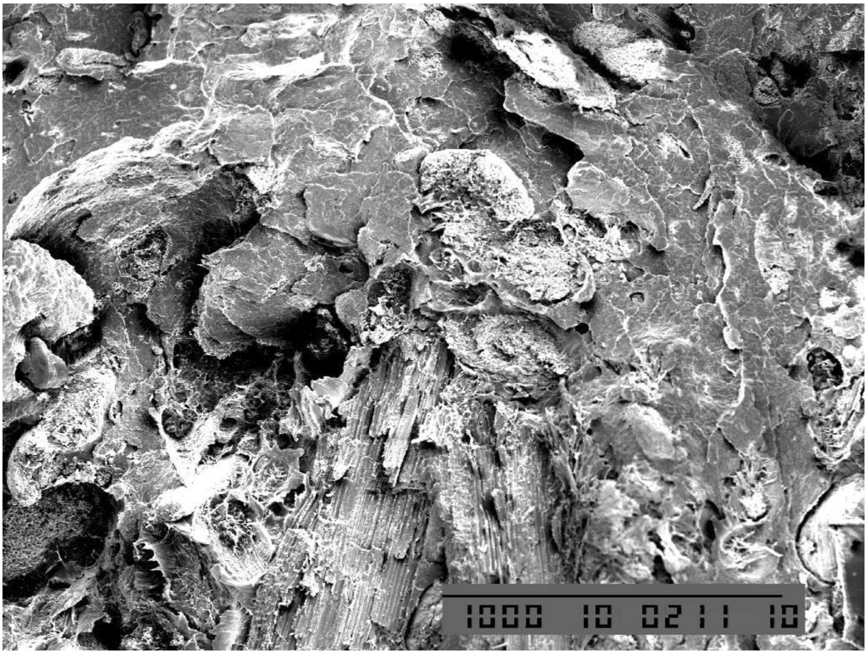

FIGURE 16 SEM micrograph of a fractured surface of a 30 wt% fiber palm-polypropylene composite with Epolene 6 wt% G-3003.

fibers and matrix That is why a higher load was transferred onto the fibers, hence the higher observed composite strength (as discussed earlier).

CONCLUSIONS

1. Existence of adhesion between the palm fiber and polypropylene to some degree resulted in increase in polypropylene modulus with fiber addition.
2. However, the fiber-matrix bond strength in palm–polypropylene system was not satisfactory, resulting in decrease in polypropylene strength with fiber addition.
3. Small fibers resulted in better composite performance due to better mixing and more uniform distribution of small fibers in the polypropylene matrix as compared to large fibers.
4. Utilizing the compatibilizer Epolene G-3003 improved the fiber-matrix adhesion, resulting in a significant improvement in composite performance.

5. Increase in compatibilizer content resulted in further increase in composite strength and the composite strength with 40 wt% fiber content and 6 wt% Epolene G-3003 reached close to the strength of unreinforced polypropylene.

REFERENCES

[1] Yam, K. L., Gogoi, B. K., Lai, C. C., and Selke, S. E., *Polym. Eng. Sci.* **30**(11), 693 (1990).

[2] Felix, J. M. and Gatenholm, P., *J. Appl. Polym. Sci.* **42**, 609 (1991).

[3] Chen, H.-S. and Porter, R. S., *J. Appl. Polym. Sci.* **54**, 1781 (1994).

[4] Jain, S. and Kumar, R., *Materials and Manufacturing Processes* **9**(5), 813 (1994).

[5] Sanadi, A. R., Young, R. A., Clemons, C., and Rowell, R. M., *J. Reinforced Plast. Compos.* **13**(1), 54 (1994).

[6] George, J., Bhagawan, S. S., Prabhakaran, N., and Thomas, S., *J. Appl. Polym. Sci.* **57**(7), 843 (1995).

[7] Devi, L. U., Bhagawan, S. S., and Thomas, S., *J. Appl. Polym. Sci.* **64**(9), 1739 (1997).

[8] Bourban, Ch., Karamuk, E., de Fondaumiere, M. J., Ruffieux, K., Mayer, J., and Wintermantel, E., *J. Environmental Polymer Degradation* **5**(3), 159 (1997).

[9] Mansour, O. Y., Kamel, S., and Nassar, M. A., *J. Appl. Polym. Sci.* **69**(5), 845 (1998).

[10] Matuana, L. M., Park, C. B., and Balatinecz, J. J., *Polym. Eng. Sci.* **38**(11), 1862 (1998).

[11] Falk, R. H., Vos, D. J., Cramer, S. M., and English, B. W., *Forest Products Journal* **51**(1), 55 (2001).

[12] Anon, *Modern Plastics* **78**(6), 49 (2001).

[13] Malvar, L. J., Pendleton, D. E., and Tichy, R., *SAMPE Journal* **37**(4), 70 (2001).

[14] Sanadi, A. R., Caulfield, D. F., and Rowell, R. M., *Plast. Eng.* **April**, 27 (1994).

[15] Bledzki, A. K., Reihmane, S., and Gassan, J., *J. Appl. Polym. Sci.* **59**(8), 1329 (1996).

[16] Sain, M. M. and Kokta, B. V., *J. Appl. Polym. Sci.* **54**, 1545 (1994).

[17] Sanadi, A. R., Caulfield, D. F., Jacobson, R. E., and Rowell, R. M., *Ind. Eng. Chem. Res.* **34**, 1889 (1995).

[18] Felix, J. M. and Gatenholm, P., *J. Appl. Polym. Sci.* **50**, 699 (1993).

[19] Gatenholm, P., Bertilsson, H., and Mathiasson, A., *J. Appl. Polym. Sci.* **49**, 197 (1993).

[20] Rowell, R. M., Tillman, A. M., and Simonson, R. A., *J. Wood Chem. Tech.* **6**, 427 (1986).

[21] Sain, M. M., Kokta, B. V., and Imbert, C., *Polym.-Plast. Technol. Eng.* **33**(1), 89 (1994).

[22] Sain, M. M., Kokta, B. V., and Maldas, D., *J. Adhesion Sci. Technol.* **7**(1), 49 (1993).

[23] Raj, R. G., Kokta, B. V., Dembele, F., and Sanschagrain, *J. Appl. Polym. Sci.* **38**, 1987 (1989).

[24] Raj, R. G., Kokta, B. V., Grouleau, G., and Daneault, C., *Poly. Plast. Technol. Eng.* **29**(4), 339 (1990).

[25] Maldas, D. and Kokta, B. V., *J. Appl. Polym. Sci.* **41**(1–2), 185 (1990).

[26] Maldas, D. and Kokta, B. V., *Polymer J.* **23**(10), 1163 (1991).

[27] Felix, J. M., Gatenholm, P., and Schreiber, H. P., *Polym. Compos.* **14**(6), 449 (1993).

[28] Belgacem, M. N., Batasille, P., and Sapieha, S., *J. Appl. Polym. Sci.* **53**, 379 (1994).

[29] Garnett, J. L. and Ng, L. T., *Radiation Physics and Chemistry* **48**(2), 217 (1996).

[30] Canche, E. G., Rodriguez, T. G., Herrera, F. P., Mendizabal, E., and Puig, J. E., *J. Appl. Poly. Sci.* **66**(2), 339 (1997).

[31] Coutinho, F. M. B., Costa, T. H. S., and Carvalho, D. L., *J. Appl. Polym. Sci.* **65**(6), 1227 (1997).

[32] Herrera-Franco, P. J. and Aguilar-Vega, M. D. J., *J. Appl. Polym. Sci.* **65**(1), 197 (1997).

[33] Karnani, R., Krishnan, M., and Narayan, R., *Polym. Eng. Sci.* **37**(2), 476 (1997).

[34] Rana, A. K., Mandal, A., Mitra, B. C., Jacobson, R., Rowell, R., and Banerjee, N., *J. Appl. Polym. Sci.* **69**, 329 (1998).

[35] Rozman, H. D., Tan, K. W., Kumar, R. N., Abubakar, A., Ismail, H., and Ishak, Z. A. M., *European Polym. J.* **36**, 1483 (2000).

[36] Gauthier, R., Joly, C., Coupas, A. C., Gauthier, H., and Escoubes, M., *Polym. Composites* **19**(3), 287 (1998).

[37] Dong, S., Sapieha, S., and Schreiber, H. P., *Polym. Eng. Sci.* **33**(6), 343 (1993).

[38] Sain, M. M. and Kokta, B. V., *J. Appl. Polym. Sci.* **48**, 2181 (1993).

[39] Maldas, D. and Kokta, B. V., *J. Adhesion Sci. Technol.* **5**(9), 727 (1991).

[40] Maldas, D. and Kokta, B. V., *J. Test. Eval.* **21**(1), 68 (1993).

[41] Raj, R. G., Kokta, B. V., Groleau, G., and Daneault, C., *Plastics and Rubber Processing and Applications* **11**(4), 215 (1989).

[42] Canche-Escamilla, G., Cauich-Cupul, J. I., Mendizabal, E., Puig, J. E., Vazquez-Torres, H., and Herrera-Franco, P. J., *Composites-A* **30**(3), 349 (1999).

[43] Urreaga, J. M., Matias, M. C., De La Orden, M. U., Munguia, M. A. L., and Sanchez, C. G., *Polymer Engineering and Science* **40**(2), 407 (2000).