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A Study on the Effect of Green Fillers in Natural Rubber

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***Abstract*: Continuous research for a green non-petroleum filler environment is an attractive challenge for modern rubber research. In recent times, rubber researchers have become increasingly aware use as green reinforcement for rubber composites. Bio-fillers based on cellulose, chitin, starch, and their composition have been interesting topics because of their eco-friendliness, low cost, and good thermo-mechanical qualities in recent decades. The hydrophilic nature of supported bio fillers in plant fibers reduces the compatibility with rubber matrix. The purpose of this work is to provide an update on cellulose, chitin, and starch, with an emphasis on their impact on the mechanical properties of natural rubber (NR)-based composites reinforced with these fillers. The major parameters of NR structural categorization will be thermal and biological deterioration. As a result of the filling's tiny size, mechanical, thermal, sorption, crystallinity, and biodegradability of bio fillers/NR composites increased. Finally, cellulose, chitin, and starch will be researched in various areas to increase the usage of NR-based chemicals. The report would aim to impart all necessary details concerning from scratch in very elliptical manner.**

***Keywords*: Cellulose, Chitin, Natural rubber, Starch.**

1. Introduction

Natural latex rubber (NRL) is an emulsion solution in which rubber particles are diffused throughout the water. In NRL, a layer of weakly charged proteins and lipids covers the rubber particles, providing gel stability. At room temperature, NRL coagulates with the addition of acids such as formic acid and other acids or salts, such as calcium chloride, calcium nitrate, and so on. Because of the continuous development of new goods and the tightening of environmental restrictions, new technologies are critical in the rubber business. The following are the most recent trends in rubber industry innovative technologies: Working with innovative, high- and high-performance elastomers is a novel development elastomer. Because of renewable and easily replaceable structures, the "green" elastomer not only saves energy but is also more environmentally friendly. As an outcome, the eco-friendly elastomer solutions are around the world. The consumption of fuel products, such as black carbon, has risen dramatically as a result of rising oil prices. Therefore, substituting renewable elastomer compounds for carbon black can help to reduce the usage of non-renewable petroleum. Currently, Biodegradable and environment friendly polymers are available in a variety of renewable resources preferred by various systems. In recent years, the number of products made from renewable resources has increased dramatically. medicine, medical implants, and other applications are among them.

A. Natural Rubber

Natural rubber is a polymer that is both significant and adaptable. Rubber latex is primarily obtained from the "Hevea Brasiliensis" rubber tree or others. Latex is a sticky colloid, with white milk which is obtained by making a cut on the bark of a tree and accumulating the extracted fluid by a process called "tapping". After that, latex was purified into a rubber that could be processed. In large areas, latex is allowed to fill in the collection cup.

The properties include, it has high tensile strength and easily adheres to other materials, apart from this NR is resistant to certain kinds of chemicals, water, fatigue, wear, tear, and abrasion.

Fig. 1. Natural rubber

In composite applications, the NR are often used as matrix material whereby it is reinforced with various reinforcing agents Among the different reinforcing agents used with NR, for overcoming the disadvantages of NR, bio-fillers are used. Bio-fillers have the benefit of being abundant, low-cost, and inexhaustible when compared to synthetic nanofillers such as carbon black (CB.

The fillers such as cellulose, starch, and chitin, hold a crucial role as far as green composite applications are concerned, because of their availability, low cost, and easy production.

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Some of the advantages of the use of agro-based fillers are pollution control, environmental protection, along with abundance in nature, and low cost. Agro-based fillers, on the other hand, have poor mechanical qualities when compared to synthetic fillers.

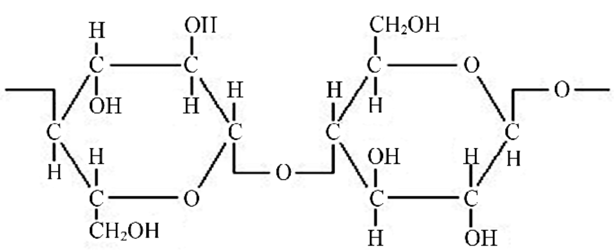
Bio-Fillers:

Petrochemical-based polymers can be replaced with cellulose, starch, and chitin. These bio- fillers are abundantly available on earth; besides, they are renewable materials since they are isolated from natural sources. These bio-fillers have great applications because of their advantages, like biodegradability, low cost, and abundance compared to synthetic fillers (carbon, glass, and aramid). Although bio-composites have numerous benefits, they also have certain drawbacks, such as moisture absorption, limited heat resistance, and poor dimensional stability.

Cellulose:

The ease of availability of cellulose makes it a popular organic filler. It also possesses excellent thermomechanical qualities, as well as low density, renewability, and biodegradability. In addition, cellulose-based materials are nontoxic because they are derived from natural sources. The composition of the cellulose have β-D-glucopyranose mer-unit, joined by β-1,4-linkages. Among the repeating units, hydrogen bonds provide stability, protective layer & linear structure to cellulose.

Sulphuric acid hydrolysis is used to extract cellulose from nano-whiskers of the Acacia Caesia plant. Textiles, composites, and flexible displays are just a few of the applications for cellulose.



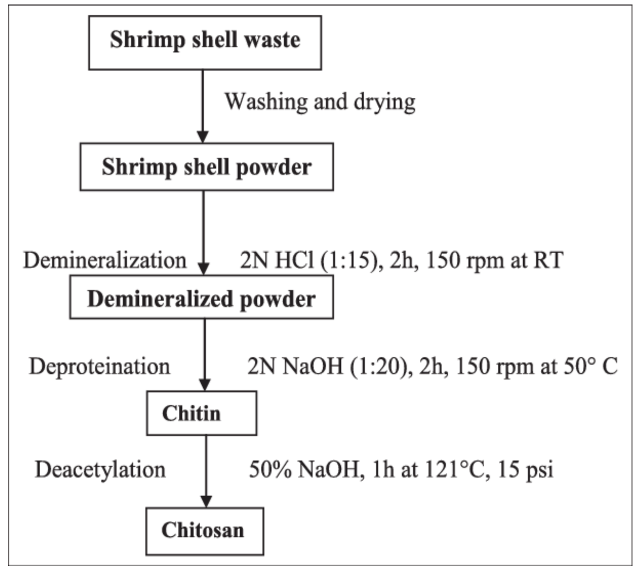
Cellulose

Due to presence of surface hydroxyl groups, Cellulose Starts agglomeration due to which it makes so difficult to get homogeneous dispersion of hydrophilic cellulose within rubber composites. Only suitable surface treatment can result in good NR composites.

Chitin:

Chitin is a linear copolymer composed of N-acetyl-d-glucosamine with a -(1–4) linkage.Chitin can be found in a variety of places, including marine animals, microorganisms, and insects. Chitin is primarily extracted from the waste of shrimp and crabs. It has variety of applications in chemical industries, biomedical, food because of its attractive properties such as biodegradability, biocompatibility and low toxicity

To separate chitin from a shellfish/shrimp shell, two key ingredients must be removed:protein and calcium carbonate, as well as a little quantity of colour and lipid that is removedduring the preceding processes.



Starch:

Starch or amylum is a polymeric carbohydrate formed from numerous glucose molecules that are joined by glycosidic bonds. It is derived from corn, potato, and rice. The two components of starch are amylose and amylopectin. Amylose is a water-soluble linear unbranched chain & the second component is amylopectin. Amylose constitutes up to 30% of starch and amylopectin 70%, depending on the botanical source. Starch is used in water treatment, pharmaceutical.

2. Composites with Natural Rubber

A. *NR-Cellulose Composite*

For the production of cellulose filled rubber composites, the well-known two-roll mixing mill process is usually used. During this process, cellulose and other curing ingredients are added to a rubber matrix step by step, then rubber sheets are prepared by compression molding under controlled temperature and pressure. Dispersing cellulose into rubber latex is done using mechanical stirring. In order to dry cellulose mixed

Table 1
Effect of cellulose on the property of rubber

Natural Rubber Composite	Effects
Cellulose nanocrystals (CNCs) filled natural rubber (NR) composites	Tensile strength of unfilled NR composite is low; however, when CTAB modified CNCs are added to NR composite, the tensile strength property improves.
MCC filled NR composites	MCC filled NR composites displayed remarkable mechanical reinforcement in the presence of maleated natural rubber (MNR) as a compatibilizer.
CNFs filled NR composites	The high fibrillated CNFs Structure Improves the tensile strength of the Natural Rubber composites.
NR latex with BC slurry	The crystallinity and tensile strength characteristics improved.
NR Latex with Isolated bagasse cellulose whiskers	The composite offers excellent mechanical characteristics and biodegrades more gradually.

rubber latex, an oven is used. A two-roll mixing mill is then used to mix different curing ingredients into a cellulose-filled rubber sample. During mixing of cellulose and rubber, cellulose typically agglomerates into the matrix. Various surface modification treatments are conducted on cellulose prior to mixing in order to obtain efficient cellulose dispersion into the rubber matrix. The hydroxyl groups on the surface of the cellulose are reacted with the functional groups of the surface modifiers using surface modification methods such as organo-silanes. As a result, because the surface hydroxyl groups are reduced, cellulose's hydrophilic feature is reduced. This is the main reason for the improved compatibility between surface modified cellulose and rubber matrix. Finally, rubber sheets' mechanical and thermal properties are calculated.

i. MCC can offer significant reinforcement in the mechanical characteristics of NR composites when just a small amount of ENR or MNR is present.
ii. CNF loading in NR composites can improve mechanical characteristics. In the NR/CNFs system, the network structure was produced via hydrogen bonding interactions between CNFs. These networks can play an important role in effectively transmitting applied stress from the NR matrix to the CNFs surface. Tensile strength decreased steadily with increased filler loading due to CNF aggregation & further the tensile strength decreased steadily with increasing filler loadingabove due to the agglomeration of CNFs.

Thermal Properties:

The most appropriate method for determining the thermal stability of rubber composites is thermogravimetric analysis (TGA). The increase in the onset decomposition temperature implies that the thermal breakdown of rubber products is being delayed; in other words, the increase in the onset composition temperature indicates that the thermal stability of rubber composites has improved.

a) The thermal stability of Natural rubber composites can be removed in the presence of ENR as compatibilizer.
b) Due to the use of the optimal amount of (2phr) of CNFs as filler, the thermal stability of the NR composite remains practically constant.

Surface modified CNCs have the most potential for use as a reinforcing filler in rubber composites among the various forms of cellulose. Thermal stability is a key characteristic for rubber productsthat will be used for a long time. Neither unmodified cellulose nor surface modified cellulose can improve the heat stability of NR compounds.

NR-Chitin Composites:

The crab's shells are rinsed completely with distilled water, and dried for 6 hours at 80 degrees Celsius. The crab shells were sieved after being reduced to powder. HCl was used to demineralize the crab shell powder. NaOH was used to deproteinize the demineralized chitin. The flow diagram for the manufacture of chitin is shown below.

Using a two-roll mill and the ASTM D 3184-80 technique for all vulcanizates, the natural rubber was masticated and blended with the additives.

Effect of Chitin on the property of natural rubber:

a) Tensile Strength

The tensile strength of NCH (Natural rubber + Chitin) decreased initially, then grew as the chitin percentage increased and reached a maximum height. At the maximum tensile strength point of a material if additional filler content is added then, reinforcing causes the material's tensile strength to diminish.

The initial drop in tensile strength can be explained by the filler's inability to support the stresses imparted by the polymer matrix at lower filler levels, & further increase can be explained that filler may support the stresses conveyed by the matrix as the filler quantity increases. The decline in tensile strength after the optimum filler ratio shows that as a material's reinforcement rises, its tensile strength improves as well, until further reinforcing causes the material's tensile strength to drop.

b) Effect on hardness

As chitin content rises, NCH hardness starts to rise as well. The hardness of the rubber greatly improved by adding chitin filler to natural rubber. Rubber's elasticity or elastic properties gets reduced when fillers are added which resulting in a more rigid rubber vulcanizate.

c) Abrasion resistance

Abrasion resistance in chitin filled Natural rubber increases with increasing filler content, beyond which abrasion resistance declines as more filler is added to the rubber. The rise in abrasion resistance with increasing filler content for NCH composites demonstrates the ability of the fillers to withstand or hinder the periodic removal of components from the surface of the vulcanizates.

Applications:

A green and pollution-free environment is essential for survival. In both structural and non-structural applications, NR-based composite materials show a high level of commercial acceptance. NR-based composites are currently widely

Table 2
Applications

Type of composites	Observation	Applications
CNCs/NR	The composites effectively removed the different types of dyes.	Composites can be used for water purification purpose
CNF/NR	NR was evenly distributed over the CNFs.	Can be used for water purification from oil and organic solvents.
ChNCs & ENR	It has good self -healing properties.	Can be used to in tires and footwear applications
NR/Starch	It has good thermo- mechanical properties.	Can be used to produce gloves.
Untreated and NaOH treated sugarcane bagasse fiber/NR	Treatment enhances the mechanical performance of the composites	composite was used to make the sandals.
Cassava starch/ NR	It reduces the moisture content and moisture absorption.	Can be used in packaging applications.

employed in the construction, building, automotive/ transportation, aerospace, and marine industries. The major requirements for these applications are high strength, renewability, stiffness, and low weight.

The applications of NR/cellulose, NR/chitin, and NR/starch composites is shown in the table 2.

3. Conclusion

This paper presented a study on the effect of green fillers in natural rubber.

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