International Journal of Sciences:
Basic and Applied Research
(IJSBAR)

ISSN 2307-4531
(Print & Online)

http://gssrr.org/index.php?journal=JournalOfBasicAndApplied

Abaca Fiber Reinforced Epoxy Composites: Evaluation of Impact Strength

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Abstract

This study focuses on the fabrication and characterization of untreated, alkali treated, acrylated, benzene diazonium chloride treated, and permanganate treated abaca fiber reinforced epoxy composites. Composites are prepared with different fiber loadings of 10%, 20%, 30%, 40%, 50% and 60% by hand lay-up technique. Impact properties for the prepared composites were investigated and comparison is made between impact properties of untreated and treated abaca fiber reinforced epoxy composites. This study revealed that treated composites showed improved impact strength compared to untreated abaca fiber reinforced epoxy composites and 40% fiber loading showed optimum properties. Amongst all the treatments carried out, benzene diazonium chloride treated fiber composites of 40% fiber loading showed maximum impact strength of 7.68 mJ/mm^2, followed by acrylic acid treatment, permanganate treatment and alkali treatment with impact strength values of 7.12 mJ/mm^2, 6.8 mJ/mm^2 and 6.5 mJ/mm^2 respectively.

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Keywords: Abaca fibers; Benzenediazonium chloride; fiber loading; impact strength; epoxy resin.

1. Introduction

Composites are materials consisting of two or more chemically different components having different physical properties. The characteristics of these composites are totally different from that of the individual components. Now a day’s natural fiber reinforced composites are gaining lots of popularity because of their structural applications. They are having high specific strength, durability and design flexibility, high fatigue resistance; light in weight, yet capable of bearing harsh loading conditions and hence are used in aerospace components like wings, tail, fuselage panels, propellers etc. They are also used in boat hulls and decks, bicycle frames racing car bodies, baseball bats, and fishing rods. The two main components of composites are matrix and reinforcement. Matrix supports the reinforcement and helps the reinforcements to stay in proper position and orientation. Matrix is less hard and more ductile and is responsible for composite toughness. Reinforcement is embedded in the matrix and is the load carrying material and strengthens the composites by imparting its properties to the matrix. It is harder than matrix. This synergistic approach imparts superior properties to the composites which the individual components are lacking. By selecting appropriate combination of matrix and reinforcement, it is possible to tailor the properties of the composites for suitable structural applications.

Polymers are the most commonly used matrix material because of the following reasons: Polymer processing does not require high pressure and temperature, the equipments required for fabrication of composites are simple, also, polymers are having less strength and stiffness which can be compensated by reinforcing materials like fibers. The advantages of using natural fibers are: they are available in abundance, have low cost, low density, renewable, biodegradable, sustainable, provides waste management solutions; wear and tear on the processing equipment is reduced as they are less abrasive and no health hazards. Even though natural fibers are having many advantages, they too suffer from certain drawbacks. The major drawback during composite fabrication is the incompatibility arising between fiber and matrix as fiber is hydrophilic and matrix is hydrophobic. This results in poor fiber matrix adhesion thereby degrading the mechanical performance. But this can be overcome by various chemical treatments like alkali treatment, acrylation, benzene diazonium chloride treatment and permanganate treatment which modifies fiber surface and improves the properties [1-9].

Natural fibers differ in their chemical composition and properties depending on the harvesting conditions and test method even for the same fiber. Natural fibers are considered as composites in which cellulose fibrils are held together in lignin and hemicelluloses matrix. The efficiency with which the natural fiber can act as reinforcement mainly depends on cellulose and its crystallinity. The major components of natural fibers are cellulose, hemicelluloses, lignin, pectin and waxes. Cellulose is a natural polymer consisting of ß – 1, 4 glycosidic linkages at C1 and C4 positions. Each repeat unit consists of three hydroxyl groups which are capable of forming hydrogen bonds. Cellulose is crystalline, linear, and resistant to alkali and oxidizing agents but hydrolyzed by acids. It provides strength, stiffness and structural stability to the fiber. The mechanical properties of natural fibers depend on its cellulose type because each type of cellulose has its own cell geometry and the geometry determines the chemical properties. Hemicelluloses are polysaccharides consisting of several 5 and 6 carbon ring sugars. They are branched, non crystalline and degree of polymerization is less than that of cellulose. They are hydrophilic, soluble in alkali and hydrolyzed by acids. Lignin is amorphous, hydrophobic and is responsible for rigidity of plants. It is a derivative of 4-hydroxy-3-methoxy phenyl propane. It is insoluble in most solvents and cannot be broken down into simpler monomer units. Pectin provides flexibility to the plants. The outer cell wall is porous and consists of pectin and other non-structural carbohydrates. The pores of the outer skin are the prime diffusion paths of water through the material. Waxes consist of different types of alcohols which are insoluble in water and in several acids such as palmitic acid and stearic acid [10, 11, 12].

The botanical name of abaca is Musa Textilis which is a species of banana grown extensively in Philippines. It is also called Manila hemp. Abaca plant belongs to the banana family Musaceae, Genus Musa and species Musa Textilis. The Republic of the Philippines is the largest producer producing around 50,000 tons per annum. Abaca fibers are used in the production of handicraft products such as fashion accessories, decorative accessories, furniture, garments, textile, packaging materials, and table-top accessories. Abaca fibers are also used in sinamay weaving. Sinamay materials are used in making gift boxes, packaging materials, decorative accessories, wall coverings, fashion accessories, footwear, tabletop accessories etc. Abaca fibers are also used in making currency and bank notes, tea bags, coffee filters, surgical caps and masks. Abaca fibers also have several medical and industrial applications: such as for orthopedic materials (joint replacements and fracture healing implants); as material composites to replace glass fibers in the manufacturing of cars, planes, yachts; for building materials such as fiberboards, ceramic tiles etc. Abaca fiber composites are used extensively in automotive industry. Mercedes Benz used abaca fiber reinforced polypropylene composites in automobile body parts. Daimler Chrysler used these abaca fiber reinforced composites in under floor protection of passenger cars [13, 14]. The use of abaca fiber instead of glass fiber reduces the weight of automotive parts, bringing about 60% savings in energy and reduces CO_2 emissions making it environment friendly. These composites are also used in construction as well as packaging industries.

Abaca is prized for its great mechanical strength, buoyancy, resistance to salt water damage, and long fiber length – up to 3 m. Ease of availability, sustainability, high tensile strength, resistance to rotting and specific flexural strength nearer to that of glass fibers makes abaca fibers superior [15].

Alkali treatment helps in the removal of hydrogen bonding in the network structure of fibers which substantially reduces moisture absorption and also changes the surface morphology [16]. Acrylation and acetylation decreased the dissemination of water into the fiber making it more hydrophobic. This is due to the replacement of hydroxyl group by hydrophobic ester groups [17**].** In permanganate treatment highly reactive permanganate ions react with cellulose hydroxyl groups forming cellulose manganate. This reduces hydrophilic nature of the fiber and also improves the mechanical inter locking with the matrix [18].

Thermoplastics and thermo sets are the two main categories of polymers which are used as matrix material in composite fabrication. Thermosetting materials undergo curing during fabrication and once they become rigid, they cannot be remolded. Epoxy resins are most commonly used matrix material because of its good adhesive nature, good electrical insulating properties, good mechanical properties and good chemical and environmental resistance.

Impact strength is the ability of a material to absorb shock and impact energy without breaking. The impact strength is calculated as the ratio of impact absorption to test specimen cross-section. Toughness is dependent upon temperature and the shape of the test specimen. Impact strength is determined by Charpy impact test. Impact strength is a very important property of a material governing the life of a structure. For example, in the case of an aircraft, impact can take place by a bird hitting a plane while it is cruising, or during takeoff and landing the aircraft may be struck by debris that is present on the runway. The capacity of the aircraft to withstand the impact depends on the strength of the material.

Impact tests are used in studying the toughness of material. A material's toughness is its ability to absorb energy during plastic deformation. Brittle materials have low toughness and they can endure a small amount of plastic deformation. The impact value of a material can also change with temperature. Generally, at lower temperatures, the impact energy of a material is decreased.

This study mainly focuses on the preparation of untreated, alkali treated, acrylated, benzene diazonium chloride treated and permanganate treated abaca reinforced epoxy composites and the effect of these treatments and the effect of fiber loading on the impact properties of the composites. Composites were prepared by hand lay-up technique method with different fiber loadings.

2. Materials and Methods

2.1 Materials

Abaca fibers were collected from the Maruthi Peach Finishing Company, Tirupur, Chennai, India and Analytical grade reagents were purchased from Qualigens Company and used as received. The epoxy resins and hardener were procured from Akolite Synthetic Resins, Mangalore.

2.2 Alkali treatment of abaca fiber

The fibers are first dewaxed by soaking in 1:2 mixtures of ethanol and benzene for 72 h, followed by washing with deionised water and then air dried. The dewaxed fibers were immersed in 6% NaOH solution at room temperature (30-32 ^{o}C) for 1h, where the total volume of the solution was 15 times the weight of the fibers. The fibers were kept in alkaline solution for 36 hours at a temperature of 30 ^{o}C; they were thoroughly washed in running water and then neutralized with 2% acetic acid solution. Lastly, the fibers were washed again in running water to remove the last traces of acid sticking to it, so that the pH of the fibers was approximately 7. Then, they were dried at room temperature for 48 hours to obtain alkali-treated fibers [15, 19].

2.3 Acrylic acid treatment of abaca fiber

Alkali treated abaca fiber was soaked in 1% acrylic acid (AA) solution at 50 ^{o}C for 1 h, then washed with distilled water thoroughly and dried in an oven for 24 h at the temperature of 70 ^{o}C [15].

2.4. Permanganate treatment of abaca fiber

The fibers were soaked in 6% NaOH for 30 min and then thoroughly washed with distilled water. These alkali pre-treated fibers were treated with 0.5% $KMnO_4$ in acetone solution for 2 min respectively, washed with water and dried in an oven at 80 ^oC [17, 18].

2.5 Preparation of benzenediazonium chloride

8 cm^3 of *concentrated* hydrochloric acid was added to a boiling tube containing 3 cm^3 of phenyl amine (aniline) and 10 cm^3 of water, the mixture was shaken until the amine has dissolved, and then the solution was cooled to 5 ^oC by cooling it in an ice bath. After that a solution of sodium nitrite (3 g in 8 cm^3 of water), previously cooled to 5 ^oC was added. The temperature of the mixture was maintained below 10 ^oC during the addition [19].

2.6 Benzenediazonium treatment of abaca fiber

The abaca fibers were chopped into a length of 10 mm, washed with distilled water, and was then dehydrated in an oven at 70 ^oC for 24 h. The dried fibers were immersed in 6% NaOH solution taken in a 2.0 L glass beaker for 10 min at about 5 ^oC. A freshly prepared diazo solution was then poured slowly into the above mixture with constant stirring. Fibers were then taken out, washed with soap solution followed by distilled water, and finally dried in an open air for 48 h [20].

2.7 *Composite fabrication*

Abaca fibers were air dried for 24 h at laboratory temperature and then dried in oven for 24 hours at 105 °C to remove free water present in the fiber. The dried fibers were subsequently cut into lengths of 10 mm and they were mixed with epoxy resin. The composite preparation process was performed in the following order using hand lay-up technique. First, the Epoxy-556 resin and the HY951 hardener were mixed in the ratio of 10:1. One half of resin was placed inside the mixing chamber for about 1 min at 20 RPM; then fibers were added over a period of 2 min. Then, the other half of the epoxy resin was placed inside the mixing chamber and the mixing speed was increased to 30 RPM for 5 min. The resulting material was compression moulded using a Santec compression moulding press. The constant load of 50 kg was applied on the mould for 24 h at laboratory temperature, 30±2 ^oC. Finally, the composites were post-cured at room temperature for 15 days. The volume fractions 10, 20, 30, 40, 50 and 60% of fiber was carefully controlled during the mixing of two ingredients. The moulds have been prepared with dimensions of 300 × 300 × 10 mm. Equation (1) was used in the composite fabrication.

$$V_f = \frac{\frac{W_f}{\rho_f}}{\frac{W_f}{\rho_f}+\frac{W_m}{\rho_m}} \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where W_f is the weight of fiber (g), W_m is the weight of matrix (g), V_f is the fiber volume fraction (%), ρ_m is the density of matrix (g/cm^3) and ρ_f is the density of fiber (g/cm^3).

2.8 Impact Strength of Composites

The prepared composite slabs were cut into required dimensions with the help of zig saw for mechanical tests. The appropriate ASTM methods were followed while preparing the specimens for test. At least five replicate specimens were tested and the results were presented as an average of tested specimens. The tests were conducted at a standard laboratory atmosphere, 30±2 ^{o}C and 65% relative humidity).

Impact energy absorbed by the specimens were determined by performing Charpy method of impact testing as per ASTM D 6110-97 with notched specimens using Instron Pendulum Tester (9050 Manual Model). The width and depth of each specimen was measured with a micrometer screw gauge to the nearest 0.01 mm, and the length was measured to the nearest 0.1 mm with digital caliper. The mean specimen dimensions were used to calculate strength. For the test, specimen of length of 55 mm with square cross section of 9.5 mm side and the U notch is made at the center of the specimen for a depth of 4 mm.

2.9 Morphological Study

Scanning electron microscopy (SEM) images of untreated, alkali treated, acrylated, and permanganate treated abaca fiber were obtained from JEOL JSM-T330, a scanning electron microscope at the accelerating voltage of 20 KV.

3. Results and discussion

3.1 Chemical treatment of fibers

Alkali treatment helps in the removal of hydrogen bonding in the network structure of fibers which substantially reduces moisture absorption. Alkali treatment reduces fiber diameter and thereby increases the aspect ratio. Increase in aspect ratio improves interfacial adhesion and increases the mechanical properties. Alkali treatment increases surface roughness resulting in better mechanical interlocking and the amount of cellulose exposed on the fiber surface. This increases the number of possible reaction sites and allows better fiber wetting. This treatment increases the amount of amorphous cellulose at the expense of crystalline cellulose. Abaca fiber morphology significantly changed due to alkali treatment [10, 11, 19].

$$\text{Fiber- OH} + \text{NaOH} \rightarrow \text{Fiber-ONa} + H_2O$$

Acrylation decreased the dissemination of water into the fiber making it more hydrophobic. Acrylic acid treated abaca fiber showed lower moisture absorption. This is due to the replacement of hydroxyl group by hydrophobic ester groups [10, 11, 15].

$$\text{Fiber- OH} + CH_2{=}CHCOOH \rightarrow \text{Fiber-}OCOCH_3 + H_2O$$

In permanganate treatment, highly reactive permanganate ions react with cellulose hydroxyl groups and forms cellulose manganate. This treatment enhances chemical interlocking at the interface and provides better adhesion with the matrix. It reduces hydrophilic nature of the fiber [10, 11].

$$\text{Fiber- OH} + KMnO_4 \rightarrow \text{Fiber- O-H –O-Mn} (=O)_2 \text{-OK}^{+I}$$

Benzene diazonium chloride treatment is also used in decreasing the hydrophilic nature of fibers by increasing fiber matrix adhesion. It undergoes coupling reaction with OH groups of the fiber forming diazo cellulose compound [13].

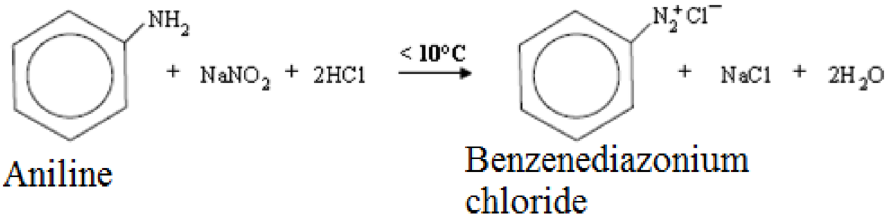

Fig. 1. Synthesis of benzene diazonium chloride

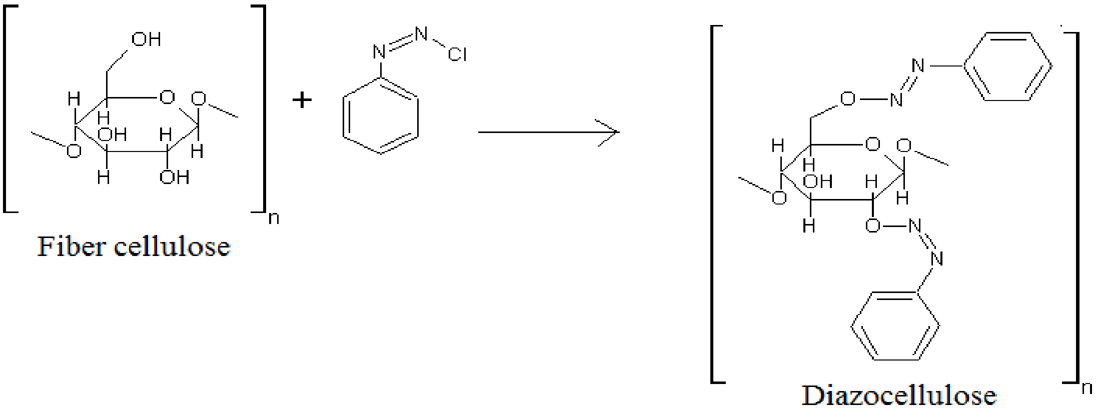

Fig. 2. Interaction between fiber cellulose and benzene diazonium chloride

3.2 Impact strength of composites

On impact, the energy of the pendulum is transferred to the test specimen and a part of the energy is consumed during fracture of the specimen. Charpy impact test determines the amount of energy absorbed by a material during fracture and is a measure of materials toughness and acts as a tool to study brittle - ductile transition. The impact strength of fiber reinforced composites depends upon fiber aspect ratio and fiber rigidity. The total energy absorbed by the composite is the sum of the energy consumed during plastic deformation and the energy needed for creating new surfaces. The impact strength of abaca fiber reinforced epoxy composites based on different fiber loadings and different chemical treatments are illustrated in Figure 3.

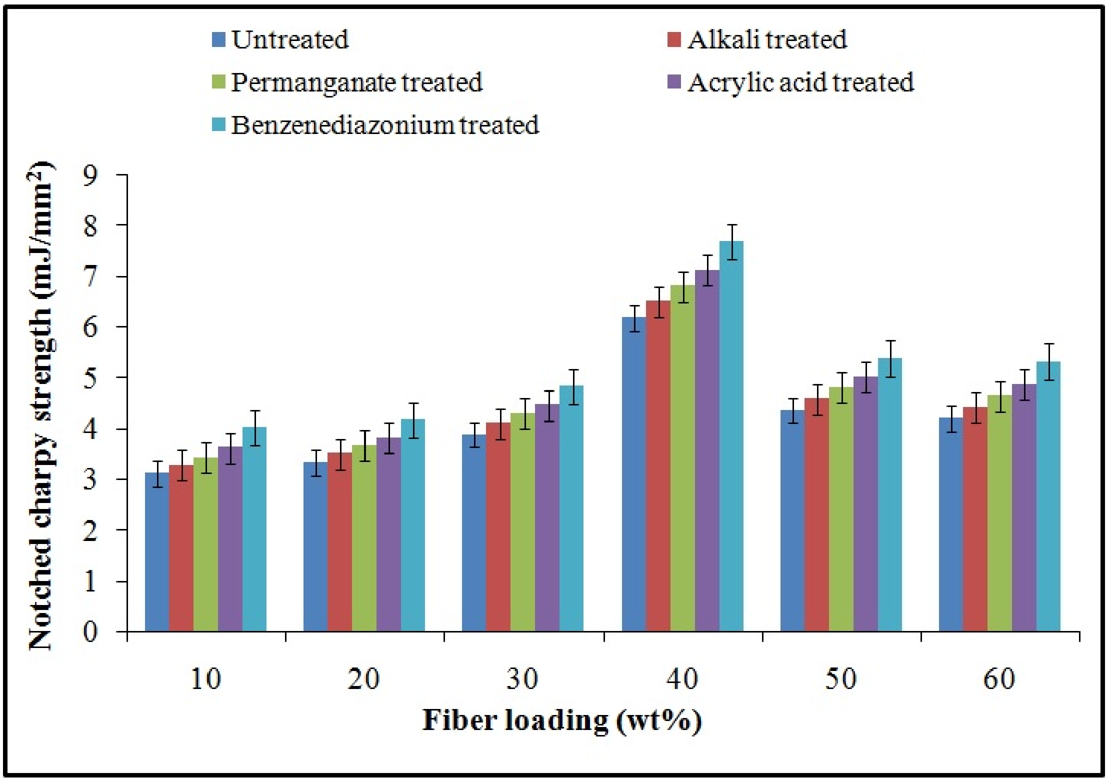

Fig . 3. Impact strengths of Abaca Epoxy Composites

In this work, abaca fiber reinforced epoxy composites were fabricated with different fiber loadings of 10%, 20%, 30%, 40%, 50% and 60% using hand lay-up technique. From the prepared composites, test specimens were cut to size according to ASTM standards and were evaluated for impact properties. It is a must to know optimum fiber loading to get good impact properties [13]. So, the variation of impact strength with different fiber loadings and different chemical treatments were studied. From figure 3 it is clear that, amongst all the composites prepared, composites with 40% fiber loading showed maximum impact strength of 7.68mJ/mm^2 for benzene diazonium chloride treatment.

Irrespective of the chemical treatment, all treatments showed higher impact strength than the untreated fiber reinforced composites. The observed impact strength values of composites can be understood in terms of chemical constituents of fiber.

In case of untreated abaca-epoxy composites the impact strengths of 30%, 40% and 50% fiber loadings are 3.88 mJ/mm^2 , 6.18 mJ/mm^2 and 4.36 mJ/mm^2 respectively. For 40% fiber loading impact strength increased by 59.27% compared to 30% fiber loading and for 50% fiber loading it decreased by 29.44% compared to 40% fiber loading.

Alkali treatment removes lignin and hemicelluloses and exposes more hydroxyl groups on the fiber surface making them available for bonding with matrix improving the adhesion between the fiber and matrix. Not only that, it also cleans the surface, reduces fiber diameter and increases the aspect ratio, increases stress transfer capacity thereby increasing the impact strength [9]. In case of alkali treated abaca-epoxy composites the impact strengths of 30%, 40% and 50% fiber loadings are 4.1 mJ/mm^2 , 6.5 mJ/mm^2 and 4.58 mJ/mm^2 respectively. For 40% fiber loading impact strength increased by 58.54% compared to 30% fiber loading and for 50% fiber loading it decreased by 29.54% compared to 40% fiber loading.

Permanganate treatment provides better interlocking at the interface by making the surface rough and improves fiber matrix adhesion. In case of permanganate treated abaca-epoxy composites the impact strengths of 30%, 40% and 50% fiber loadings are 4.3 mJ/mm^{2,} 6.8 mJ/mm^2 and 4.8 mJ/mm^2 respectively. For 40% fiber loading impact strength increased by 58.14% compared to 30% fiber loading and for 50% fiber loading it decreased by 29.41% compared to 40% fiber loading.

The reaction between fiber and acrylic acid enhances stress transfer capacity at the interface and improves the impact strength. In case of acrylated abaca-epoxy composites the impact strengths of 30%, 40% and 50% fiber loadings are 4.46 mJ/mm^{2,} 7.12 mJ/mm^2 and 5.02 mJ/mm^2 respectively. For 40% fiber loading impact strength increased by 59.64% compared to 30% fiber loading and for 50% fiber loading it decreased by 29.49% compared to 40% fiber loading.

The coupling reaction between fiber and benzene diazonium chloride facilitates fiber matrix adhesion and improves the impact strength. In case of benzene diazonium chloride treated abaca-epoxy composites the impact strengths of 30%, 40% and 50% fiber loadings are 4.82 mJ/mm^2 , 7.68 mJ/mm^2 and 5.38 mJ/mm^2 respectively. For 40% fiber loading impact strength increased by 59.34% compared to 30% fiber loading and for 50% fiber loading it decreased by 29.95% compared to 40% fiber loading.

In case of all chemically treated abaca-epoxy composites the impact strength increased upto 40% fiber loading, and afterwards for higher fiber loading impact strength decreased. At 40% fiber loading it showed maximum value. This is because at 40% fiber loading, there is better fiber distribution in matrix, less fiber fractures and effective transfer of load from matrix to fibers. As fiber loading increased beyond 40%, impact strength decreased due to poor interfacial adhesion and inefficient stress transfer from matrix to fibers [21, 22].

In comparison to untreated fiber composites of 40% fiber loading, alkali, permanganate, acrylated and benzene diazonium chloride treated fiber composites showed 5.17%, 10.03%, 15.21% and 24.27% increase in impact strength respectively. From these results it is very clear that chemical treatments are effective in improving the mechanical properties and benzene diazonium chloride treated fiber composites of 40% fiber loading showed maximum impact strength. So, these composites are best suited for applications where high impact strength is required.

Md. Rezaur Rahman et al showed that impact strength increased with increase in fiber loading [2, 23, 24]. One of the reasons for decrease in impact strength is fiber pullout. As fiber loading increases, more force is required to pullout the fibers thereby increasing the impact strength. The decrease in impact strength may be due to the micro spaces between the fiber and the matrix which initiates micro cracks on impact and results in crack propagation leading to failure [25].

3.3 Morphological Study

The SEM image of untreated abaca fibers (Figure 4) shows a network structure of the fiber in which the fibrils are held together by hemicelluloses and lignin. Untreated fiber reinforced composites, showed poor mechanical performance because of easy fiber pullout from the interfacial region due to poor compatibility between the fiber and matrix.

The SEM photographs of alkali treated, acrylated and permanganate treated abaca fibers clearly indicate that compared to untreated fibers, the surface roughness of treated fibers increased due to partial removal of hemicelluloses and lignin [19, 26]. This improved fiber matrix adhesion and stress transfer efficiency which in turn improved the mechanical performance of the composites [3].

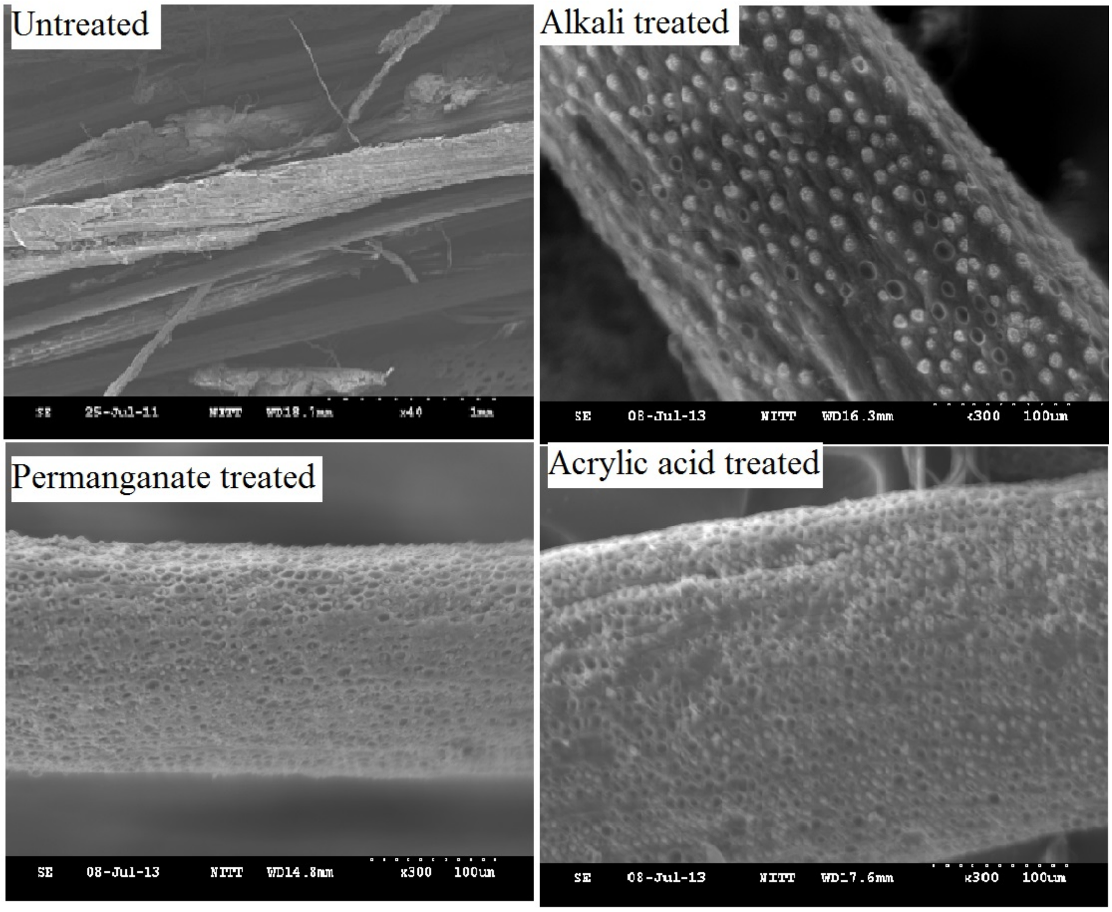

Fig. 4. SEM images of abaca fiber

4. Conclusions

From these studies we can conclude that chemical treatments are very effective in modifying the fiber surface, reducing the hydrophilic nature of the fiber, improving fiber matrix adhesion there by increasing the mechanical performance of the composites. Untreated Composites with 40% fiber loading showed maximum impact strength of 7.68 mJ/mm^2. For untreated as well as for all chemically treated abaca-epoxy composites the impact strength increased with increase in fiber loading from 10% to 40% and beyond 40% it showed a decline. Amongst all the treatments carried out, benzene diazonium chloride treated fiber composites of 40% fiber loading showed maximum impact strength of 7.68 mJ/mm^2, followed by acrylic acid treatment, permanganate treatment and alkali treatment with impact strength values of 7.12 mJ/mm^2, 6.8 mJ/mm^2 and 6.5 mJ/mm^2 respectively.

Acknowledgements

The first author would like to thank the Management of KLE Society's BVB College of Engineering and Technology, Hubli, Karnataka, India for the kind encouragement and constant support provided. She sincerely thanks Prof. Ashok Shettar, Principal, KLE Society's BVB College of Engineering and Technology, without his encouragement and motivation, this work would not have been possible.

The author, Basavaraju Bennehalli is thankful to the Vision Group on Science & Technology, Department of IT, BT and Science & Technology, Government of Karnataka for financial support in the form of sanctioning a Research Project to carry out the present investigation (grant number VGST/CISEE/2012-13/282 dated March 16, 2013).

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