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Advances in natural rubber seed shell utilization in polymer technology

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ABSTRACT

The valorization of rubber seed shell into useful materials for industrial applications in polymer technology is of great interest. The usefulness of this material is borne out of the ease of processing; it's readily available at low or no cost, and less abrasive to equipment. Literature and research reports have shown that rubber seed shell has gained applications in the adhesive industry as reinforcing additive, in electrode manufacturing, as filler in polymer composites, as sorbent in the uptake of heavy metals during waste water treatments, as starting material in the production carbon materials for value added products for the industry. However, there are still outstanding prospects in the utilization of this material in various areas of polymer technology such as a lingo-cellulosic source for the production of biodegradable foams, polymer gels, second generation bio-plastics and biofuels, and as surfactants. This review examines the results of a retrospective and prospective study in polymer technology of the latent properties inherent in rubber seed shell with particular emphasis on its utilization in polymer technology.

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1. Introduction

The growing interests in natural fibers has led to significant adjustments geared towards enhancing the quality of natural fibers; to make it either at par or superior to synthetic fibers. Hence, natural fibers are fast evolving material for use in reinforcing composites [1]. The utilization of natural fiber reinforced composites as construction material is essential for achieving sustainability in addition to the enormous benefits in terms of cost effectiveness, maintenance and durability. The natural rubber, initially homegrown from the Amazon Valley forest, are being grown mainly in Southeast Asia, particularly for countries like Malaysia and Indonesia, as well as in Nigeria. Also, rubber trees have been cultivated in over fourteen million acres of land [1].

The seed shell of rubber is essentially lingo-cellulosic agricultural by-product of the rubber tree [2]. The economic significance is mainly based on the exploitation of the rubber latex with minimum consideration on the value of the seed shell of the rubber tree. While major progress has been recorded in the use of the modified rubber seed shell in water and wastewater treatment, minimum research reports are recorded in literature of the usefulness of this material in other areas.

Biomass such as the seed shell of the rubber tree is available in abundance and renewable but of limited value before now [2]. These

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lingo-cellulosic by-products based on research output in polymer technology have been shown to be the major source of chemicals, fibers, as well as other industrial products [3]. These lingo-cellulosic materials are available in large quantities at very minimum cost. The utilization of this plant biomass for the production of value added industrial materials has created a source of income for small holder farmers [2].

The worth of a by-product depends on its potential usage and availability [4]. Ligno-cellulosic are useful valorization materials for several industrial applications based on the composition and properties [3]. Rice and wheat straws, as well as corn stalks; although to a limited extent, are conventional methods utilized for paper and pulp making, and for regenerated cellulose fibers as reported in literature. Pineapple, coir and banana leaves have been utilized as natural cellulose fiber source for composites, textiles and paper [5]-[9]. Research reports have shown the production of natural cellulose fibers appropriate for textile and other industrial usages from corn stalks and husks [9].

The choice of usage of lingo-cellulosics to make ethanol and other sugars through biomass pretreatment, saacharification hydrolysis, fermentation, purification has been as a result of the rise in fuel costs and scarcity of petroleum sources [10], [11]. Moreover, the earth's climatic change is the result of rising concentrations of greenhouse gases resulting primarily from fossil fuel combustion into the atmosphere [12]. Among several contaminates, the removal of organic compound are mainly considered for waste water as these compounds can precipitate at low quantities [13]. These organic pollutants such as volatile organic compounds, pesticides, and deter-gents; are detrimental to health causing cancer, flu symptoms such as chills, fever, and muscle ache, breathing harm [14]. More intense exposures trigger tracheo-bronchitis, pneumonia, diarrhea, stomach pain and serious vomiting, bone fracture, reproductive failure and potentially infertility.

This review examines the results of a retrospective and prospective study in polymer technology of the potentials of the seed shell of the rubber tree with particular emphasis on the utilization in polymer technology.

2. Composition of the Seed Shell of the Rubber Tree

The characterization of materials gives an insight to the nature and composition of the material, which helps in determining suitable utilization of the material. The chemical composition of a lingo-cellulosic is based on the nature of the particular plants. Cellulose, lignin and hemicellulose are the three major components of any lingo-cellulosic source. The ratio of these constituents in a fiber depends on the source of the fiber, the age, and the extraction conditions utilized to obtain the material [3]. The seed shell of the rubber tree has been characterized in terms of the following contents: holocellulose, lignin, ash, hemicellulose and cellulose; according to standard techniques [2].

The characteristics of the seed shell of rubber tree and the extractive cellulose yield are presented in Table 1 to Table 3. The shells were removed from the seeds, air-dried and condensed to small unspecified sizes. Eight samples of 1kg each were weighed and carbonized at varying temperatures from 100 - 800°C, for three hours each; using the technique described in [5]. Then, the carbonized shell samples were milled to fine powder, and sieved through a mesh size of 150μm. The particles, which passed through were collected and characterized using standard methods and the temperature with

Table 1 Characteristics of powdered rubber seed shell (PRSS) and extractive cellulose yield [1]

Ash (%)	Lignin (%)	Hemicellulose (%)	Cellulose (%)	Holocellulose (%)	Cellulose yield (%)
0.7-0.9	7-21	19-25	38-65	57-90	55.52

Table 2 Some physical characteristics of rubber seed shell and carbon black (N330) [13]

Parameters	RSS	CRSS	CBN 330
Loss on Ignition at 875^{0}C (%)	32.71	59.18	92.80
Moisture Content at 125^{0}C (%)	10.02	6.35	2.41
pH of Slurry	8.50	8.50	6.50
Iodine Adsorption Number (mg/g)	50.17	57.31	81.24
Oil Absorption (g/100g)	51.02	54.11	55.13
Particle Size (nm)	<150	<150	>35

RSS-Rubber seed shell, CRSS-Carbonized rubber seed shell, CB (N330)-Carbon black

Table 3 Quantitative elemental composition of rubber seed shell [5]

Components	RSS	CRSS
Potassium (%)	3.34	3.91
Sodium (%)	1.65	2.07
Calcium (%)	2.45	3.06
Magnesium (%)	0.02	0.32
Chloride (%)	1.80	0.96
Ammonia-N (%)	2.00	1.13
Phosphate-P (%)	0.43	0.02

ND-Not Detected

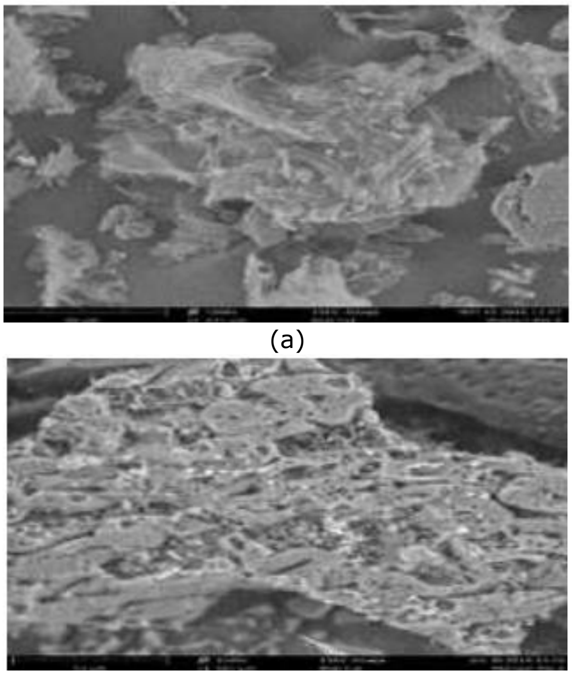
the optimum values are reported in Table 2. The elemental analysis were carried out using flame photometer and prescribed methods. The results are as shown in Table 3.

The results in Tables 1 - 3 showed the characteristics of the RSS, CRSS and the control in the study N330 carbon. The pH showed alkalinity, the likely reason being that; as the residual material is being lost on combustion, the metal content activity increases and hence alkalinity. The trend in the pH of carbon shows that high temperature makes the carbon alkaline.

Elemental analysis showed pronounced presence of the alkaline metals in the CRSS than the RSS as a result of the carbonization and the significant reduction in the presence of the non-metals in the CRSS compared to the RSS. The iodine adsorption number and the oil adsorption number in Table 2, showed significant difference between the CRSS and RSS when compared to N330 carbon. The iodine adsorption number elicits the surface area of the material [1]. This parameter indicates the macrostructure of filler and reflects the reaction and adsorption abilities [15].

3. Morphology of Rubber Seed Shell

Fig. 1 shows the microscopic morphology structure of the raw rubber seed shell and the activated rubber seed shell carbon structural images with magnification up to 300 times [16]. The structure of the fresh seed shell of the rubber tree in Fig. 1(a) showed the raw material before activation has no observable pores. However, the scanning electron microscopy (SEM) image clearly showed the presence of fine pores on the surface upon activation at 700°C. On activation, a greater distribution of pores appeared to be active sites for adsorption to take place more promptly [16].



(a)

(b)

Fig. 1 SEM of rubber seed shell (a) fresh seed shell, (b) activated seed shell [16]

4. Utilization Potentials of Rubber Seed Shell

4.1. As Filler in Polymers

One of the techniques of achieving an extension of service life of polymers is by incorporating additives into the polymer matrix, which when added to the base polymer facilitate easy processing, enhance service properties and lower the cost of the product [17].

Filler has been described as one of the main additives utilized in polymers that has marked effect and influence on polymeric materials [18]. Generally, filler operates to adjust the physical and chemical properties of vulcanizates, improve processing and reduce cost. Fillers in rubber compounds perform a central function in the physico-mechanical properties, and exercise control over the product's cost. Carbon black has been established as one of the most important classical reinforcing filler, particularly for the polymer industry [19]. However, carbon black, being a derivative of petroleum is non-renewable; in addition, it is expensive. Therefore, extensive research and improvement have been carried out in polymer technology to evaluate alternatives of filler with renewable raw materials as such rubber seed shell.

Table 4 Mechanical properties of natural rubber vulcanizates filled with the seed shell at filler loading and with carbon black at different loading [6]

Filler Loading (Phr)	Tensile strength (Mpa)	Modulus at 100%	Elongation at break (%)	Compression set (%)	Hardness (IRHD)	Abrasion resistance index
10	7.33(6.34) *20.64	1.41(1.10) *2.54	500(510) *660.40	27.25(31.40) *21.28	42.10(39.40) *45.40	40.08(42.42) *36.80
20	12.45(9.30) *26.64	1.79(1.30) *3.08	469.13(433.68) *560.10	22.77(25.15) *15.02	48.90(41.30) *50.34	44.08(40.18) *41.62
30	16.92(12.95) *28.58	2.28(1.65) *5.74	421.75(400.78) *500.09	18.79(22.04) *13.43	54.20(43.70) *56.60	43.90(39.71) *38.55
40	21.64(17.11) *34.14	2.64(1.96) *6.23	409.90(377.50) *301.04	13.68(18.40) *9.66	60.30(52.10) *60.38	40.18(39.76) *39.67
50	24.90(20.78) *39.01	3.00(2.20) *6.04	390.07(350.00) *275.22	12.23(15.65) *8.42	65.70(57.30) *66.50	42.50(40.04) *40.26

Mechanical properties of natural rubber vulcanizates filled with RSS at filler loading in parentheses.
**Mechanical properties of natural rubber vulcanizates filled with carbon black at different loading.*

The utilization of Nano carbon from seed shell of the rubber tree to produce vulcanizates materials, which are economical with synthetic composites has gained attention over the recent decades, due to low cost, accessibility of materials, easy processing, high volume utilizations and its less abrasiveness to equipment [20]. The Nano carbon was produced by pyrolysis using a locally fabricated tubular rector as shown in chemical vapor deposition processes.

The impact of the incorporation of the seed shell of the rubber tree and its carbon on the physico-chemical properties of natural rubber vulcanizates was reported [21], values are as shown in Tables 4-6. The research reports showed that the seed shell of the rubber tree is a suitable starting material for the preparation of carbon as fillers for polymers [15].

Further research reports in the chemical modification of the RSS due to poor resistance to moisture absorption and lack of good interfacial adhesion has made the utilization of natural fiber-reinforced composites attractive as been reported [6], [15], values of results are as shown in Tables 5 and 6. Table 7 shows the modulus at 100% (MPa) values for untreated and treated shell powder filled vulcanizates.

The rubber seed shell were modified using the activation and carbonization in a single step by carrying out the thermal decomposition of the rubber seed shells that was previously impregnated with ammonium chloride as activating agent and also the modification can be done using by mercerization and acetylation of the rubber seed shells.

Table 5 Tensile Strength (MPa) values for untreated and treated shell powder filled vulcanizates [6]

Samples	Loading/ Conc.		5%	10%	15%	20%	30%
	Nil	7.50					
Gum	20	5.41					
U	40	6.14					
	20		10.13	10.85	11.68	10.02	9.30
M	40		12.11	13.45	16.12	10.58	9.67
MA	20		10.13	16.50	25.45	34.98	37.40
	40		18.80	19.32	32.11	36.24	40.54
MB	20		18.95	20.36	13.52	11.11	9.92
	40		26.05	28.78	19.68	12.80	10.00
MP	20		18.54	30.88	21.87	13.56	11.11
	40		23.98	44.55	22.25	14.09	13.32

(U: Control; M: Mercerization, MA: Acetylation; MB: Benzonylation; MP: Peroxidation)

Table 6 Elongation at Break (%) values for untreated and treated shell powder filled vulcanizates

Samples	Loading/ Conc.		5%	10%	15%	20%	30%
Gum	Nil	880					
U	20	560					
	40	430					
M	20		548	510	488	540	570
	40		390	366	325	385	440
MA	20		533	513	476	451	381
	40		381	349	330	301	249
MB	20		365	348	505	496	556
	40		305	321	395	377	510
MP	20		376	298	401	400	501
	40		311	240	344	328	329

Table 7 Modulus at 100% (MPa) values for untreated and treated shell powder filled vulcanizates

Samples	Loading/ Conc.		5%	10%	15%	20%	30%
Gum	Nil	0.80					
U	20	1.40					
	40	1.62					
M	20		1.11	1.20	1.54	1.04	1.45
	40		2.21	2.67	2.81	2.64	2.34
MA	20		1.40	1.94	2.02	451	381
	40		2.22	2.44	3.47	2.46	2.55
MB	20		2.65	2.95	2.77	4.59	4.25
	40		3.87	3.76	2.89	2.87	2.41
MP	20		3.99	5.35	3.91	3.21	2.31
	40		4.32	6.11	4.44	3.66	3.56

Literature reports on the chemical modification of the fibers has revealed that it can clean the fiber surface, prevent the moisture absorption process, and chemically adjust the surface by reducing the hydroxyl groups possibly involved in the hydrogen bonding within the cellulose molecule [6], [23].

The results of Tables 5-7 showed the significant performance in the mechanical features of the chemically modified rubber seed shell when compared to the non-modified rubber seed shell. The results showed peak performance at 15% concentration of modification.

4.2. Waste water treatment

The mass transfer limitations in the use of chitosan, cellulose and other natural polymers for water purification has been solved with the use of carbon composites prepared from rubber seed shell has been reported [24]. The study was aimed at producing a value added biosorption composite material which could be a profitable technology for the uptake of organic compounds and heavy metals from aqueous media. The composite bio-sorbent prepared was done by coating chitosan onto acid treated carbon from seed shell of the rubber tree [4]. The extent of adsorption was assessed by evaluating the extent of adsorption of selected transition metals such as chromium, zinc, cadmium, and lead metal ions, as well as catechol and 4-chlorophenol from aqueous media under equilibrium conditions with uptake efficiency values (93% Cr, 86% Zn, 76%Pb, 82% Cd, 75% 4-chlorophenol, and 38% catechol) [4]. Metal ion concentrations were deduced using Perkin Elmer bulk scientific (Analyst 200) Atomic Absorption spectrophotometer (AAS). Sorption was carried out by agitating 1.0g of the adsorbents respectively for 180 mins in 100ml solutions containing varying concentrations of Zn, Cr, Cd, and Pb ions (A mechanical shaker set at 150rpm agitation speed was used). The dilution factor was used for varying concentrations of the metal ions from 10 to 50mg/l for the sorption experiment. The concentration of the heavy metal ions due to sorption were deduced by AAS after filtration. The absorbed metal ions concentration in the sorbents were determined by AAS as the difference between the initial metal ions concentration in solution and unsorbed metal ions concentration.

The removal of catechol and 4-chlorophenol from aqueous solution was estimated from measuring changes in the values of chemical oxygen demand, which is a commonly established parameter to indicate the pollution strength of wastewater.

The aqueous media samples were prepared for the organics (catechol and 4-Chlorophenol), by dissolving 11.01g of catechol, and 12.85g of 4-chlorophenol in 1L deionized water respectively, representing 0.1M stock solution from which 10ml solution was taken and diluted to 100ml as stimulate waste water sample at ambient temperature.

The graft polymerization of polyacrylonitrile onto seed shell-cellulosic of rubber (*hevea Brasiliensis*) and its assessment for heavy metal uptake from aqueous medium has been reported [2]. The report showed that hydrolysis enhances the sorption affinity of grafted cellulose toward water and metal ions. The results show that graft copolymerization of acrylonitrile onto cellulosic material derived from RSS can be initiated effectively with ceric ammonium nitrate, an initiator [25].

4.3. In Adhesives

The reinforcement of polyvinyl alcohol adhesive using rubber seed shell cellulose micro-fiber is of great practical and technical importance. Rubber seed shell was evaluated as reinforcing filler for the adhesive prepared and achieved more significant results when compared with other microfiber such as groundnut husk [26]. The physico-mechanical properties such as bond strength, peel strength test were measured and compared with those obtained for a non-filled adhesive.

It was found that the rubber seed shell cellulose micro-fiber (CMF) residue filled adhesive compared favorably with the non-filled adhesive in terms of cohesive and tensile strength. Consequently, the agricultural wastes-filled adhesive can be a substitute for investigation and can replace the non-filled adhesive for better cohesive properties.

4.4. In Electrode Materials

The use of rubber seed shell in the production of electrode materials has been reported in [27]. In the study, activated carbon derived from derived from rubber seed shell via pyrolysis process. Reduced graphene oxide (rGO) was used as an additive material in order to study the effect of the rGO in capacitive behavior. The synthesized rGO was successfully produced through the electrochemical exfoliation method then further chemically reduced the solution using hydrazine hydrate. The results showed that using rubber seed shell for the production of reduced graphene oxide in electrode manufacturing revealed that the spread of a small amount of rGO in enhancing the capacitive behavior has potentially showed its ability to be implemented as a conductive electrode material in super capacitor applications.

5. Conclusion

This study indicates the promising potential of rubber seed shell in Polymer Technology with applications in areas where agricultural waste has been applied as a substitute for petroleum-based materials. This has made the seed shell of the rubber tree a valuable material. Despite numerous chemical and physical studies that have been reported on rubber seed shell utilization in polymer Technology; there are still numerous areas that calls for the use of this material in the industry; such as in hydrogel, paint production, in the synthesis of second generation ethanol, bio-plastics, biofuels and Chemicals such as butane, ethylene, ethane etc. However, the usefulness of this material of the rubber tree is based on the following: readily accessibility of materials, low cost, easy processing, high volume usage and its less abrasiveness to equipment.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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