

Utilization of Bowstring Hemp Fiber as a Filler in Natural Rubber Compounds

E. Osabohien, S. H. O. Egboh

Department of Chemistry, Delta State University, Abraka, Nigeria

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ABSTRACT: The cure characteristics and physico-mechanical properties of natural rubber (standard Nigerian rubber) vulcanizates filled with the fiber of bowstring hemp (*Sansevieria liberica*) and carbon black were investigated. The results showed that the scorch and cure times decreased, whereas the maximum torques increased, with increasing filler loadings for both bowstring hemp fiber and carbon black filled vulcanizates. The tensile strength of both bowstring hemp fiber and carbon black filled vulcanizates increased to a maximum at a 40 phr filler concentration before decreasing. The elongation at break and rebound resilience decreased, whereas the modulus, spe-

cific gravity, abrasion resistance, and hardness increased, with increasing filler contents. The carbon black/natural rubber vulcanizates had higher tensile strength, which was about 1.5 times that of bowstring hemp fiber/natural rubber vulcanizates. This superiority in the tensile strength was probably due to the higher moisture content and larger particle size of the bowstring hemp fiber. However, the bowstring hemp fiber/natural rubber vulcanizates showed superior hardness. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 107: 210–214, 2008

Key words: compounding; fillers; reinforcement; rubber

INTRODUCTION

It has been well documented that the addition of sufficient quantities of filler particles such as carbon black (CB) to a polymer matrix greatly enhances the physical and mechanical properties of the resulting vulcanizates, which are used in a wide range of commercial and engineering applications. Generally, fillers are classified into two types: reinforcing (or active) and nonreinforcing (inert or diluent) fillers. A reinforcing filler is one that improves the modulus and failure properties (tensile strength, tear resistance, and abrasion resistance) of the final vulcanizates, whereas inert fillers add little or nothing to the tensile strength or tear or abrasion resistance but are frequently added to a polymer matrix to extend the volume of the stocks into which they are incorporated and hence reduce the overall production cost or aid processing.^{1–5}

CB has remained the most widely used reinforcing filler for most polymers; however, efforts to develop fillers from plant sources that can achieve a reinforcing potential similar to that of CB and thus substitute for it have been intensified in recent years.^{6–8} CB is produced from a nonrenewable resource, petroleum; it is expensive and imparts black coloration

to any polymeric material to which it is added. The utilization of plant-based fibers as fillers for polymers is based on the premise that they are renewable, are available in commercial quantities at relatively low cost, and can be added to polymers to form a biodegradable matrix.⁴ There is also the possibility of recycling the materials.⁹ Several studies have indicated the successful utilization of flax fibers,⁴ melon seed shells,⁹ cocoa pod husks,⁸ rubber seed shells,⁸ and rice husks¹¹ among others as fillers in natural rubber compounds. The industrial use of plant fibers as fillers has also increased in many polymer manufacturing sectors, such as household wares, packaging, toys, automobiles, building, and construction, because natural-fiber-reinforced polymer composites are lightweight, non-corrosive, nonabrasive, temperature-resistant, easy to recycle, and environmentally friendly and have high mechanical properties.⁴

Sansevieria liberica bowstring hemp belongs to the family *Agavaceae* and is one of the bowstring hemp species with concave, short petioled leaves containing over 2% fiber. It has long rhizomes with long fibrous roots and a rapid rate of growth. The leaves are short, with a low number of leaves per unit of area, and provide yields of about 1500 kg of fiber per hectare per annum. A number of species of bowstring hemp, such as *Sansevieria cylindrical*, *Sansevieria guineensis*, *Sansevieria trifasciata*, *Sansevieria roxburghiana*, *Sansevieria zeylanica*, *Sansevieria ehrenbergii*, and *Sansevieria longiflora*, are grown as ornamental

Correspondence to: E. Osabohien (osabohieneemma@yahoo.com).

plants. They are widely distributed throughout the tropics. Fibers for local use are obtained from the leaves of some species in various countries for making string, nets, coarse fabrics, and bows.¹⁰ The leaves and roots of *S. liberica* have been put to medical use in parts of Nigeria in the treatment of asthma, sexual weakness, hypertension, diarrhea, abdominal pains, hemorrhoids, and wounds of the foot. The plant is also prescribed as a remedy for monorrhagia, colic, and gonorrhoea. A paste of the herb is used to treat eczema, piles, and snake bites.¹²

This research work is aimed at investigating the effects of bowstring hemp fiber (BHF) on the cure characteristics and physicochemical properties of natural rubber vulcanizates. It is intended to develop a local filler that is lower cost, renewable, and available and that can be used as a substitute or supplement for CB fillers in natural rubber articles. Nigeria is endowed with rich agricultural resources, including different species of bowstring hemp. The end uses of these resources are of paramount importance for technological growth.

EXPERIMENTAL

Materials

The materials used in this study included BHF derived from the leaves of bowstring hemp collected from Abraka, Nigeria, and natural rubber [Standard Nigerian Rubber 10 (SNR10)] obtained from Foot Wear Accessories, Manufacturing and Distribution (formerly Bata Multinational Co., Benin City, Nigeria). Industrial-grade CB (N330) was obtained from Nigerian National Petroleum Company (Warri, Nigeria). Industrial-grade compounding ingredients and rubber test equipment were obtained from the Department of Polymer Technology, Auchu Polytechnic (Auchi, Nigeria), and Dunlop, Plc. (Ikeja, Lagos).

Methods

Preparation of the BHF filler

BHF was obtained from bowstring hemp leaves through the soaking of the leaves in clean water for about 1 week in a clean plastic bowl and the removal of the greenish part by washing with water. The resulting whitish fiber was air-dried for few days, pulverized with a corona grinding machine, and screened through a sieve with a mesh diameter of 212 nm.

Characterization of the natural rubber and BHF

The natural rubber used in this study (SNR10) was characterized in terms of its dirt, ash, and nitrogen contents, volatile matter, plasticity retention index,

and Mooney viscosity with standard techniques.^{3,13} The prepared BHF filler was also characterized in terms of the moisture content, loss on ignition, iodine adsorption number, pH of its aqueous slurry, density, and metallic composition particle diameter in comparison with those of N330 with standard methods.^{3,8,14,15}

Compounding and curing of the SNR10 mixes

The recipe used in the formulation of SNR10 is given later in Table III. The compounds were made with 0–70 phr filler loadings. Each of the formulations was masticated, and the mixing was carried out with a laboratory two-roll mill (160 × 320 mm²) kept below 80°C.

The SNR10 compounds were cured by compression molding with a steam press machine at a pressure of 150 kg/cm at 180°C for about 30 min. The cure characteristics of the vulcanizates were determined with a Monsanto MDR 2000 rheometer. The scorch and cure times and torques were determined from the resulting rheographs.

Determination of the physicochemical properties of the vulcanizates

The tensile properties (tensile strength, modulus, and elongation at break) of the vulcanizates were measured with a Monsanto Instron 4301 tensometer at a crosshead speed of 500 mm/min with dumbbell-shaped test specimens in accordance with ASTM D 638-99,¹⁶ whereas a Wallace Croydon R2A resilience tester was used to determine the rebound resilience. The specific gravity and hardness of the vulcanizates were measured with a Monsanto Densitron 2000 and a Wallace Croydon hardness tester, respectively. The abrasion resistance was measured in accordance with the procedure described in BS 903 part A9.¹⁷

RESULTS AND DISCUSSION

The results of this research work are shown in Tables I–VII. The properties of the BHF-filled natural rubber are compared with those of N330.

TABLE I
Characteristics of SNR10, SAR10, and SMR5

Parameter	SNR10	SAR10	SMR5
Dirt content (%)	0.01	0.02	0.05
Ash content (%)	0.25	0.32	0.50
Nitrogen content (%)	0.20	0.23	0.70
Volatile matter (%)	0.25	0.40	1.00
Plasticity retention index	71.00	67.00	—
Mooney viscosity at 100°C	76.00	70.00	60.00

TABLE II
Physicochemical Properties of BHF and N330

Parameter	BHF	N330
Moisture content at 125°C (%)	2.15	1.10
Loss on ignition at 1000°C (%)	92.50	94.50
Iodine adsorption number (mg/g)	54.30	80.10
Oil absorption (cm ³ /100 g)	165.20	133.50
pH of the aqueous slurry	7.10	6.90
Density (g/cm)	1.50	1.80
Sodium (%)	3.01	Trace
Potassium (%)	0.92	Trace
Calcium (%)	0.05	Trace
Magnesium (%)	0.06	Trace
Iron (%)	0.03	Trace
Silica (%)	0.20	0.30
Particle diameter range (nm)	20–212	30–35

Physicochemical properties of SNR10 and the fillers

Tables I and II summarize the physicochemical properties of SNR10 and the fillers. According to Table I, the values of the physical properties of SNR10 compare favorably with those of other standard natural rubbers such as Standard African Rubber 10 (SAR10)¹⁸ and Standard Malaysian Rubber 5 (SMR5).¹⁹ It shows higher purity because of its lower dirt, ash, nitrogen, and volatile matter contents. The high values of the plasticity retention index and the Mooney viscosity indicate a high resistance of the rubber to aging and flow, respectively.³ The physicochemical properties of the BHF and N330 fillers are shown in Table II. The loss on ignition at 1000°C is slightly higher for N330 than BHF, and this implies that N330 has more carbon or combustible materials than BHF. The results further reveal that N330 has a higher iodine adsorption number and consequently a larger surface area than BHF.³ The amount of carbon that a filler has, its surface area, shape, and surface structure, and the pH of its aqueous slurry are

TABLE III
Typical Recipe for the SNR10 Compound

Ingredient	phr
Natural rubber (SNR10)	100.0
Zinc oxide	4.0
Stearic acid	2.0
Filler ^a	0–70
Processing oil	2.0
CBS ^b	2.0
TMQ ^c	1.5
Sulfur	1.5

^a The fillers were BHF and N330; the filler loadings were 0, 10, 20, 30, 40, 50, 60, and 70 phr.

^b *N*-Cyclohexyl-2-benzothiazylsulfenamide.

^c 2,2,4-Trimethyl-1,2-dihydroquinoline.

TABLE IV
Cure Characteristics of SNR10 Filled with BHF

Filler (phr)	0	10	20	30	40	50	60	70
t_{s2} (s) ^a	34.2	27.0	24.6	23.8	23.4	23.3	23.2	23.0
t_{90} (s) ^b	41.4	40.6	40.1	38.6	38.6	37.6	37.4	37.4
Maximum torque (lb in.)	6.47	8.79	10.51	11.53	13.68	13.87	13.93	14.30

^a Scorch time.

^b Cure time.

the primary determinants of its reinforcing potential.^{11,20} The higher the carbon content is and the larger the surface area of the filler particles is, the higher the possibility is of interactions between the polymer matrix and the filler and the higher the reinforcement potential is.^{2,3,5,21} The N330 filler shows slight acidity according to the pH of its aqueous slurries (Table I). It has been shown that acidic fillers generally retard cure rates.^{2,9} BHF has a slightly higher moisture content at 125°C than N330. A high moisture content has been shown to lead to a poor dispersion of a filler in a polymer matrix and hence poor filler/polymer matrix adhesion.^{2,4,8,11}

Plant fibers contain cellulose and lignin in addition to other carbonaceous and mineral matter, and some have been reported to affect the properties of the polymer to which they are added. Several studies have revealed that lignin imparts low mechanical properties such as tensile strength, modulus, and hardness to natural rubber compounds.⁹

Cure characteristics of the SNR10 compounds

Tables IV and V show the cure characteristics of the SNR10 vulcanizates filled separately with BHF and N330 fillers at different filler loadings. The scorch and cure times for both the BHF- and N330-filled natural rubber systems decrease with increasing filler loading. Similar observations have been reported

TABLE V
Cure Characteristics of SNR10 Filled with N330

Filler (phr)	0	10	20	30	40	50	60	70
t_{s2} (s) ^a	34.2	32.4	28.2	26.4	25.8	24.8	24.2	24.0
t_{90} (s) ^b	41.4	49.2	46.2	43.8	43.2	42.2	41.2	40.2
Maximum torque (lb in.)	6.47	7.57	8.93	11.07	11.13	11.26	11.87	13.18

^a Scorch time.

^b Cure time.

TABLE VI
Physicomechanical Properties of SNR10 Filled with BHF

Filler (phr)	0	10	20	30	40	50	60	70
Tensile strength (MPa)	9.48	13.09	15.18	17.03	20.41	18.52	15.01	12.81
Modulus at 100% strain (MPa)	1.13	2.58	2.60	2.63	2.88	3.16	3.28	3.46
Elongation at break (%)	886.0	605.1	573.8	535.6	491.2	470.9	390.2	361.3
Specific gravity	1.006	1.008	1.016	1.037	1.048	1.062	1.077	1.084
Rebound resilience (%)	87.7	83.7	80.6	76.1	74.7	72.4	70.1	68.8
Hardness (International Rubber Hardness Degree)	44	56	68	72	73	74	76	80
Abrasion resistance index	40.1	40.1	41.0	41.4	42.1	43.0	43.3	44.1

in earlier studies.^{6,8,11,22} Ishak and Bakar¹¹ attributed the difference in the cure characteristics of vulcanizates to the fact that each filler possesses different properties, such as the surface area, surface reactivity, particle size, and metal oxide content. Both BHF- and CB-filled SNR10 compounds experience cure enhancement with increasing filler content. BHF-filled SNR10 shows a faster cure rate because the BHF filler has higher levels of metal and moisture and a lower surface area, that is, a larger particle size (Table II), which could affect the cure rates.¹¹ The maximum torque (T_{\max}) increases regularly with increasing filler content. The increase in T_{\max} implies an increase in the number of crosslinks formed with increasing filler content. These crosslinks restrict the molecular mobility of the polymer chains. BHF-filled vulcanizates have slightly higher T_{\max} values than CB-filled vulcanizates and hence a higher restriction of the molecular mobility of polymer chains because of good filler/rubber matrix adhesion as a result of the polymer (fiber)/polymer (rubber) interactions; this implies higher stiffness (modulus) and higher hardness for the BHF-filled vulcanizates (Tables VI and VII).^{23,24}

Physicomechanical properties of SNR10

Tables VI and VII summarize the physicomechanical properties of BHF- and N330-filled vulcanizates. The tensile strength of both the BHF- and N330-filled vulcanizates increases to a maximum at 40 phr and

decreases thereafter. However, the BHF-filled vulcanizates have lower tensile strength than the N330-filled vulcanizates, and this implies that BHF has a lower reinforcement efficiency that may be due to its higher moisture content and larger particle size (lower surface area; Table II).^{4,9,12}

The modulus at 100% strain increases with increasing filler content for both vulcanizates. The higher modulus observed in the BHF-filled vulcanizates versus the CB-filled vulcanizates could be due to some kind of improved fiber/rubber molecular interactions because both are macromolecules.^{24,25} The elongation at break for both the BHF- and N330-filled vulcanizates decreases with the filler content. The decrease in the elongation at break could be due to the attractive forces between the fillers and the polymer molecules leading to the formation of a crosslinked network that restricts the free mobility of the polymer chains and thus increases the resistance to stretching upon the application of strain.³ However, the BHF-filled vulcanizates have higher elongations at break than the N330-filled vulcanizates, probably because of the fiber/rubber matrix adherence resulting from the polymer (fiber)/polymer (rubber) interactions, which leads to an increase in the strain under stress.²⁵

The specific gravity, abrasion resistance, and hardness of both types of vulcanizates increases with an increasing filler loading, but the BHF-filled vulcanizates show slight superiority over the N330-filled vulcanizates, particularly with respect to the specific gravity and hardness properties, in applications

TABLE VII
Physicomechanical Properties of SNR10 Filled with N330

Filler (phr)	0	10	20	30	40	50	60	70
Tensile strength (MPa)	9.48	18.52	27.24	30.53	35.60	31.24	25.20	24.48
Modulus at 100% strain (MPa)	1.13	2.12	2.55	2.96	3.48	3.54	3.98	4.54
Elongation at break (%)	886.0	604.2	572.1	500.9	450.4	382.5	325.2	308.1
Specific gravity	1.006	1.015	1.046	1.069	1.089	1.104	1.119	1.139
Rebound resilience (%)	87.7	83.7	82.6	80.6	79.9	76.1	72.4	70.1
Hardness (IRHD)	44	45	50	56	58	61	65	67
Abrasion resistance index	40.1	41.2	42.1	43.8	44.6	45.5	45.6	45.8

requiring weight reduction. The BHF filler (Table II) has a lower density and hence a lower specific gravity, which implies lighter weight rubber articles for applications requiring weight reduction, which is advantageous. The results also show that the rebound resilience of both the BHF- and N330-filled vulcanizates decreases with an increasing filler loading. The slightly lower resilience of the BHF-filled vulcanizates implies slightly higher hysteresis and heat build-up, and hence they would be more difficult to process.³ The reinforcing potential of this BHF for natural rubber has been found to be superior to that of pineapple leaf fiber by a factor of about 1.5 times.²⁶

CONCLUSIONS

The results of this study show that the incorporation of BHF as a filler influences both the cure characteristics and physicomechanical properties of natural rubber vulcanizates but imparts inferior tensile strength in comparison with a commercial-grade CB filler (N330). This has been attributed to its higher level of moisture and larger particle size, both of which could lead to poor dispersion of the fiber in the polymer matrix and thus reduce the fiber/rubber matrix interfacial bonding and strength. Related studies have indicated that the reinforcing potential of a filler could be improved by carbonization and particle size reduction or by a chemical pretreatment of the filler.^{4,11,27} Therefore, if modern methods of wet and dry grinding techniques are employed, a finer particle size BHF filler could be obtained that could improve the strength properties. However, BHF-filled rubber vulcanizates could be useful in products such as hoses, shoe soles, mats, and sealing articles, for which its superior stiffness, hardness, and specific gravity are of immense benefit.

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References

1. Bergstrom, J. S.; Boyce, M. C. *Rubber Chem Technol* 1999, 72, 633.
2. Hofmann, W. *Rubber Technology Handbook*; Hanser/Gardner: Cincinnati, OH, 1989; p 217.
3. *Rubber Technology and Manufacture*, 2nd ed.; Blow, C. M.; Hepburn, C., Eds.; Butterworth: London, 1982; p 1.
4. Wang, B.; Panigrahi, S.; Tabil, L.; Crerar, W.; Powell, T.; Kolybaba, M.; Sokhansanj, S. Presented at the CSE/ASAE Annual Intersectional Meeting, Fargo, ND, Oct 2–3, 2003.
5. Medalia, A. I. *Rubber Chem Technol* 1977, 50, 1.
6. Wang, M. *Rubber Chem Technol* 1999, 72, 430.
7. Sreeja, T. D.; Kutly, S. K. N. *J Elastomers Plast* 2001, 33, 225.
8. Imanah, J. E. Ph.D. Thesis, University of Benin, 2003; p 61.
9. Iyasele, J. U.; Okieimen, F. E. *Proc Int Conf Chem Soc Nig Benin City* 2004, 27, 272.
10. Purseglove, J. W. *Tropical Crops, Monocotyledons*; Longman: London, 1975; Vols. 1 and 2, p 33.
11. Ishak, Z. A. M.; Bakar, A. A. *Eur Polym J* 1995, 31, 259.
12. Gill, L. S. *Ethnomedical Uses of Plants in Nigeria*; Uniben: Benin City, Nigeria, 1992; p 209.
13. *Rubber Research Institute of Malaya. Test Methods for Standard Malaysian Rubber*; Bulletin 7; Kuala Lumpur, 1970; p 1.
14. *Association of Official Analytical Chemists. Official Methods of Analysis*, 11 ed.; Washington, DC, 1970; p 4.
15. Christian, G. D. *Analytical Chemistry*, 3rd ed.; Wiley: New York, 1980; p 400.
16. *Standard Test Method for Tensile Properties of Elastomers*; ASTM D 638-99; American Society for Testing and Materials: West Conshohocken, PA, 1983; 7: 1.
17. *Determination of Abrasion Resistance: British Standard Methods of Testing Vulcanized Rubber*; BS 903 Part A9; British Standard Institutions (BSI), London, 1982.
18. Egbon, E. E.; Akinlabi, A. K.; Asia, I. O. *Adv Nat Appl Sci Res* 2003, 1, 177.
19. *Rubber Technology*, 2nd ed.; Morton, M., Ed.; Van Nostrand Reinhold: New York, 1973; p 152.
20. Matar, S.; Hatch, L. F. *Chemistry of Petrochemical Processes*; Gulf: Houston, 1997; p 114.
21. Hepburn, C. *Plast Rubber Int* 1984, 9, 11.
22. Oladimeji, M. O.; Matthew, G. O.; Adeosun, B. F. *Proc Int Conf Chem Soc Nig Benin City* 2004, 27, 345.
23. Wolff, S.; Wang, M. J.; Tan, E. H. Presented at the Meeting of the Rubber Division of the American Chemical Society, Denver, Colorado, May 18, 1993.
24. Tan, E. H.; Wolff, S.; Hadderman, M.; Grewatta, H. P.; Wang, M. J. *Rubber Chem Technol* 1993, 66, 594.
25. Billmeyer, F. W., Jr. *Textbook of Polymer Science*, 2nd ed.; Wiley: New York, 1971; p 391.
26. Osabohien, E.; Egbob, S. H. O.; Okoh, B. E. *Biosci Biotechnol Res Asia* 2006, 3, 111.
27. Yakubu, M. K.; Ogbuse, L. A.; Adekunle, A. T. *Proc Annu Tech Conf Polym Inst Nig Zaria* 2004, 15, 99.