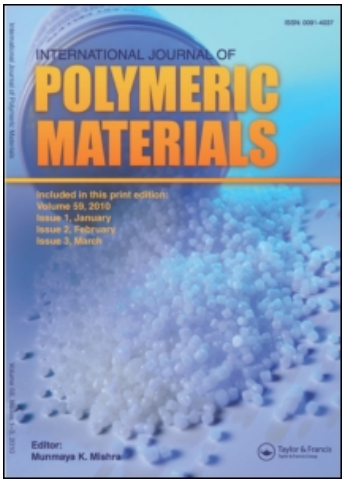

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

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To cite this Article Murty, Vedula M.(1983) 'Short Glass Fiber-Natural Rubber Composites', International Journal of Polymeric Materials, 10: 2, 149 — 158
To link to this Article: DOI: 10.1080/00914038308080266
URL: http://dx.doi.org/10.1080/00914038308080266

Intern. J. Polymeric Mater., 1983, Vol. 10, pp. 149–158
0091–4037/83/1002–0149 $18.50/0

Printed in the United Kingdom

Short Glass Fiber–Natural Rubber Composites

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(Received November 17, 1982)

Effect of fiber concentration on the physical properties such as tensile strength, tear strength and elongation at break, flexing resistance, heat buildup, compression set, permanent set, hardness, resilience and aging resistance of natural rubber filled with treated short glass fiber has been studied. The effect of carbon black on the physical properties and processing characteristics of the fiber–rubber composites has also been studied at different fiber concentrations. Scanning electron microscopy studies have been made in order to assess the failure criteria.

INTRODUCTION

Recently[1–2] we have reported the results of our studies on short jute fiber reinforced natural rubber composites and the effect of particulate fillers on short jute fiber–natural rubber composites. Glass fibers have found a wide range of uses in plastics and in certain rubber products such as tires. But as a short fiber, glass fiber is found to be inefficient because of high modulus which leads to breakage during mixing.[3–5] In the earlier studies[6] it is reported that short fibers act as reinforcing fillers only when they are added beyond a certain volume loading. It is also important that the fibers must have some critical aspect ratios for reinforcing the rubber matrix.[7] In this paper we have studied the effect of fiber concentration vis-a-vis fiber breakage on the physical properties of the rubber

composites. Reinforcing carbon black is added to the mixes in order to study its effect vis-a-vis the effect of short fibers. Scanning electron microscopy has been used to assess the dispersion and bonding of the fibers to the rubber matrix.

EXPERIMENTAL

Treated glass fiber as supplied by M/s. Fiberglass Pilkington Ltd. was used.

Mixing was done on a 150 mm × 330 mm open mixing mill with a nip gap of 0.25 mm. During mixing care was taken to ensure that in all mixes fiber orientation was the same in grain direction. Mixes were vulcanized at 150°C at their respective optimum cure times, as obtained from Monsanto Rheometer R-100. The method of preparation of the vulcanizates was the same as reported earlier.[8] Tensile testing and tear testing were done according to ASTM methods D412-52T and D624-54 respectively. Flexing was carried out at 70°C in a De–Mattia flexing machine according to ASTM D430-73. Heat buildup and permanent set measurements were made with a Goodrich flexometer according to ASTM D623-61, method A. Compression set measurements were made according to ASTM D395-61 method B. Shore A hardness was determined according to ASTM D676-57. Resilience was measured at 35°C using a Dunlop Tripsometer. Abrasion resistance was measured with a Croydon Akron Abrader and the samples were abraded for 500 cycles. The orientation of the fibers was maximum along the grain direction. All the tests except abrasion, resilience, and hardness were carried out both along and across the grain direction.

Green compounds (unvulcanized) were dissolved in benzene and the fibers were extracted. The measurement of fiber length distribution was done under an optical microscope. Green strength was measured using the method developed by Foldi.[9] The compounds were pressed at 120°C to remove tack and the strength at yield point was taken as the green strength. Mill shrinkage was measured according to ASTM D1917-62T.

The dumbell shaped specimens were aged at 100°C for 48 h and the physical properties of the aged samples were determined for studying the aging resistance.

The samples were made to fail under tensile and tear tests and the failed specimens were kept carefully in a desiccator without touching the surface. The fracture surfaces were sputter coated with gold within 72 h. of testing. The scanning electron microscopy (SEM) studies were made using a Philips 500 model.

RESULTS AND DISCUSSION

Table I gives the formulations and physical properties of the mixes A to G. In mixes B to G fiber concentration was varied. As in the case of other fibers, glass fibers also exhibit a minimum concentration of fibers[1,3,7] below which no reinforcement can be achieved using glass fibers. In the present case, the minimum concentration of glass fibers is 25 phr which is higher than that for jute fibers.[1] This high value of the minimum loading for glass fiber may be due to their severe breakage and poor bonding with rubber matrix. The anisotropy in tensile strength is observed only beyond 25 phr loading which again confirms that these fibers act as reinforcing fillers beyond 25 phr. Both the value of elongation at break and tear strength provide similar conclusions. Mixes (G_{10}, G_{25}, G_{75}) consisting of different fiber concentrations have been chosen for further study. Control stocks G_0, and F_0, both with and without black, have also been included in the studies.

TABLE I

Physical properties of mixes

Mix[a]		A[b]	B	C	D	E	F	G
Glass fiber		25	0	5	15	25	50	75
Optimum cure time, min		10.5	9.5	9.5	10.0	10.0	10.0	9.2
Tensile strength, MP_a	L	16.18	21.68	18.71	13.67	9.38	10.29	11.73
	T	—	21.45	18.12	13.15	9.32	7.89	7.00
Elongation	L	500	600	550	450	200	100	90
at break, %	T	—	600	520	450	350	380	150
Tear Strength, kN/m	L	39.16	32.22	36.88	41.10	33.87	31.14	41.76
	T	—	30.12	36.28	42.15	40.73	43.90	40.73

[a] Base recipe: NR 100, ZnO 5, Stearic acid 2, Resorcinol 5, Silica 5, Hexa 3.2, Sulfur 2, CBS 0.8.

[b] Mix A does not contain bonding agents, (silica, resorcinol, hexa).

TABLE II
Processing characteristics[a]

Glass fiber (Phr)	0	10	25	75	0	10	25	75
Carbon black (Phr) (N 330)	0	0	0	0	20	20	20	20
Mill shrinkage, %	57	35.4	21.8	12.7	48	26.8	12.6	18.7
Green strength, MPa	0	0	—	0.24	—	0.22	—	0.51

[a] Base recipe: NR 100, ZnO 5, Stearic acid 2, Silica 5, Resorcinol 5, Sulfur 2, CBS 2. Hexa 3.2.

Processing characteristics

Processing characteristics of the composites are reported in Table II. Mill shrinkage is decreased continuously with the increase in fiber loading in the absence of black. But in the presence of black up to a loading of 25 phr mill shrinkage decreases and at higher loading (mix F_{75}) there is slight increase in mill shrinkage due to high compound viscosity which results in fiber breakage. Green strength is improved by the addition of black as observed in the case of jute fiber reinforced composites.[2] The mill shrinkage is higher and green strength is lower for glass fiber reinforced composites as compared to jute fiber reinforced composites. Distribution of fiber length after mixing is shown in Figure 1. Fiber breakage is very severe in case glass fibers and fiber length decreases from 9 mm to 0.75 mm in the case of G_{10} and to 0.45 mm in the case of G_{25} and G_{75}. With the increase in fiber loading from 10 to 25 phr, due to the increase in compound viscosity, the breakage is increased and beyond that no more breakage of fiber occurred. The addition of carbon black further enhances the breakage of fibers and the fiber length in the case of black filled compounds is lower than that of corresponding mixes without black (Table II). It is interesting to note that fiber diameter has dropped down during mixing from 0.33 mm to 0.01 mm. It may be because the fiber was treated with a bonding agent and the coating increases the diameter. Subsequent mixing removes the coating and the diameter decreases. As discussed later, this may be the reason for not observing any difference in technical properties between treated and untreated fibers. The diameter of untreated fiber is 0.01 mm, which is very close to the diameter of the treated fiber after mixing. In the case of black filled compounds fiber diameter could not be measured accurately as the fibers are

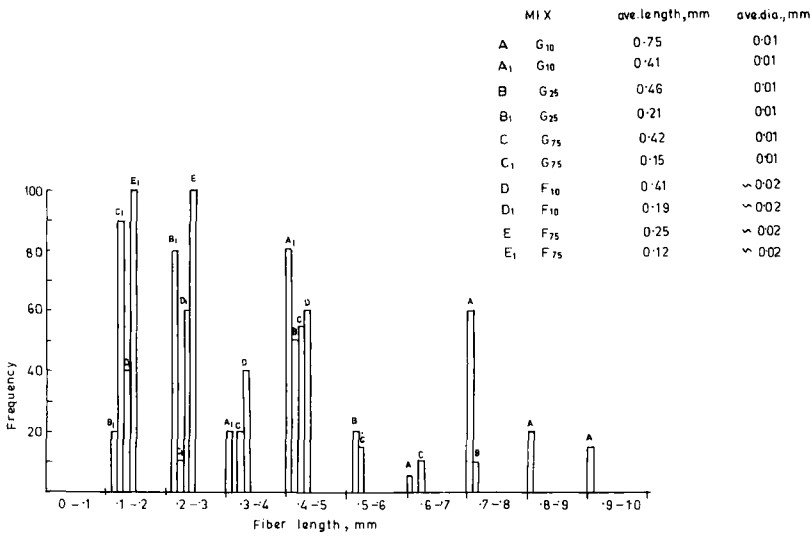

FIGURE 1 Histogram showing the fiber length distribution after mixing.

covered with a carbon black layer even after several hours of extraction. It is also interesting to note in all the cases two distributions of fiber length were observed. The distributions of fiber length are given in Figure 1.

Physical properties

Variation of tensile strength with volume loading of fiber in case black filled composites is similar to the observations made by us earlier[2] for carbon black filled jute fiber rubber composites. In the case of mix F_{75} because of severe fiber breakage there was no improvement in the tensile strength value measured in longitudinal direction. As the values of tensile strength in transverse direction does not depend much on the fiber length there is not much difference in these values.

Tear strength is improved by the addition of fiber but is mostly independent of fiber concentration and orientation. It was explained earlier[1–2] that the improvement in tear strength is due to the fact that fibers obstruct tear paths from proceeding straight.

TABLE III

Physical properties of the mixes

Mix[a]		G_0	G_{10}	G_{25}	G_{75}	F_0	F_{10}	F_{25}	F_{75}
Glass fiber (Phr)		0	10	25	75	0	10	25	75
Carbon black (N 330) (Phr)		0	0	0	0	20	20	20	20
Tensile Strength, MP_a	L	21.68	15.68	9.38	11.73	13.59	10.03	11.71	10.89
	T	21.45	15.22	9.32	7.00	15.16	9.27	8.00	8.79
Tear Strength, kN/m	L	32.22	38.64	33.67	41.76	30.40	35.36	35.87	29.31
	T	30.12	42.46	40.73	40.73	38.44	34.18	37.08	28.12
Elongation at break, %	L	600	380	200	90	450	300	200	70
	T	600	380	350	150	450	350	320	220
Hardness, Shore A	—	45	52	60	75	55	65	70	82
Flex resistance,	L	40.5	12.5	7.7	1.0	34.4	8.3	1.6	1.1
(kilo cycles)	T	40.5	22.0	7.7	2.4	34.4	8.3	3.6	1.8
Compression set, %	L	37.2	25.1	26.4	33.6	37.8	32.8	30.7	42.1
	T	35.9	28.3	26.4	38.2	37.7	34.2	30.6	42.6
Resilience, %	—	73.0	73.5	70.2	59.7	56.4	61.1	56.4	51.8
Heat buildup (ΔT°C)	L	—	10	—	29	15	19	22	39
	T	10	10	—	30	14	17	21	41
Permanent set, %	L	—	0.74	—	2.5	1.8	2.0	2.7	7.1
	T	0.78	0.45	—	1.2	1.7	1.4	0.9	3.6
Abrasion Loss, cc/500 rev.	—	0.54	0.89	1.41	1.41	0.48	0.79	1.04	1.32
Percent tensile strength retained after aging	L	5.5	12.4	30.4	43.1	17.9	33.1	38.1	54.3
Percent tear strength retained after aging	L	22.2	24.1	38.9	45.5	27.9	36.8	45.6	61.1
Percent elongation at break retained after aging	L	4.3	73.6	75.0	77.7	51.1	60.0	45.0	71.5

[a] Base recipe: NR 100, ZnO 5, Stearic acid 2, Silica 5, Resorcinol 5, Sulfur 2, CBS 2, Hexa 3.2.

Elongation at break drops down with the increase in fiber concentration and anisotropy of this property is observed when the fiber loading is beyond 25 phr both in the presence and absence of carbon black.

Flexing resistance drops down with the increase in fiber loading. Flex resistance is higher in the transverse direction since the stiffness becomes higher in the longitudinal direction. These observations are similar to our earlier observations with jute fiber-rubber composites.[1]

Heat buildup increases with the increase in fiber loading. The variation of heat buildup is similar to the variation of hardness with fiber loading. Heat buildup is lower in the case of composites with fibers oriented transversely as expected.

Permanent set increases with the increase in fiber loading in the

case of composites both with longitudinally oriented and transversely oriented fibers. However, permanent set is higher for the composites with fibers aligned longitudinally. The higher values for the composites aligned longitudinally may be due to the buckling that takes place invariably when the closely packed fibers are compressed in the direction of their alignment. When the samples were observed visually after the testing the wrinkles were observed on the surface of the samples with fibers aligned longitudinally, while the samples with transversely aligned fibers failed, forming deep cracks on the surface. However, compression set remained almost independent of fiber loading. Resilience as expected decreases with the increase in fiber loading. Carbon black further reduces the resilience values.

Abrasion resistance, as in the case of jute fiber, decreases with the addition of fibers, while the addition of carbon black increases the abrasion resistance. The higher abrasion loss in glass fiber reinforced composites is mainly due to the loss of fibers.[10] The increase in abrasion resistance in black filled mixes is due to the reinforcement offered by the carbon black.

Percent retention of tensile strength and tear strength values after aging at 100°C for 48 h increases with increase in fiber concentration. Comparison of the values after aging of the composites indicates that the effect of fiber on the aging retention property is dominant. In the case of fiber-rubber composites, fibers carry the load and rubber matrix transfers the load to the fibers. As fibers do not deteriorate by aging it is expected that fiber-rubber composites exhibit better aging resistance.

SEM studies

The scan areas and fracture surfaces are shown in Figure 2. Figure 3 gives the scanning electron micrograph of the tensile fracture of mix A containing no bonding agent. In this case debonding is obvious, with long stems of fiber protruding out of the rubber matrix. These observations are similar to our earlier observations with untreated glass fiber with bonding agent.[10] Figure 4 shows the photomicrograph of tensile fracture of mix G_{25} containing 25 phr fiber and bonding agent. There is an improvement in the bonding as the holes made by the detached fibers are not always circular indicating the

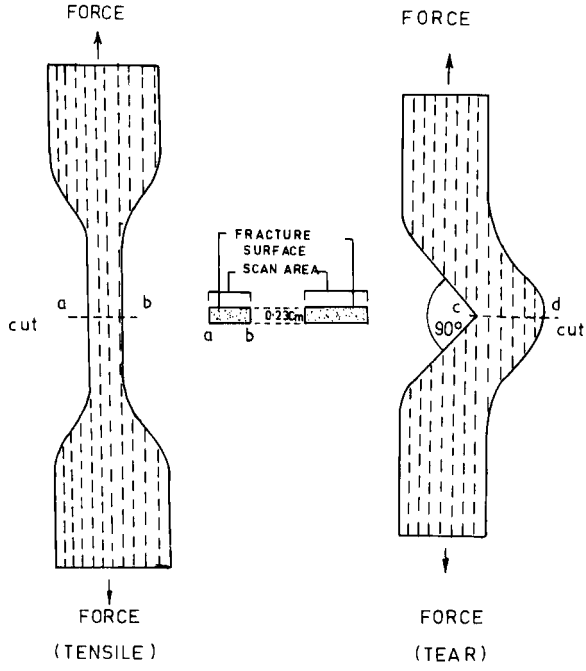

FIGURE 2 Fracture surfaces and scan areas.

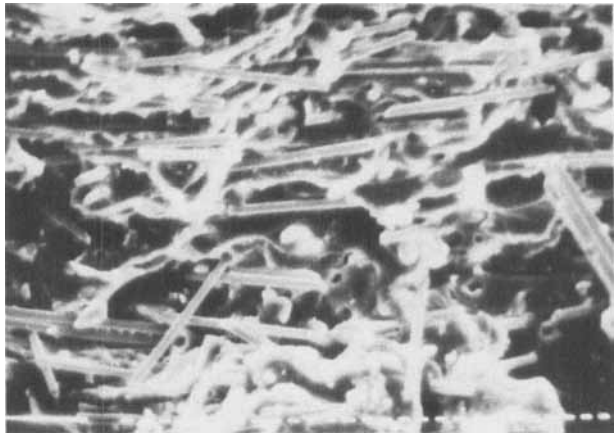

FIGURE 3 Photomicrograph of tensile fracture surface of mix A (200 ×).

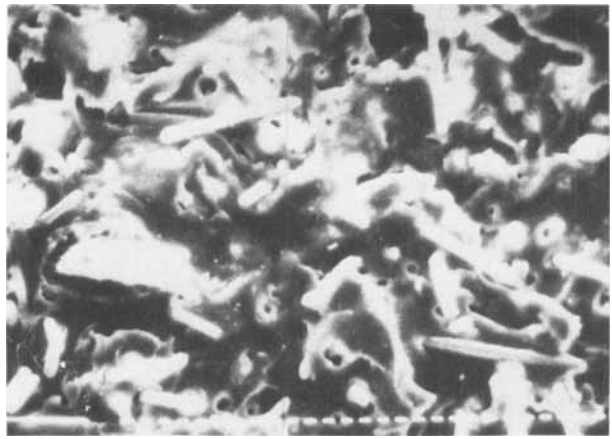

FIGURE 4 Photomicrograph of tensile fracture surface of mix G_{25} (200 ×).

removal of a certain amount of rubber along with the fiber. Some rubber compound can also be seen on the detached fibers (Figure 4). Tear fracture surface is shown in Figure 5. There are cracks on the surface. Figure 5 shows the crumbled matrix surface between the cracks. This observation is in contradiction to our earlier observation with jute fiber reinforced composites but is similar to that with untreated glass[10] fiber rubber composites. It is apparent

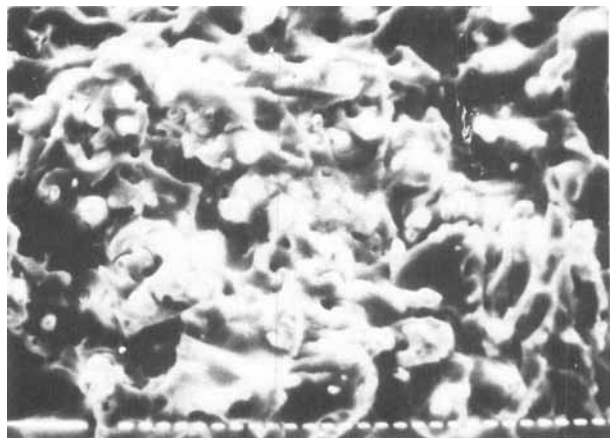

FIGURE 5 Photomicrograph of tear fracture surface of mix G_{25} (200 ×).

that bonding between glass fibers and natural rubber matrix is not complete as is the case with jute fiber and natural rubber. In the latter case tensile failure is mainly due to the fiber breakage.[1]

CONCLUSIONS

The following conclusions can be drawn from the above results and discussion

1) Short glass fibers show anisotropy in technical properties only when added beyond a loading of 25 phr.

2) Processing properties like mill shrinkage and green strength are improved by the addition of fibers and addition of carbon black further improves the properties. Fiber breakage is very severe in the present case. Addition of carbon black enhances the fiber breakage and consequently some technical properties are not improved by the addition of carbon black.

3) Fiber dispersion is good. There is some improvement in bonding between fibers and rubber matrix by the addition of bonding agents, (hydrated silica, resorcinol and hexa).

Acknowledgement

The author sincerely acknowledges the suggestions received from Dr. Sadhan K. De.

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