

Impact Behavior of Unidirectional Polyethylene–Glass Fibers: Resol/Vinyl Acetate-2-Ethylhexylacrylate Interpenetrating Polymer Network Systems

Chandan Datta, Diya Basu, Amaranth Banerjee

Department of Polymer Science and Technology, 92 A.P.C. Road, University of Calcutta, Calcutta 700 009, India

Received 24 July 2002; 13 April 2003

ABSTRACT: Resol was solution blended with vinyl acetate-2-ethylhexylacrylate (VAc–EHA) resin in an aqueous medium at a 90-10 w/w ratio with hexamethoxymethylmelamine (HMMM) as crosslinker. Here we aimed to study the impact behavior of unidirectional laminates cast from (Resol/VAc–EHA/HMMM)/glass fiber (GF), (Resol/VAc–EHA/HMMM)/polyethylene fiber (PEF), and (Resol/VAc–EHA/HMMM)/GF/PEF (hybrid) and to study the role of PEF ply/plies in hybrid laminates toward the impact behavior, as dependent on the relative position of the ply/plies. A

brittle failure mode was observed in the GF-reinforced laminates, which tended to the ductile failure mode, with the incorporation of PEF ply/plies. Again, the impact fracture mode of GF was minimized by the placement of PEF ply/plies at the impacted side of the hybrid laminates. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 339–342, 2004

Key words: Interpenetrating networks (IPN); hybrid laminates; impact strength; Resol

INTRODUCTION

Hybrid composites possess some unique features that can be used to meet different design requirements with respect to strength, stiffness, and impact resistance. A key parameter in hybrid composite structures is the arrangement of fibers within the hybrid, as demonstrated by studies on hybrid systems based on carbon fibers, glass fibers, (GFs) or aramid fibers.^{1–5} Polyethylene fiber (PEF), a tough reinforcing fiber, is currently produced by the solution (gel) spinning of ultrahigh-molecular-weight polyethylene (PE) and possesses good mechanical properties in terms of high specific strength and stiffness.⁶ Moreover, these PEFs possess a relatively high work-to-break ratio (i.e., good impact properties) compared to carbon fibers, GFs, and aramid fibers. Because of these properties, PEFs have a high potential for various applications, notably for those that require good impact properties.^{7–12}

Phenolic resin is well known for its good heat- and fire-resistance properties, but the impact properties of this resin are not at all up to the mark. Hence, we took a full interpenetrating polymer network system of Resol and vinyl acetate-2-ethylhexylacrylate (VAc–EHA). However, this resin could not compete with

metals with respect to properties at lower temperatures. High-performance PEF used as reinforcement offset this demerit, which made this composite comparable to metal, even with respect to mechanical properties.

This work was undertaken with the following two objectives: to study the impact behavior of unidirectional laminates cast from [Resol/VAc–EHA/hexamethoxymethylmelamine (HMMM)]/GF, (Resol/VAc–EHA/HMMM)/PEF, and (Resol/VAc–EHA/HMMM)-GF/PEF (hybrid) and to study the role of PEF ply/plies in hybrid laminates toward the impact behavior, dependent on the relative position of the ply/plies.

MATERIALS

Resol was prepared by a method cited in the literature.^{13,14} The hardener of resole was *p*-toluolsulfuric acid 0982 H from Bakelite AG. The VAc–EHA copolymer was obtained from Macromoles (India). PEF (spectra 900, 1200 den) was supplied by Allied-Signal Corp. (Petersburg, FL). GF (433 BF-225) was supplied by Owens Corning Fiberglass Corp. (OH).

The surface of GF had already been treated with a standard treatment and was used directly to make composites.

CHROMIC ACID TREATMENT OF HIGH-PERFORMANCE PE FIBERS

For the use of treated fiber in making composites, the fibers were immersed in chromic acid at room tem-

Correspondence to: A. Banerjee

Contract grant sponsor: Council of Scientific & Industrial Research [to C.D. (Simer Research Fellow) and D.B.].

perature for 15 min,¹⁵ after which they were immediately rinsed in distilled water followed by a washing in running water for 2 h. The fibers were then further immersed in distilled water and dried in an air oven at 40°C for 5 h. For these treatments, the standard composition of chromic acid adopted¹⁶⁻²⁰ was 7 parts by weight $K_2Cr_2O_7$, 150 parts by weight concentrated H_2SO_4 , and 7 parts by weight H_2O .

EXPERIMENTAL

The individual polymers (emulsions) were first separately diluted with distilled water to maintain a solid content of 50 wt % for convenience, under well-stirred conditions. Then, a weighed amount of Resol was taken in a three-necked round-bottomed flask. The VAc-EHA copolymer was then accurately weighed into the flask, and the contents were stirred to give a homogeneous mixture in the desired blend ratio of the components. PTSA-²¹ was thoroughly mixed with 7 wt % (based on Resol) for 20 min. Then, HMMM was added at 20 wt % of VAc-EHA copolymer.^{22,23} The prepregs of GF and PEF were prepared previously. When the formation of the bubbles ceased, the viscous mass was poured into a glass mold prepared by the clipping together of two glass plates separated by a Teflon gasket in between, the thickness of which controlled the thickness of the sample sheet formed. It was then initially kept at room temperature for about 24 h and then heated at 80°C for 4 h. Thus, the samples were produced.

Impact properties

Izod impact strength was determined with ASTM D 256. The unnotched samples for measurements were cut to 70 × 10 × 3 mm dimensions. In all cases, 12 specimens were tested and average values were reported.

RESULTS AND DISCUSSION

Figure 1 shows the schematic arrangement of the hybrid laminates. Figures 2 and 3 show the variation of impact strength with the volume fraction of the fibers (V_f). As the number of plies was increased from one to four, the impact strength gradually increased. The impact strength of polyethylene-fiber-reinforced laminates (PEFRCs) was 2–2.5 times that of glass-fiber-reinforced laminates (GFRCs) at the same V_f indicating the superior energy-absorbing capacity of ductile PEF. A nonlinear variation of the impact strength with V_f was observed at higher V_f values

The impact strength in the curve for GFRC rose less steeply compared to the PEFRC curve with increasing V_f . Cracks were generated at the impacted surface (compression side), which contributed to the impact

Nomenclature	Arrangement of different plies	
	————— PEF	- - - - - GF
GP(31)	U ----- ----- ----- ----- ----- L	
GP(22)	U ----- ----- ----- ----- L	
GP(13)	U ----- ----- ----- ----- L	
I(31)	U ----- ----- ----- ----- ----- L	
I(22)	U ----- ----- ----- ----- L	
I(13)	U ----- ----- ----- ----- L	

a

Load direction	Sample designation
	GP(31)/U
	GP(31)/L

b

Figure 1 Schematic representation of hybrid laminates: (a) nomenclature and geometry of hybrid laminates and (b) load direction with sample designation.

failure by crack propagation. For GRFC, the initiated crack was propagated more easily from the impacted surface to the outermost surface (tension surface) due to the brittle characteristics of GF (i.e., low fraction propagating energy). As a result, the curve flattened

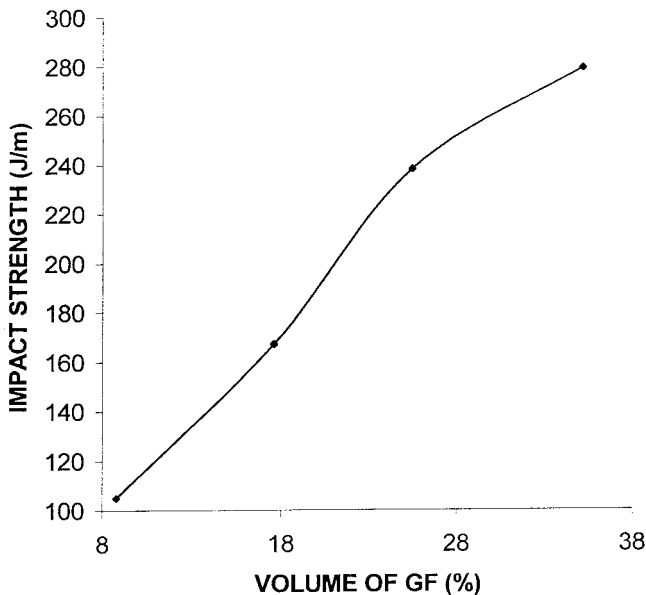


Figure 2 Variation of impact strength with volume of GF (%).

as the V_f increased. However, for PEFRC, the PEF absorbed the energy (which was transferred from the striking edge) and also resisted crack propagation. This was more pronounced as the PEF plies were increased from one to four plies. Due to these reasons, the curve became steeper as the V_f increased.

An interesting feature of this study was that the impact behavior changed remarkably when the position of the PEF and GF ply/plies were altered in the hybrid laminates. In Figure 4, the impact strength is

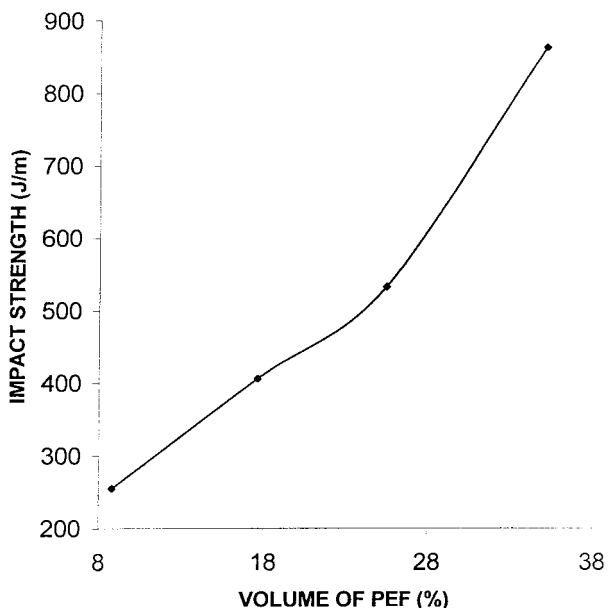


Figure 3 Variation of impact strength with volume of PEF (%).

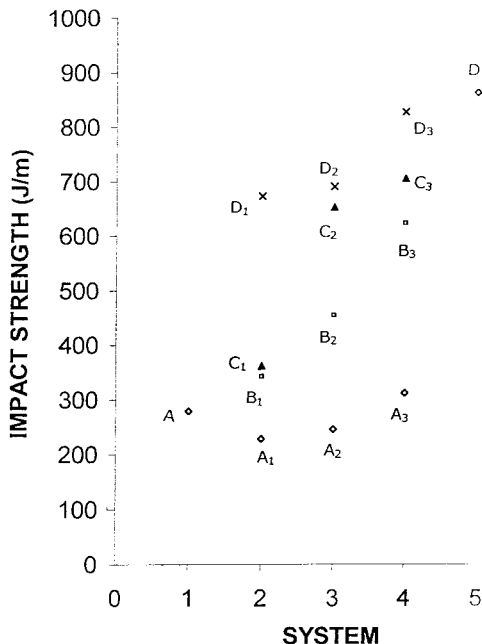


Figure 4 Plot of the impact strength versus systems 1–5. System 1: A, G₄; System 2: A₁, I(31)/L; B₁, GP(31)/L; C₁, GP(31)/U; D₁, I(31)/U; System 3: A₂, I(22)/L; B₂, GP(22)/L; C₂, GP(22)/U; D₂, I(22)/U; System 4: A₃, I(13)/L; B₃, GP(13)/L; C₃, GP(13)/U; D₃, I(13)/U; System 5: D, P₄.

plotted against systems 1–5. All of the systems in this figure contained approximately the same total V_f . When three GF plies and one PEF ply were mixed together (system 2), the impact strength increased from 230 ± 10 to 675 ± 12 J/m [I(31)/U]; the samples GP(31)/L and GP(31)/U showed values of 345 ± 15 and 385 ± 10 J/m, respectively. The same feature was reflected in systems 3 and 4.

From these studies, we concluded that when the striking edge hit the side containing the PEF ply/plies of the PEF-rich side of the hybrid composites, the impact strength was always higher. If the case was just reversed, the impact strength showed a lower value compared to the former one. When impact shock was applied to the beam, the crack was generated on the compression side, which was transferred to the tension side. This impact behavior may have been due to the superior shock-absorbing capability and efficient crack-resisting characteristics of the PEF ply/plies compared to the GF ply/plies.

A brittle failure mode was observed in the GFRC, which tended to the ductile failure mode by the incorporation of the PEF ply/plies. Almost no PEF fracture occurred in PEFRC and hybrid specimens. Figure 5 shows the optical micrographs of the fracture surfaces of the hybrid laminates. When the PEF ply was at the compression side, the fibers did not fracture, but extension and buckling took place [Fig. 5(b)].

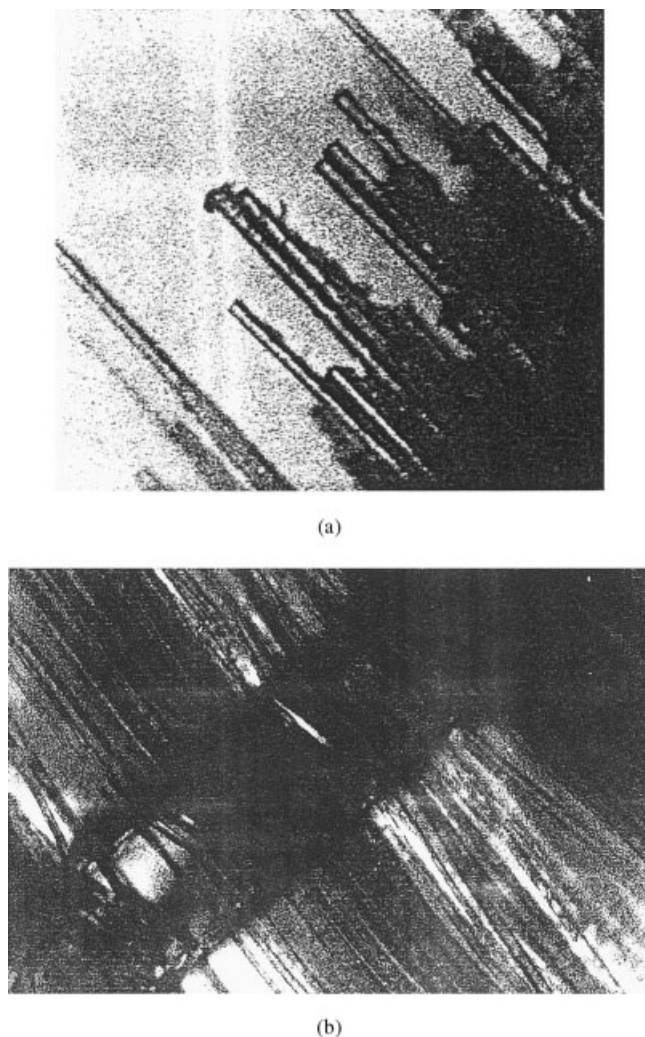


Figure 5 Optical micrograph of the fracture surface: (a) GF and (b) PEF at the impacted surface.

CONCLUSIONS

From this investigation, we drew the following conclusions:

1. The impact performance of GFRC was significantly improved by hybridization with PEF.

2. The impact fracture mode of GF was minimized by the placement of PEF ply/plies at the impacted side of the hybrid laminates.
3. Previously, work has been done with PEF with matrix systems such as epoxy resin and thermoplastics acrylic resin. However the introduction of phenolic resin as a matrix in PEFRCs was novel, and its application field will also, be versatile. The properties and performance of this composite are superior to the other existing composites of this nature.

References

1. Wagner, H. D.; Roman, I.; Marom, G. *J Mater Sci* 1982, 17, 1359.
2. Mallick, P. K.; Broutman, L. J. *J Testing Evaluation*, 1977, 5, 190.
3. Dorey, G.; Sidney, G. R.; Hutchings, J. *Composites*, 1978, 9, 25.
4. Morom, G.; Fischer, S.; Tuler, F. R.; Wagner, H. D. *J Mater Sci*, 1978, 13, 1419.
5. Peijs, A. A. J.; Venderbosch, R. W. *J Mater Sci Lett* 1991, 10, 122.
6. Iemstra, P. J.; Kirschbaum, R.; Ohta, T.; Yasuda, H. *Developments in Oriented Polymer*; Elsevier, London, 1987; Vol. 2, p 39.
7. Adams, D. F.; Zimmerman, R. S.; Chang, H. W. *SAMPE J* 1985, 21, 44.
8. Chang, H. W.; Lin, L. C.; Bhatnagar, A. *Proceedings of the 31st International SAMPE Symposium*, 1986; p 859.
9. Zimmerman, R. S.; Adams, D. F.; *Proceedings of the 32nd International SAMPE Symposium*, 1987; p 1461.
10. Peijs, A. A. J. M.; Catsman, P.; Govaert, L. E.; Lemstra, P. J. *Composites* 1990, 21, 513.
11. Peijs, A. A. J. M.; Venderbosch, R. W.; Lemstra, P. J. *Composites* 1990, 21, 522.
12. Peijs, A. A. J. M.; De Kok, J. M. M. *Composites* 1993, 24, 19.
13. Steiner, P. T. *J Appl Polym Sci* 1975, 7, 19.
14. Fyfe, C. A. *J Appl Polym Sci* 1983, 28, 2611.
15. Peijs, A. A. J. M.; Catsman, P.; Govaert, L. E.; Lemstra, P. J. *Composites* 1990, 21, 513.
16. Ladizesky, N. H.; Ward, I. M. *J Mater Sci* 1983, 18, 533.
17. Ladizesky, N. H.; Ward, I. M. *J Mater Sci* 1989, 24, 3763.
18. Silverstein, M. S.; Breuer, O. *J Mater Sci* 1993, 28, 4153.
19. Silverstein, M. S.; Breuer, O. *J Mater Sci* 1993, 28, 4718.
20. Andreopoulos, A. G.; Liolios, K.; Patrikis, A. *J Mater Sci* 1993, 28, 5002.
21. Wolfrum, J.; Ehrenstein, G. W. *J Appl Polym Sci* 1995, 74, 3173.
22. Saha, N.; Banerjee, A. N. *J Appl Polym Sci* 1996, 62, 1193.
23. Mukhopadhyay, S.; Basu, D.; Banerjee, A. N.; Mitra, B. C. *J Appl Polym Sci* 1997, 63, 985.