Composite Materials Prepared from Waste Textile Fiber

STUART A. BATEMAN, DONG YANG WU

CSIRO Building, Construction Engineering, P.O. Box 56, Highett, Victoria 3190, Australia

ABSTRACT: Composite materials were prepared utilizing whole carpet and LDPE. The addition of compatibilizers during processing was shown to provide an improvement in the mechanical performance of such materials. It is believed that the addition of a reactive compatibilizer to the system increases both the compatibility between components of the composite material and interfacial interactions, through chemical reactions between functional groups on the compatibilizer and the surface of the carpet fibers under the processing conditions. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 81: 3178–3185, 2001

Key words: carpet; polyolefin; composite; interfacial adhesion; reactive processing

INTRODUCTION

Waste fibers from the textile industry represent an environmental challenge considering that annually 2 billion kilograms of carpet is removed from homes and businesses in the United States alone, and 16 million kilograms of additional carpet waste is generated from carpet manufacture and laying.[1] The clothing industry also adds to this problem, with millions of kilograms of waste textiles generated from garment manufacture each year.[2,3] Unfortunately, 80% of textile waste is disposed to landfills, with the remainder recycled into products requiring low-performance characteristics such as soundproofing, padding, and insulation. This fact is somewhat surprising considering that virgin textile fibers generally provide high-performance characteristics and durability for their intended application.

With the tightening of government regulations regarding the use of landfills, new technologies and processes are emerging to better utilize these fibrous resources. Such technologies include depolymerization of synthetic carpet fibers via hydrolysis[4] or ammonolysis[5,6] to recover valuable monomers such as caprolactam. Following purification, the monomers may be repolymerized to form new polymers having similar performance characteristics to those prepared from virgin sources with a significant energy saving. Waste carpet has also been investigated as a wood substitute similar to fibrous particleboard,[2,3] for the reinforcement of concrete and earth,[7] and, recently, the reuse of polypropylene carpet has been investigated through injection-molding techniques.[1]

In this study, an alternative approach for the reuse of waste textiles was investigated whereby carpet fibers were utilized to reinforce polyethylene. Such materials have the potential to be used in the automotive and building industries. New methodology for the processing of carpet-containing composites using conventional plastics processing equipment was also developed. Such methodology avoids the need for carpet shredding and its associated problems of low bulk density

---

Table I   Chemical Composition of Compatibilizers Trialed

Code	Compatibilizer	Composition
PE-*co*-GMA	Polyethylene-*co*-glycidylmethacrylate	8% wt GMA
PE-*co*-MA-*co*-GMA	Polyethylene-*co*-methylacrylate-*co*-glycidyl methacrylate	25% wt MA 8% wt GMA
PE-*co*-EA-*co*-MAh	Polyethylene-*co*-ethylacrylate-*co*-maleic anhydride	9% wt EA 3% wt MAh
PE-*g*-MAh (**1**)	Polyethylene-*g*-maleic anhydride	0.85% wt MAh
PE-*g*-MAh (**2**)	Polyethylene-*g*-maleic anhydride	3% wt MAh

and feeding difficulties onto the screw of processing equipment.

It is well known that the addition of small amounts of compatibilizers during the processing of polymer blends or composites can improve the performance of the resulting material.[8–12] This improvement occurs due to a reduction in interfacial tension and an increase in adhesion between the phases, which allows more efficient stress transfer between the respective polymer domains or across the fiber–matrix interface.[8–12] Similarly, it was theorized that the addition of certain additives/compatibilizers during the processing of carpet fiber-reinforced polyethylene would improve its mechanical properties. Any improvement in performance brought about by the addition of such additives has the potential to greatly increase the range of applications available to carpet fiber–polyolefin composite materials.

EXPERIMENTAL

Materials

Low-density polyethylene (LDPE) of film-extrusion grade (LD4200) was obtained from Kemcor (Melbourne, Australia). Recycled LDPE flakes were obtained from waste-packaging material. The carpet utilized in the trial contained approximately 50% w/w nylon 6,6 face fibers, an

SBR and calcium carbonate adhesive, and a jute backing layer. All other chemicals including the different compatibilizers were obtained from the Aldrich Chemical Co. (Sidney, Australia). An abbreviation and compositional list of the compatibilizers trialed is provided in Table I.

Sample Preparation

Carpet squares (30 cm^2 approximately) were cut and laminated with recycled polyethylene in a Pongrass 16-MPa press equipped with water-cooled platens at 150°C. The laminated sheets were then sliced using a guillotine and pelletized using an Axxon AB pelletizer to obtain pellets of a suitable dimensions for extrusion.

Compounding of the carpet fiber–LDPE composite material was carried out on a single-screw Axxon R40F bench-top extruder with a vent stage using a 32 *L:D* ratio screw and the temperature profile indicated in Table II. Additives, LDPE, and the pelletized carpet laminate were dried at 60°C for at least 12 h and mixed thoroughly prior to extrusion using a Bear planetary mixer. The screw speed was set at 120 rpm, which correlated to a residence time of approximately 2 min. A single-aperture string die was utilized with the carpet composite string pelletized following cooling in a water bath. The temperature profile and residence time was chosen to obtain an acceptable extrudate without causing melting of the nylon face

Table II   Carpet Fiber–LDPE Composite Processing Conditions

Zone	Hopper	2	3	4	5	Die
Temperature (°C) extruder	170	185	190	205	205	205
Temperature (°C) injection molder	190	200	200	200	200	70

Figure 1 Variation in tensile modulus of carpet–LDPE composites with percentage carpet.
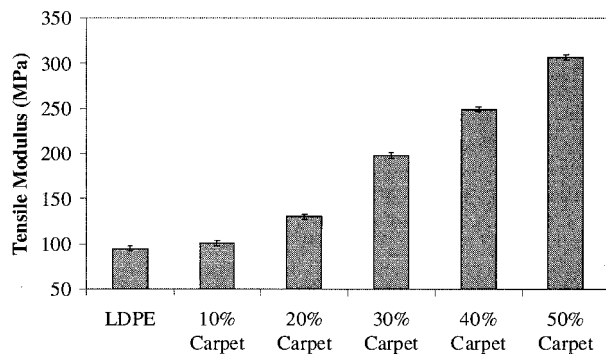

Figure 3 Variation in ultimate tensile strength of carpet–LDPE composites with percentage carpet.

fibers or excessive decomposition of the jute backing. These processing conditions were also intended to allow sufficient time and temperature for chemical reactions to occur between reactive functionalities on the carpet and compatibilizer if employed.

Tensile test specimens (ASTM D 638–97, type 1) were molded using an 80-tone Battenfeld injection molder (BA 800 CDC). The temperature profile for molding is also provided in Table II. An injection pressure of 1300 psi and a holding pressure gradient between 1000 and 1100 psi was used. The compounded samples were mixed thoroughly and dried at 60°C for at least 12 h in a desiccated oven prior to molding.

Tensile Testing

Tensile testing was carried out using an Instron tensile-testing apparatus (5565) utilizing a 5-kN load cell according to the ASTM D 638–97 stan-
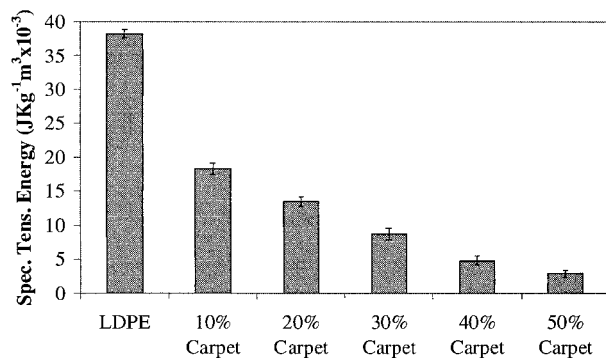
dard. A crosshead speed of 50 mm/min was used for all specimens employing an external extensometer with a minima of six specimens tested per sample. Ultimate tensile strength, elongation at break, and tensile modulus values were recorded and calculated. Specific tensile energy values for the composite materials, being derived from integration over the entire stress-versus-strain curve, are also shown. Composite density values were obtained by employing the ASTM D 792–91 test method for the density and specific gravity of plastics.

Scanning Electron Microscopy (SEM) and High-temperature Gel Permeation Chromatography (HTGPC)

Micrographs were recorded on a Philips XL 30 field-emission SEM. The carbon-coated samples were analyzed at a working distance of

Figure 2 Variation in specific tensile energy of carpet–LDPE composites with percentage carpet.

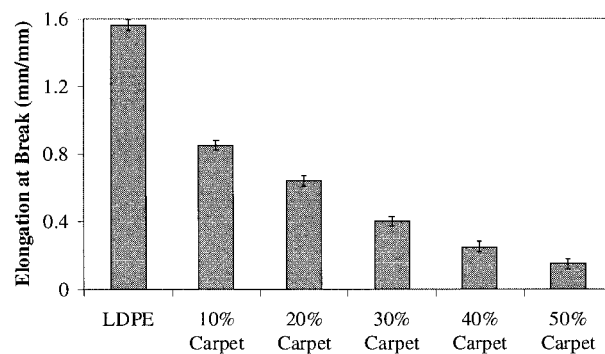**Figure 4** Variation in elongation at maximum load of carpet–LDPE composites with percentage carpet.
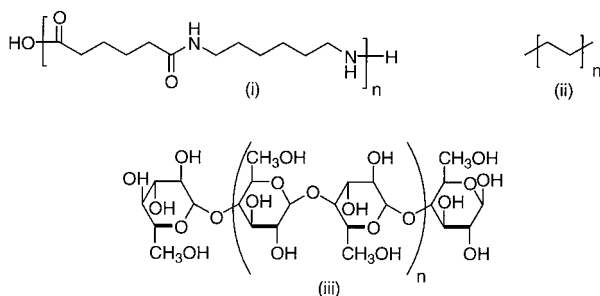
Figure 5   Structures: (**i**) nylon 6,6; (**ii**) PE; (**iii**) cellulose.

approximately 10 mm using an electron potential of 5 kV.

HTGPC was carried out using a Waters 150C chromatograph. Duplicate samples were prepared at a concentration of approximately 0.3% w/v in trichlorobenzene and passed through a bank of three 10 A Styragel HT columns at 140°C. The change in the refractive index of the eluant was recorded with the time.

RESULTS AND DISCUSSION

The variation in tensile properties with respect to carpet-fiber loading is shown in Figures 1–4. Increasing the fiber loading increased the tensile modulus from around 100 MPa for virgin LDPE to 350 MPa for samples containing 50% w/w carpet fiber. However, this increase in stiffness was achieved at the expense of the specific tensile energy at break, with these values decreasing from 0.038 J kg^{-1} m^{-3} for the matrix to 0.003 J kg^{-1} m^{-3} for samples containing 50% carpet. A reduction in the tensile strength, the elongation, and, hence, the specific tensile energy at break of the samples is not surprising considering that 75% of the mass of the carpet employed in the trial was either nylon 6,6 or jute, which, itself, is composed of mainly cellulose and hemicellulose.[13]

Comparing the highly polar chemical structure of these fibers to that of polyethylene (PE) (Fig. 5) would suggest that there is limited chemical compatibility between them. As such, the carpet fibers merely act as a filler with little stress transfer possible across the fiber–matrix interface owing to low levels of interfacial interactions between the carpet fibers and the PE matrix.

Commercially available PE copolymers containing ester, anhydride, and/or epoxy substituents were chosen as compatibilizers, since these substituents are known to be reactive with nylon chain ends and amide groups via exchange reactions and with the hydroxyl groups associated with cellulose-type structures (Fig. 6).[8,14–16] Initially, the compatibilizers were screened for their propensity to improve the performance of carpet fiber-reinforced LDPE using a 5% w/w loading of the compatibilizers (Table III). The impact of the compatibilizers on the performance of the parent matrix was also investigated, with the results from this study presented in Table IV.

Composite materials that incorporated PE-*g*-maleic anhydride (Mah) (**1**) were shown to provide a marked improvement in mechanical performance compared with the other compatibilizers trialed. PE-*g*-MAh (**2**) gave much poorer results in performance even though it had a higher level of grafted MAh groups than had PE–MAh (**1**) (3.0% compared with 0.85% w/w). Composite materials containing the remaining types of compatibilizers (PE-*co*-GMA, PE-*co*-MA-*co*-GMA, and PE-*co*-EA-*co*-MAh) provided improvements in tensile strength and strain values over unmodified and PE-*g*-MAh (**2**), albeit less than PE-*g*-MAh (**1**), their performance also not correlating with the number of reactive functional groups associated with their chemical structure.

HTGPC analysis of the compatibilizers (Fig. 7) suggested that PE-*g*-MAh (**2**) had a significantly lower molecular weight distribution compared with the other copolymers, as indicated by its
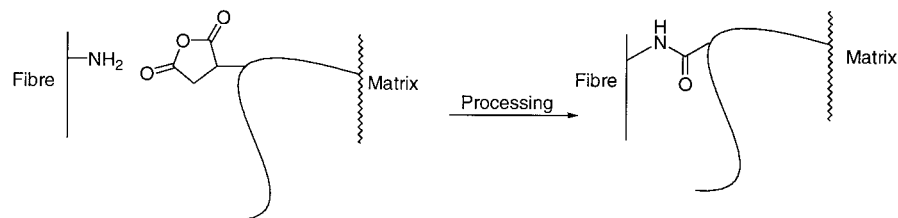

Figure 6   Schematic of one possible graft reaction between the nylon amine chain end group and Mah-modified PE.

Table III   Variation in Tensile Performance of 30% Carpet Fiber-reinforced LDPE Composites with the Addition of 5% w/w Compatibilizer

Sample	Tensile Modulus[a] (MPa)	Specific Tensile Energy[a] ($J kg^{-1} m^{-3} \times 10^{-3}$)	UTS[a] (MPa)	Strain[a] (mm/mm)
Untreated	203 (5)	8.8 (0.6)	10.5 (0.1)	0.42 (0.02)
PE-*co*-GMA	200 (9)	10.8 (0.6)	14.0 (0.1)	0.45 (0.03)
PE-*co*-MA-*co*-GMA	172 (5)	8.5 (0.5)	10.0 (0.1)	0.42 (0.01)
PE-*co*-EA-*co*-MAh	188 (7)	10.4 (0.7)	12.9 (0.1)	0.45 (0.05)
PE-*g*-MAh (**1**)	221 (7)	14.3 (0.6)	16.6 (0.1)	0.55 (0.05)
PE-*g*-MAh (**2**)	206 (6)	8.4 (0.6)	12.7 (0.1)	0.36 (0.03)

[a] Standard deviation is provided in parentheses.

longer retention time and, thus, smaller hydrodynamic volume. The remaining compatibilizers surveyed had broad molecular weight distributions similar in average hydrodynamic volume to PE-*g*-MAh (**1**), which, itself, was smaller than was the LDPE matrix used in the study. This reduced molecular weight distribution explains the poor performance of PE-MAh (**2**) as a compatibilizer, since it limits the extent of attractive van der Waals interactions and chain entanglements possible with the matrix once the compatibilizer has reacted with the fiber surface. Hence, the strength of the fiber–matrix linkage is reduced, which results in poorer mechanical performance of composite materials prepared with it compared with PE-*g*-MAh (**1**).

The compatibilizers PE-*co*-GMA, PE-*co*-MA-*co*-GMA, and PE-*co*-EA-*co*-MAh are highly functionalized, containing between 8 and 33% reactive monomer within their structures and, at a 5% w/w loading, do not appear to have a detrimental impact on the tensile properties of the virgin LDPE matrix (Table IV). However, in a carpet fiber-reinforced LDPE composite material, such

high levels of functionalization on the compatibilizer backbone may result in it interacting strongly with the carpet fibers, but not having sufficient regions compatible with the LDPE matrix to form a strong interfacial linkage. Such behavior may explain the reduced mechanical performance of these compatibilizers compared with a carpet fiber–LDPE composite utilizing PE-*g*-MAh (**1**), since less efficient stress transfer is possible between the reinforcing fibers and the matrix in these instances. Thus, lightly functionalized PE-*g*-MAh (**1**) provided the best balance of molecular weight distribution and functionality in this study.

Figures 8–11 provide information pertaining to the effects of fiber concentration and PE-*g*-MAh (**1**) loading on the performance of the composite material. In general, the addition of even small quantities (1%) of the MAh compatibilizer increased the performance of the carpet fiber-reinforced composite material in terms of ultimate tensile strength, elongation at maximum load, and specific tensile energy, although the tensile

Table IV   Variation in the Tensile Performance of LDPE with 5% w/w Compatibilizer

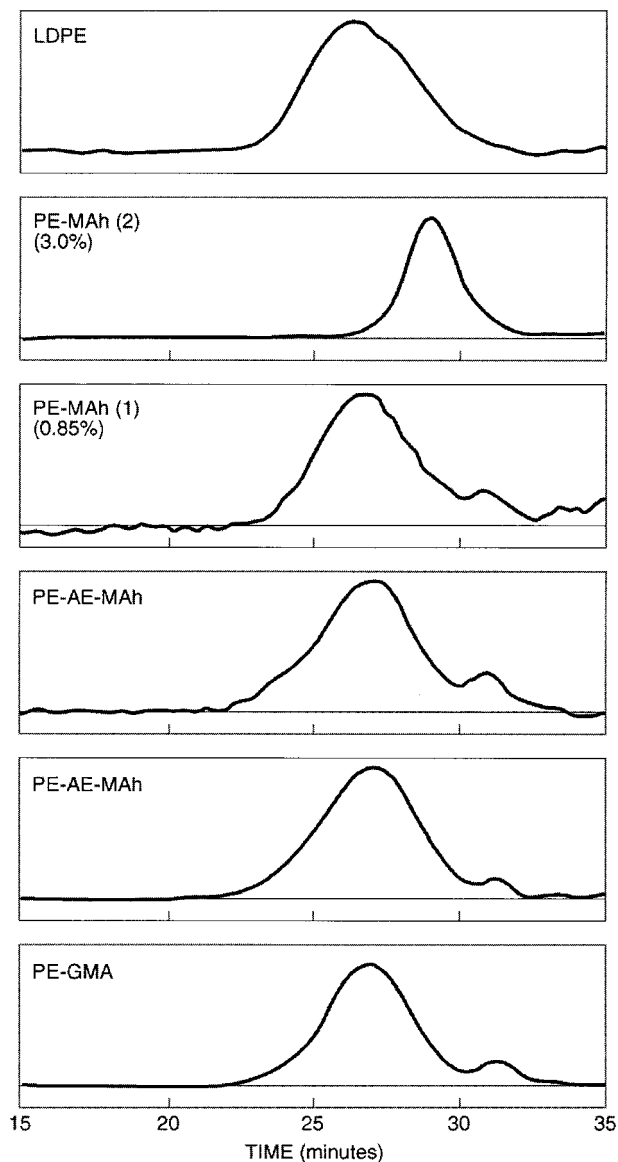| Sample	Tensile Modulus[a] (MPa)	Specific Tensile Energy[a] ($J kg^{-1} m^{-3} \times 10^{-3}$)	UTS[a] (MPa)	Strain[a] (mm/mm)
Untreated	87.7 (1.9)	38.2 (0.5)	11.5 (0.1)	1.75 (0.03)
PE-*co*-GMA	83.5 (3.4)	39.9 (2.0)	11.4 (0.1)	1.93 (0.02)
PE-*co*-MA-*co*-GMA	80.0 (0.8)	38.5 (0.3)	11.2 (0.1)	1.72 (0.02)
PE-*co*-EA-*co*-MAh	82.2 (3.6)	38.3 (1.7)	11.4 (0.1)	1.71 (0.03)
PE-*g*-MAh-1	87.7 (3.5)	41.5 (1.3)	11.7 (0.1)	1.94 (0.01)
PE-*g*-MAh-2	83.8 (1.5)	36.5 (0.6)	11.1 (0.1)	1.64 (0.02)

[a] Standard deviation is provided in parentheses.

Figure 7 HTGPC chromatograms of LDPE and the compatibilizers used to form carpet fiber–LDPE composite materials.
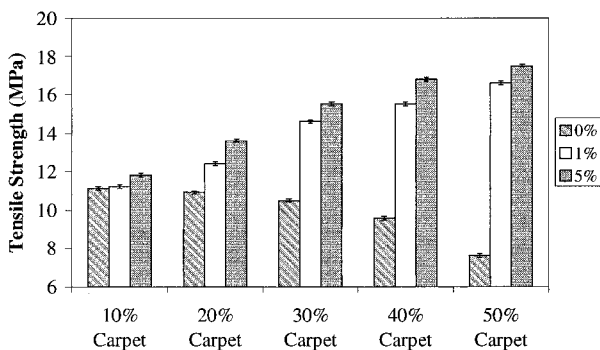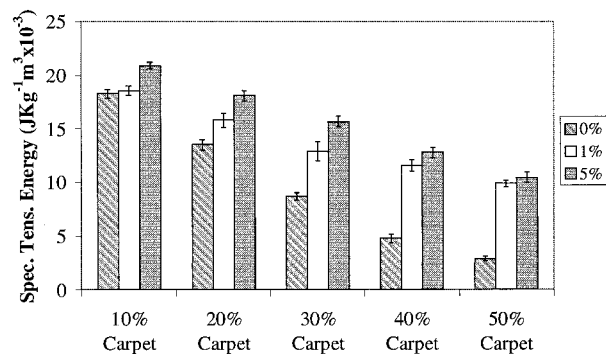

Figure 9 Specific tensile energy of carpet fiber–LDPE composite materials modified with various concentrations of PE-*g*-MAh (**1**).

modulus value was not significantly affected by the addition of the compatibilizer.

In the case of ultimate tensile strength, the addition of the PE-*g*-MAh (**1**) compatibilizer reversed the trend of decreasing performance with increased fiber loading; as previously described in the absence of the compatibilizer, composite materials exceeded the ultimate tensile strength of LDPE (12 MPa). Elongation-at-break values were also improved with samples containing 5% compatibilizer and a 50% fiber loading, providing strain values comparable to those of noncompatibilized samples with a 30% fiber loading. These improvements in performance may be explained in terms of a stronger interfacial interaction between the matrix and the fibers, resulting in better stress transfer between them.[10–13] Indeed, increasing the level of the compatibilizer improved the performance of the material regardless of the fiber concentration, as might be expected.
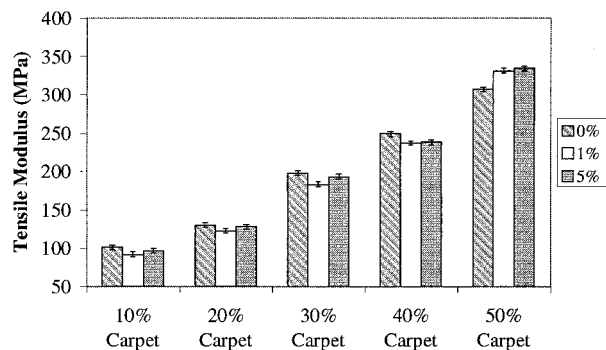

Figure 8 Ultimate tensile strength of carpet fiber–LDPE composite materials modified with various concentrations of PE-*g*-MAh (**1**).

Figure 10 Tensile modulus of carpet Fiber–LDPE composite materials modified with various concentrations of PE-*g*-MAh (**1**).

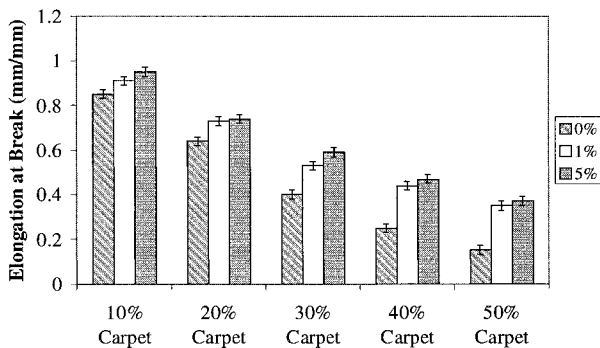**Figure 11**   Elongation at break of carpet fiber–LDPE composite materials modified with various concentrations of PE-*g*-MAh (**1**).

Figure 13   Fracture surface of noncompatibilized carpet fiber–LDPE composite material (1500×).

SEM analysis of the specimens supported the hypothesis of increased fiber–matrix interaction with the addition of PE-*g*-MAh (**1**). Figure 12 presents the fracture surface of a noncompatibilized 30% carpet fiber–LDPE composite specimen following tensile testing. Closer inspection of a carpet-fiber bundle (Fig. 13) shows that fibers are pulled away from the matrix, indicating little fiber–matrix interaction. Conversely, SEM analysis of samples containing the MAh compatibilizer (Fig. 14) show that the LDPE matrix interacted strongly with the fiber surface. Increased magnification of the sample (Fig. 15) shows that the LDPE matrix yielded under the tensile strain but remained attached to the fiber surface through fracture. The SEM results confirm that improved interfacial compatibility and interactions resulted from the addition of the compatibilizer.
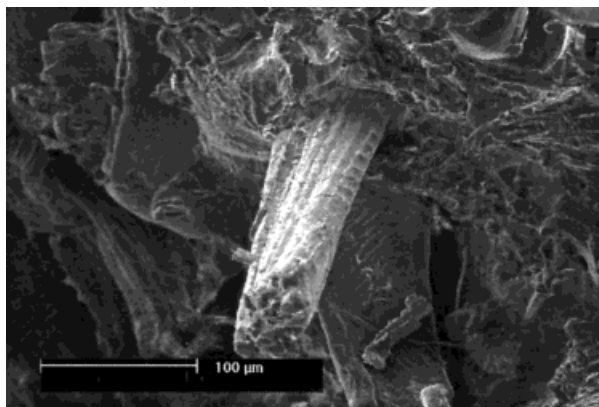
CONCLUSIONS

A new process for the preparation of carpet fiber-reinforced composite materials was reported using conventional plastics processing equipment. The use of as little as 1% w/w of an MAh-modified PE copolymer was shown to improve the tensile properties of such materials as a result of increased interfacial compatibility and enhanced interfacial adhesion.

Figure 12   Fracture surface of noncompatibilized carpet fiber–LDPE composite material (250×).
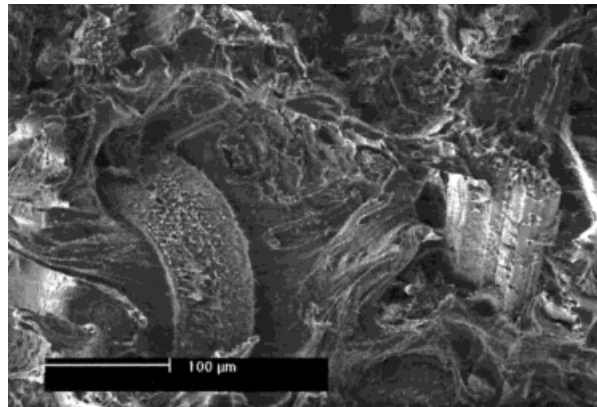

Figure 14   Fracture surface of carpet fiber–LDPE composite material with PE-*g*-MAh (**1**) (200×).
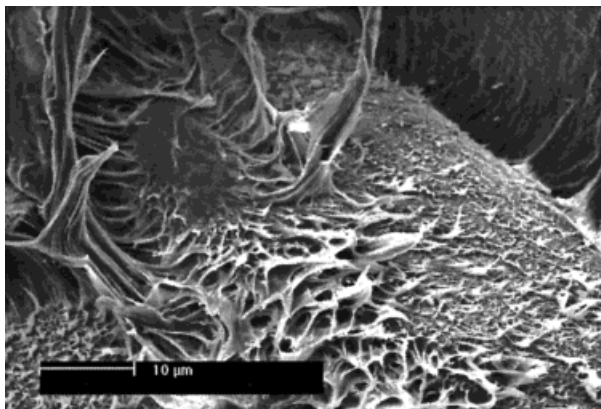
Figure 15 Fracture surface of carpet fiber–LDPE composite material with PE-*g*-MAh (**1**) (1500×).

REFERENCES

1. Kumar, S.; Zang, Y.; Muzzy, J. D. Polym-Plast Technol Eng 1999, 38, 485–498.
2. Kotlair, A. M.; Fountain, D. P. U.S. Patent 5 626 939 (1997).
3. Funger, B.; Gruber, H. U.S. Patent 5 662 994 (1997).
4. Brawn, M.; Levy, A. B.; Sifniades, S. Polym-Plast Technol Eng 1999, 38, 471–484.
5. Gallagher, F.G. U.S. Patent 5 710 315 (1996).
6. Gallagher, F.G. U.S. Patent 5 532 404 (1998).
7. Wang, Y.; Frost, J.D. In Symposium Proceedings—Recovery and Effective Reuse of Discarded Materials and Byproducts for Construction of Highway Facilities, October 1993.
8. Datta, S.; Lohse, D. J. Polymeric Compatibilizers—Uses and Benefits in Polymer Blends; Hanser: New York, 1996.
9. Shultz, J.; Lavielle, L.; Martin, C. J Adhes 1987, 23, 45–60.
10. Wu, S. Polymer Interface and Adhesion; Marcel Dekker: New York, 1982.
11. Lesko, J. J.; Jayaramen, K.; Reifsnider, K. L. Key Eng Mater 1996, 116–117, 61–86.
12. Mander, E.; Grundke, K.; Jacobasch, H.-J.; Wachinger, G. Composites 1994, 25, 739–744.
13. Encyclopedia of Polymer Science and Engineering; Kroschwitz, J.I., Ed.; Wiley: New York, 1985; Vol. 8.
14. The Chemistry and Technology of Cellulosic Co-Polymers; Hebeish, A.; Guthrie, J. T., Eds.; Springer-Verlag: New York, 1981.
15. Bledzki, A. K.; Reihmane, S.; Gassan, J. Polym-Plast Technol Eng 1998, 37, 451–468.
16. March, J. Advanced Organic Chemistry, 3rd ed.; Wiley-Interscience: New York, 1985.