

Effect of Chemical Treatments on Tensile Strength of Areca Fiber Reinforced Natural Rubber Composites

S. Dhanalakshmi^{1,2}, P. Ramadevi^{1,2} and B. Basavaraju^{3*}

¹Research Scholar, Department of Chemistry, Jawaharlal Nehru Technological University, Hyderabad, Telangana, India

²Department of Chemistry, K.L.E Technological University, BVB Engineering College Campus, Hubballi, Karnataka, India

^{3*}Department of Chemistry, Visvesvaraya Technological University, Alva's Institute of Engineering and Technology, Mijar, Karnataka, India.

Abstract: Every day, every second in different part of the world, new class of environmental friendly greener materials like natural fiber reinforced polymer composites with superior properties are being produced in order to improve livelihoods of people. In this study, untreated, alkali treated, potassium permanganate treated, benzoyl chloride treated and acrylic acid treated areca fiber reinforced natural rubber composites were fabricated by using heat press machine under 40%, 50%, 60% and 70% fiber loadings at a compounding temperature of 130 °C. The aim of this research work is to study the influence of chemical treatments on the tensile strength of areca fiber reinforced natural rubber composites. The investigation of tensile strength of untreated and all chemically treated areca fiber reinforced natural rubber composites was done by following ASTM standard procedures. It is observed that with increase in fiber loading up to 60%, the tensile strength of untreated and all chemically treated areca fiber reinforced natural rubber composites were increased and then, they showed a decline. Further, amongst all untreated and chemically treated areca fiber reinforced natural rubber composites, the highest tensile strength was shown by acrylic acid treated areca fiber reinforced natural rubber composites with 60% fiber loading.

Keywords: Areca fibers, Chemical treatments, Natural Rubber Composites, SEM analysis, Tensile strength.

I. Introduction

Now-a-days, natural lignocellulosic fiber reinforced natural rubber composites find incredible importance in its end-use applications, since these natural fiber reinforced natural rubber composites exhibit the combined behaviour of the soft, elastic rubber matrix and the stiff, strong fibrous reinforcement. Rubber is a versatile and adaptable thermoplastic elastomeric material that has been successfully used for many years and it combines the easy processing ability of thermoplastics and elastic behaviour of rubbers. The chemical structure of natural rubber is given in Fig.1 and it is a linear polymer of an unsaturated hydrocarbon called isoprene (2-methyl-1,3-butadiene) and there may be as many as 11,000 to 20,000 isoprene units in a polymer chain of natural rubber.

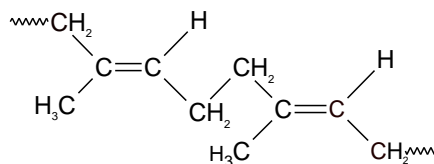


Fig. 1 Chemical structure of natural rubber

Natural Rubber is defined as a material that is capable of recovering from large deformations quickly and forcibly. It is obtained from a tropical rubber tree (*Hevea brasiliensis*) in the form of milky white fluid known as field latex. Latex is obtained by making a cut in the rubber tree and it contains 30%-40% of rubber as a colloidal solution in water. Crude rubber is a tough and elastic solid and it becomes soft and sticky as the temperature rises. Elasticity is the most important property of natural rubber and when stretched it expands and attains its original state when released and this property is due to its coil-like structure. The specific gravity of natural rubber is 0.915. Natural rubber has several advantages such as low cost, low hysteresis, high resilience, excellent dynamic properties and fatigue resistance [1]. The natural resilience of rubber is related to molecular flexibility, amorphous structure and very low intermolecular attraction. Hence natural rubber exhibits swift and easy response to force and release of force. Natural Rubber is produced by plants and hence it is renewable, inexpensive and creates no health hazard problems. Natural rubber possesses high tensile strength and modulus due to strain-induced crystallization and it also possesses good crack propagation resistance. Toughness of rubber can change the mechanical and physical properties. However, raw natural rubber has poor mechanical

properties. To improve its mechanical properties, ingredients such as accelerators, activators, cross linking agents, and fillers are commonly mixed into rubber to obtain suitable properties [2]. In particular, the incorporation of fillers in natural rubber matrix leads to significant improvement in mechanical properties [3]. But, the most important problem observed in the fabrication of natural fiber reinforced natural rubber composites is incompatibility between the hydrophilic nature of the fiber and hydrophobic nature of rubber which in turn leads to poor mechanical properties for natural rubber composites [4]. This problem can be solved by surface modification of natural fibers by various chemical treatments which help in reducing the hydrophilicity of natural fibers and in producing good adhesion between the treated fiber and polymer matrix. Consequently, there will be improvement in mechanical properties of resultant chemically treated natural fiber reinforced natural rubber composites.

Among many natural fibers, areca fiber appears to be a budding fiber, because it is cheap, available in large quantities and a very high potential recurrent crop. Areca fibers have a great prospective in the composite field. The botanical name of areca is *Areca Catechu* Linnaeus and it belongs to the *Arecaceae* (*Palmae*), palm family and *Arecoideae* subfamily. Karnataka is India's largest areca nut producing state which has a share of around 50% areca productions in the country. In Dakshina Kannada, areca is one of the major crops and is cultivated in 27,600 hectares with an annual production of about 40,000 tones. Hence, enormous quantity of unmanaged areca husk is available for further processing. The areca husk is a hard fibrous material covering the endosperm and constitutes about 60-80% of the total weight and volume of the areca fruit [5-8]. The husk fiber is composed of 55.82% cellulose, 34.28% hemicelluloses, 6.82% lignin, 1.80% moisture content and 1.28 % ash content [9]. At present, this highly cellulosic material is being used as a fuel in areca nut process. Thus, the use of this husk fiber as structural material requires a detailed study on physical, chemical and mechanical characteristics. Surface modifications of natural fibers by various chemical treatments are one of the most significant areas in the field of technical utilization of natural fiber reinforced polymer composites in various industrial sectors.

Alkali treatment improves the fiber-matrix adhesion due to the removal of natural and artificial impurities [10]. Mercerization improved the fiber wetting by fibrillation which increases the effective surface area available for the contact with the wet polymer matrix [11]. Permanganate treatment of natural fibers showed enhanced chemical interlocking at the interface and provided better adhesion with the matrix [12]. 6% and 33% improvement on tensile strength and moisture resistance properties were reported for benzoylated flax fiber reinforced low density polyethylene composites [13]. Reduced hydrophilic nature was shown by acrylated abaca fibers. Good adhesion and better mechanical performance was also observed for acrylated abaca - polypropylene composites [14]. An increase of 42.2% for tensile strength and 13.9% for flexural strength was reported for the acrylated jute fiber reinforced epoxy-phenolic matrix composites [15]. Mercerized, permanganate treated, benzoylated and acrylated areca - epoxy composites with 60% fiber loading showed 98.83%, 108.48%, 120.90% and 126.90% increase in flexural strength values respectively when compared to untreated areca-epoxy composites with same 60% fiber loading [9]. The mechanical behaviour of areca composites with 60% fiber loading was studied only by few researchers and they found to have a good flexural strength and adhesion tensile properties [16-20]. However, no literature is available on the influence of chemical treatments on the areca fiber reinforced natural rubber composite properties.

In this research work, areca fibers were treated with sodium hydroxide, potassium permanganate, benzoyl chloride and acrylic acid to impart good adhesion between the areca fibers and natural rubber matrix so that areca fiber reinforced natural rubber composites with improved properties can be obtained. Then, untreated and above said chemically treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings were fabricated by using heat press machine at a compounding temperature of 130 °C. The investigation of tensile strength of untreated and all chemically treated areca fiber reinforced natural rubber composites was done by following ASTM standard procedures and then, the influence of chemical treatments on the tensile strength of areca fiber reinforced natural rubber composites was studied.

II. Materials and Methods

2.1 Materials

Areca empty fruits were obtained from Madhu Farm House, Nilagal, Davangere, Karnataka, India. The analytical grade reagents were purchased from Qualigens Company and used as received. Natural Rubber (NR) granules which were used as matrix material in this work for the preparation of areca fiber reinforced thermoplastic elastomeric polymer composites was obtained from Akolite Synthetic Resins, Mangalore. Natural rubber granules used for the study was ISNR 5 (Indian Standard Natural Rubber-5) grade and light in color.

2.2 Fiber Extraction

The dried areca empty fruits were soaked in de-ionized water for about five days. This process is called retting; allowing the fiber to be removed from the fruit easily. The fibers were removed from the fruit

and separated with a comb. After drying in the room temperature, the fibers were combed in a carding frame to further separate the fibers into an individual state. Then a sieve was used to remove broken fibers and impurities. The resulting fibers were treated in the condition (temperature 30 °C, RH 70%) for 72 h before the chemical treatment [6, 21].

2.3 Alkali treatment

Areca fibers were soaked in a stainless steel vessel containing 6% NaOH at room temperature (30-32 °C) for 1h. The alkali treated fibers were immersed in distilled water for 24 h to remove the residual NaOH. Final washing was done with distilled water containing little amount of acetic acid. Then alkali treated areca fibers were dehydrated in an oven at 70 °C for 3 h [7, 8, 22-23].

2.4 Potassium permanganate treatment

Areca fibers, pre-treated with 6% alkali were immersed in 0.5% KMnO₄ in acetone solution for 30 min. The permanganate treated fibers were then decanted and dried in air [7, 12, 22].

2.5 Benzoyl chloride treatment

During benzoylation treatment, 6% alkali pre-treatment was used to activate the hydroxyl groups of the areca fibers. This alkali pre-treated areca fibers were soaked in 6% NaOH and agitated with benzoyl chloride for 15 min. The treated areca fibers were soaked in ethanol solution for 1 h to remove benzoyl chloride that adhered to the fiber surface. Then, the treated areca fibers were washed thoroughly using distilled water and dried in air [13, 22].

2.6 Acrylic acid treatment

6% alkali treated areca fibers were immersed in 5% acrylic acid solution at 50 °C for a period of 1 h and then acrylic acid treated areca fibers were washed thoroughly using distilled water and dried in air [22, 24-25].

2.7 Fabrication of areca fiber reinforced natural rubber composites

The fabrication of untreated, alkali treated, potassium permanganate treated, benzoyl chloride treated and acrylic acid treated areca fiber reinforced natural rubber composite plates was carried out by compression molding technique. The weight fractions, 40%, 50%, 60% and 70% of areca fiber was carefully controlled during the mixing of two ingredients. The resulting material was compression molded to the dimension of 300 x 300 x 5.0 mm³. The composite preparation process was performed in the following order. First, the heat press was pre-heated to 60 °C. Then the pressure was set as 0 MPa and the temperature was raised to 100 °C. After that the pressure was raised to 5 MPa and temperature was raised to 130 °C. Further, raised the pressure to 15 MPa, maintained the pressure and temperature for 30 min. Finally, lowered the pressure to 0 MPa, lowered the temperature to 30 °C and composite plate was removed from the heat press. The specimens were post cured for 48 h before the test.

2.8 Tensile strength

Untreated and all chemically treated areca fiber reinforced natural rubber composite specimens prepared for the tensile strength test were cut and the measurement was carried out according to ASTM D3039 standard procedure at a standard laboratory atmosphere of 30±2 °C and 65% relative humidity (RH 65%). A rectangular shape specimen with the total length of 250 mm, gauge length of 150 mm, width of 50 mm and a uniform thickness of 5.0 mm is considered for the test. The specimen was loaded in the universal testing machine until the failure of the specimen occurs. At least 5 replicate specimens were tested and the results were presented as an average of tested specimens.

2.9 Morphological study

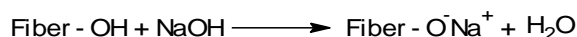
The SEM images of untreated, alkali treated, potassium permanganate treated, benzoyl chloride treated and acrylic acid treated areca fibers were taken by using JEOL JSM-T330A Scanning Electron Microscope at the accelerating voltage of 20 KV to characterize the morphological investigations of areca fibers.

III. Results And Discussion

3.1 Chemical treatment of areca fibers

Chemical treatment facilitates efficient coupling with polymeric matrix by exposing more reactive groups on the fiber surface. In this study, surface modification of areca fiber is done with suitable chemical treatments in order to optimize the effective interfacial bonding between the areca fibers and natural rubber matrix so that the areca fiber reinforced natural rubber composites with improved properties can be prepared.

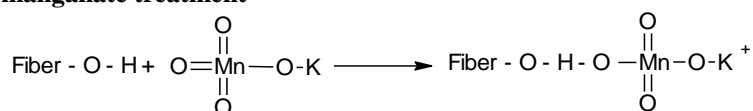
3.1.1 Alkali treatment



Scheme 1. Reaction between areca fibers and sodium hydroxide

Fiber-cell-O-Na groups are formed between the cellulose molecular chains due to the alkali treatment of areca fibers and there is increase in the amount of amorphous cellulose at the expense of crystalline cellulose. The effect of alkali on cellulose fiber is a swelling reaction and there is removal of hydrogen bonding present in the network structure and new reactive hydrogen bonds are formed between the cellular molecular chains. Hence, the natural crystalline structure of the cellulose relaxes and this provides more access to penetration of chemicals. As a result, hydrophilic hydroxyl groups are reduced and the fibers moisture resistance property is increased and effective fiber surface area for good adhesion with the matrix is increased [7-8, 22-23, 26-28].

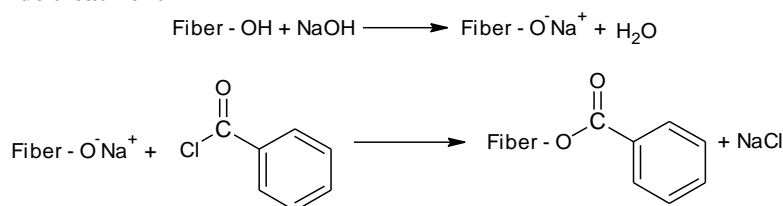
3.1.2 Potassium permanganate treatment



Scheme 2. Reaction between areca fibers and potassium permanganate

Highly reactive permanganate ions (Mn^{3+}) react with cellulose hydroxyl groups and form cellulose manganate. Permanganate ions also react with the lignin constituents and carve the areca fiber surface. As a result, areca fiber surface becomes physically rough and this reduces hydrophilic nature of the areca fibers and improves chemical interlocking at the interface and provides better adhesion with the polymeric matrix [12, 22, 29].

3.1.3 Benzoyl chloride treatment



Scheme 3. Reaction between areca fibers and benzoyl chloride

When benzoyl chloride reacts with areca fibers, an ester linkage is formed. This benzoylation treatment results in the reduction of hydrophilicity of areca fibers and hence made the areca fibers to become more compatible with polymer matrix. Benzoylation treatment also enhances thermal stability of the fibers, improves chemical interlocking at the interface and provides effective fiber surface area for good adhesion with the matrix [13, 22, 26, 30].

3.1.4 Acrylic acid treatment



Scheme 4. Reaction between areca fibers and acrylic acid

Acrylic acid provides more access of reactive cellulose macro radicals to the polymerization medium by reacting with the cellulosic hydroxyl groups of the areca fibers. An ester linkage is formed with the areca fibers by the carboxylic acid present in acrylic acid. Moisture absorption of areca fibers is reduced due to the replacement of hydrophilic hydroxyl groups by hydrophobic ester groups and effective fiber surface area for good adhesion with the matrix is increased [14, 22, 31].

3.2 Possible hypothetical models of interface of areca – Natural Rubber composites

Natural fibre reinforced polymer composites consist of natural fibers of high strength and modulus embedded into a polymeric matrix with distinct interfaces between them. The reinforcement natural lignocellulosic areca fiber and the thermoplastic elastomeric polymeric matrix natural rubber phase are chemically and/or mechanically combined or otherwise indistinct at the interface. The very much essential

factor to achieve good fiber reinforcement is the existence of good interfacial bonding between the fibre and polymeric matrix in the composites. In the composites, polymeric matrix molecules may be anchored to the fiber surface by chemical reaction or adsorption and this determine the extent of interfacial adhesion. The interfacial strength of composites depends upon the surface topology of the fibre and it is a very important factor.

Chemical treatments such as alkali treatment, potassium permanganate treatment, benzoyl chloride treatment and acrylic acid treatment exposed more reactive groups on the areca fiber surface and provided effective areca fiber surface area for good adhesion with the natural rubber matrix. Further, chemically treated areca fiber surface became more hydrophobic and there is improvement in surface characteristics such as wetting, adhesion and porosity of areca fibers. Also, there is improvement in interfacial adhesion between the treated areca fiber surface and the natural rubber matrix [7-8, 12-14, 22-23, 26-31]. Based on the hypothetical models proposed by Joseph P. V [32] and Manikandan Nair K. C. [33], the possible hypothetical models of interface of untreated, alkali treated, potassium permanganate treated, benzoyl chloride treated and acrylic acid treated areca fiber – natural rubber composites may be represented as follows.

3.2.1 Interface of untreated areca fiber – Natural Rubber composite

A possible hypothetical model of interface of untreated areca fiber surface having hydroxyl groups with the natural rubber thermoplastic elastomeric matrix is shown in Fig.2.

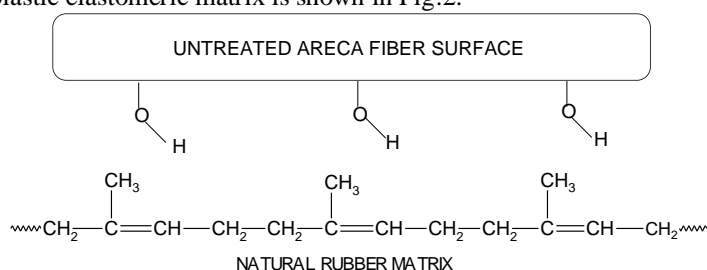


Fig. 2 A hypothetical model of interface of untreated areca fibre – natural rubber composite

3.2.2 Interface of alkali treated areca fiber – Natural Rubber composite

A possible hypothetical model of interface of alkali treated areca fiber surface having reactive -ONa groups with the natural rubber thermoplastic elastomeric matrix is shown in Fig. 3.

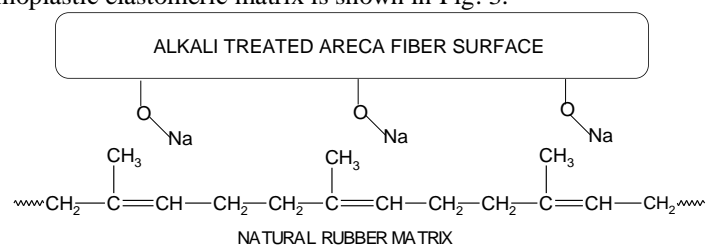


Fig. 3 A hypothetical model of interface of alkali treated areca fibre – natural rubber composite

3.2.3 Interface of potassium permanganate treated areca fiber – Natural Rubber composite

A possible hypothetical model of interface of potassium permanganate treated areca fiber surface with the natural rubber thermoplastic elastomeric matrix is shown in Fig. 4.

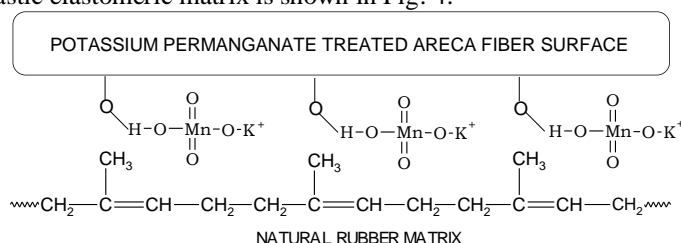


Fig. 4 A hypothetical model of interface of permanganate treated areca fibre – natural rubber composite

3.2.4 Interface of benzoyl chloride treated areca fiber – Natural Rubber composite

A possible hypothetical model of interface of benzoylated areca fiber surface having reactive $\text{C}_6\text{H}_5\text{COO-}$ groups with the natural rubber thermoplastic elastomeric matrix is shown in Fig. 5.

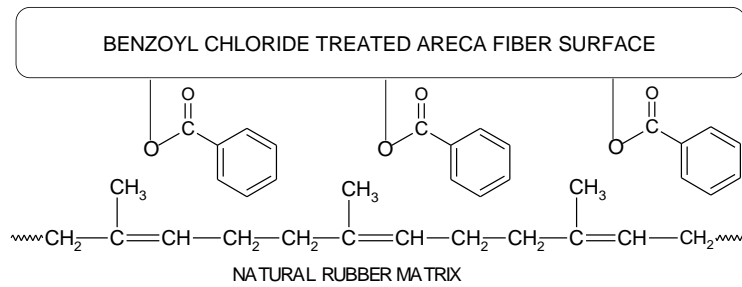


Fig. 5 A hypothetical model of interface of benzoylated areca fibre – natural rubber composite

3.2.5 Interface of acrylic acid treated areca fiber – Natural Rubber composite

A possible hypothetical model of interface of acrylic acid treated areca fiber surface having reactive $\text{CH}_2=\text{CHCOO}-$ groups with the natural rubber thermoplastic elastomeric matrix is shown in Fig. 6.

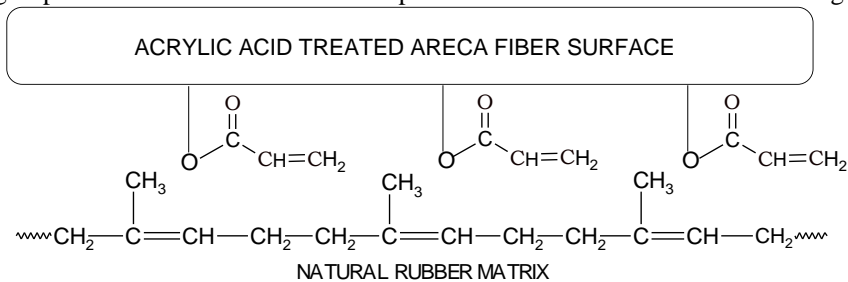


Fig. 6 A hypothetical model of interface of acrylic acid treated areca fibre – natural rubber composite

3.3 Tensile strength of areca fiber reinforced natural rubber composites

The tensile strength is defined as the force per unit area (N/mm^2 or MPa) required for breaking the composite material. This testing has been done until it complete failure or break. The effect of chemical treatments on tensile strength of untreated and all chemically treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are pictorially represented in Fig. 7.

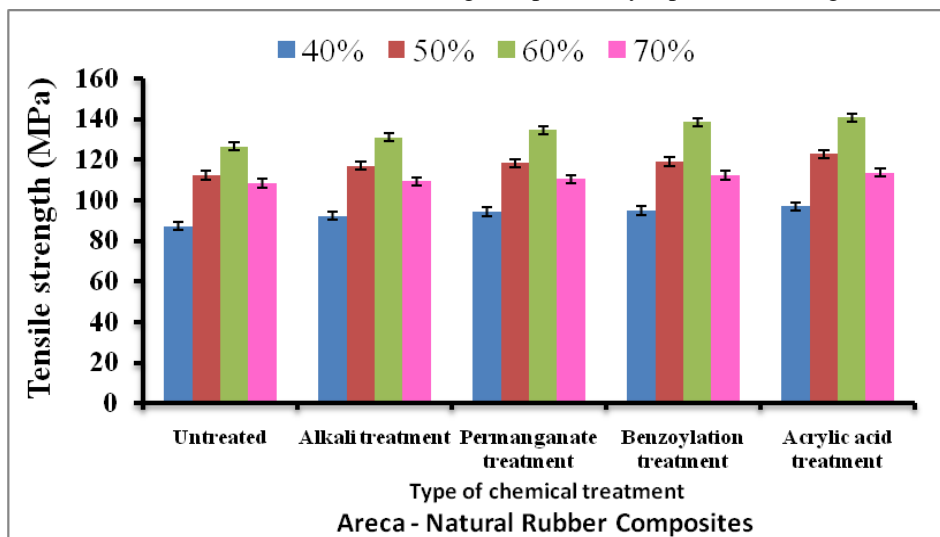


Fig. 7 Tensile strength of areca – natural rubber composites with different fiber loadings

An immense influence on tensile strength of areca fiber reinforced natural rubber composites was observed with respect to fiber content and chemical modifications of areca fibers. The tensile strength values of untreated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are 87.24 MPa, 112.28 MPa, 126.48 MPa and 108.32 MPa respectively. The tensile strength of untreated areca fiber reinforced natural rubber composites with 60% fiber loading increased by 44.97% compared to 40% fiber loading, 12.65% compared to 50% fiber loading and for 70% fiber loading, tensile strength decreased by 14.35% when compared to 60% fiber loading.

The tensile strength values of alkali treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are 92.26 MPa, 116.82 MPa, 130.82 MPa and 109.26 MPa respectively. The

tensile strength of sodium hydroxide treated areca fiber reinforced natural rubber composites with 60% fiber loading increased by 41.79% compared to 40% fiber loading, 11.98% compared to 50% fiber loading and for 70% fiber loading, tensile strength decreased by 16.48% when compared to 60% fiber loading.

The tensile strength values of potassium permanganate treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are 94.28 MPa, 118.28 MPa, 134.56 MPa and 110.52 MPa respectively. The tensile strength of potassium permanganate treated areca fiber reinforced natural rubber composites with 60% fiber loading increased by 42.72% compared to 40% fiber loading, 13.76% compared to 50% fiber loading and for 70% fiber loading, tensile strength decreased by 17.87% when compared to 60% fiber loading.

The tensile strength values of benzoyl chloride treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are 94.86 MPa, 118.98 MPa, 138.52 MPa and 112.26 MPa respectively. The tensile strength of benzoyl chloride treated areca fiber reinforced natural rubber composites with 60% fiber loading increased by 46.03% compared to 40% fiber loading, 16.42% compared to 50% fiber loading and for 70% fiber loading, tensile strength decreased by 18.96% when compared to 60% fiber loading.

The tensile strength values of acrylic acid treated areca fiber reinforced natural rubber composites with 40%, 50%, 60% and 70% fiber loadings are 96.86 MPa, 122.84 MPa, 140.82 MPa and 113.56 MPa respectively. The tensile strength of acrylic acid treated areca fiber reinforced natural rubber composites with 60% fiber loading increased by 45.39% compared to 40% fiber loading, 14.64% compared to 50% fiber loading and for 70% fiber loading, tensile strength decreased by 19.36% when compared to 60% fiber loading.

Sodium hydroxide, potassium permanganate, benzoyl chloride and acrylic acid treated areca fiber reinforced natural rubber composites with 60% fiber loading showed 3.43%, 6.39%, 9.52% and 11.34% increase in tensile strength values respectively when compared to untreated areca fiber reinforced natural rubber composites with same 60% fiber loading.

3.4 Effect of chemical treatments on tensile strength of areca fiber reinforced natural rubber composites

The hydrophilic nature of untreated areca fibers and the hydrophobic nature of natural rubber matrix resulted in incompatibility between the areca fibers and the natural rubber matrix. Hence, the untreated areca fiber reinforced natural rubber composites indicated lower tensile strength values when compared to all chemically treated areca fiber reinforced natural rubber composites with corresponding fiber loadings.

Alkali treatment of areca fibers takes out certain portion of hemicelluloses, lignin, adhesive pectin, waxy epidermal tissue and oil covering materials and reduced fiber diameter and thereby increased aspect ratio. As a result effective fiber surface area for good adhesion with the matrix is increased [8, 10-11, 22-23, 26-28].

Permanganate ions react with the lignin constituents and carve the fiber surface. As a result, areca fiber surface became rough. This improved chemical interlocking at the interface and provided better adhesion with the polymeric resin [12, 22, 29, 34].

Benzoylation treatment results in an introduction of ester linkage with areca fibers and it improved chemical interlocking at the interface and provided effective fiber surface area for good adhesion with the matrix [13, 22, 26, 30, 34].

During acrylic acid treatment, most of the hemicelluloses and lignin are removed and there is replacement of hydroxyl group by hydrophobic ester groups in the areca fiber. As a result, effective fiber surface area for good adhesion with the matrix is increased and stress transfer capacity at the interface is enhanced [14, 22, 24, 33].

Hence, alkali treated, potassium permanganate treated, benzoyl chloride treated and acrylic acid treated areca fiber reinforced natural rubber composites showed improved tensile strength values when compared with that of untreated areca fiber reinforced natural rubber composites.

With increase in fiber loading from 40% to 60%, the tensile strength values of untreated and all chemically treated areca fiber reinforced natural rubber composites increased and beyond 60% fiber loading, the tensile strength showed a decline. That is areca fiber reinforced natural rubber composites exhibited maximum tensile strength values at 60% fiber loading. This is because of better fiber distribution in matrix, less fiber fractures and effective transfer of load from matrix to fibers at 60% fiber loading. It has also been reported that high fiber content increases the probability of fiber agglomeration and further it results in regions of stress concentration and requires less energy for crack propagation [35]. Hence, with increase in fiber loading from 40% to 60%, more force is required to pullout the fibers and thereby increases the tensile strength.

The observed increase in tensile strength values with chemical modification and as well as with increase in fiber loading up to 60% is in good agreement with the results reported in literature [7, 9, 13, 15-20]. These results suggest that the areca fiber reinforced natural rubber composites are capable of showing higher tensile strength values because of strong interfacial bonding between the treated areca fibers and natural rubber matrix.

Further, in the cases of composites with 40% and 50% fiber loadings, lesser areca fiber fraction is used and areca fibers are serving as the crucial reinforcement in the natural rubber composites and hence, the reduced fiber amount resulted in a significant decrease in tensile strength compared to areca fiber reinforced natural rubber composites with 60% fiber loading [36].

The decrease in tensile strength values for untreated as well as chemically treated areca fiber reinforced natural rubber composites beyond 60% fiber loading is due to fact that the melted rubber could not reach each of the areca fibers surface because of the smaller amount of matrix material and also, there is poor interfacial adhesion and inefficient stress transfer from matrix to fibers at 70% fiber loading [37]. The factors contributing to the lower tensile strength values at 70% fiber loading may also be due to the random alignment of short areca fibers and the presence of voids in the areca fiber reinforced natural rubber composites [38].

These results clearly evidenced that chemical treatments are very effective in surface modification of the areca fibers and in improving the tensile strength of chemically treated areca fiber reinforced natural rubber composites. Amongst all the chemical treatments carried out, acrylic acid treated areca fiber reinforced natural rubber composites of 60% fiber loading showed maximum tensile strength. So, these chemically treated areca fiber reinforced natural rubber composites with 60% fiber loadings are best suitable for applications where high tensile strength is required.

3.5 Scanning Electron Microscopic image analysis of areca fibers

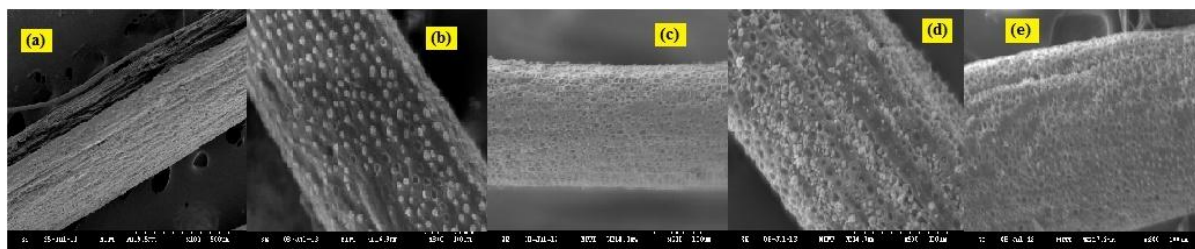


Fig. 8 SEM images of areca fibers: (a) untreated; (b) alkali treated; (c) potassium permanganate treated; (d) benzoyl chloride treated; (e) acrylic acid treated

The SEM image of untreated areca fibers is shown in Fig.8 (a). It clearly shows a network structure in which the fibrils are bound together by hemicelluloses and lignin. It also shows the presence of longitudinally oriented unit cells with almost parallel orientations. The intracellular gap is filled up by the adhesives, lignin and fatty substances and these hold the unit cells firmly in the untreated areca fibers [7-8, 22].

The SEM image of alkali treated areca fibers is shown in Fig.8 (b). It clearly shows a large number of pinholes or pits and a rough surface on the treated areca fiber surface. The observed topographical changes are due to the removal of waxy epidermal tissue, adhesive pectin and hemicelluloses [22-23]. SEM image of potassium permanganate treated areca fibers is shown in Fig.8 (c). Permanganate ions react with the lignin constituents and carve the fiber surface. As a result, areca fiber surface became physically rough. A large number of pinholes are also observed on the treated fiber surface [7, 22, 29]. The SEM image of benzoyl chloride treated areca fibers is shown in Fig.8 (d). It clearly shows a large number of pinholes and a rough surface. This is due to the removal of waxy epidermal tissue, adhesive pectin and hemicelluloses by alkali pre-treatment and further removal of fatty deposits from the areca fibers by the reaction of benzoyl chloride [22, 30]. The SEM image of acrylic acid treated areca fibers is shown in Fig.8 (e). Acrylic acid treatment removed most of the hemicelluloses and lignin and destroyed the cellulose structure. As a result, large numbers of pits and rough surface are observed on the surface of the areca fibers [14, 22, 31].

The topographical changes observed after alkali treatment, potassium permanganate treatment, benzoyl chloride treatment and acrylic acid treatment when compared to untreated areca fibers confirmed the chemical modification of areca fibers by NaOH, KMnO_4 , $\text{C}_6\text{H}_5\text{COCl}$ and $\text{H}_2\text{C}=\text{CHCOOH}$.

IV. Conclusions

Due to chemical treatments, there is introduction of some reactive groups into the fiber surface. And also, the impurities like pectin, fat and lignin present in the fiber can be removed by chemical treatments. Further, a rougher fiber surface may result after the chemical treatments. From these studies, it is clearly concluded that chemical treatments of areca fibers are of greater importance in modifying the fiber surface, in reducing the hydrophilic nature of the fiber, in enhancing the fiber - matrix adhesion and thereby increasing the properties of the polymer composites. Tensile strength values of areca fiber reinforced natural rubber composites are increased after chemical modification. Highest values of tensile strength is observed at 60% fiber loading for untreated and all chemically treated areca fiber reinforced natural rubber composites. Amongst all the chemical treatments carried out, acrylic acid treated areca fiber reinforced natural rubber composites of 60%

fiber loading showed maximum tensile strength values followed by benzoyl chloride treatment, permanganate treatment and alkali treatment at same 60% fiber loading. Hence, the areca fiber reinforced natural rubber composites can be considered as a very promising material for fabrication of light weight materials and can be effectively used in industrial sector like automobile body building, office furniture, packaging industry, partition panels, etc.

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