NATURAL FIBER REINFORCED POLYMER COMPOSITES

Ajmal Hussain Barbhuiya[1] and Kochi Ismail*[2]
[1]*Regional Institute of Science and Technology, Killing Road, 9th Mile, Ri-Bhoi, Meghalaya-793101*
[2]*C/o. Department of Chemistry, North-Eastern Hill University, Shillong 793022*
*For Correspondence. (kinehu@hotmail.com)

Abstract: Hybrid composites of epoxy matrix and four mixed fibers were prepared. The epoxy matrix is from BADGE and the hardener PPA-7040 mixed in 1:1 volume ratio. The four mixed fibers used for reinforcement are (i) murta bast + jute, (ii) murta bast + luffa, (iii) murta bast + coir, and (iv) murta bast + betel nut fiber. The hybrid composites were analyzed and characterized on the basis of density, water absorption, thermal gravimetric analysis, tensile/flexural/compressive/impact strengths and hardness studies. Murta and jute form the best mixture for making hybrid composite as this composite possess superior physical and mechanical properties. The amount of water absorbed by a hybrid composite does not appear to have any correlation with the chemical composition of the fibers used for reinforcement.

1. Introduction:

Materials of various types are required in our day-to-day life for diverse applications. In the development of any technology, the development of materials with specific properties is a basic requirement. Material science or development of target materials has therefore always been in the centre stage of research and development. One way of achieving a target material with specific properties is by fabrication of composites. A composite is a material made from two or more constituent materials having significantly different physical or chemical properties that, when combined, produce a material with characteristics different from the individual components.

Composites exist in nature, which are classified as natural composites. Natural composites exist in both animals and plants. For example, wood and bone are actually composites. Wood contains long cellulose fibers held together by lignin, a much weaker substance. The two weak substances, cellulose and lignin, together form a much stronger wood. The bone in human body is made from a hard but brittle material called hydroxyapatite (which is mainly calcium phosphate) and a soft and flexible material called collagen, which is a protein.

The benefits of composites were known to people from early days and hence composites were prepared according to the requirement using human skill and expertise, which are classified as man-made composites. People prepared composites by mixing mud with straw for construction purpose. Gypsum mortar and lime mortar were used in ancient days for construction. Concrete is a mixture of stone chips, cement, sand and water. It has good compressive strength. On adding metal rods or wires to the concrete, bending strength of concrete increases. Concrete containing such rods or wires is called reinforced concrete or more commonly as reinforced cement concrete (RCC). Another example of early man-made composite is plywood prepared by gluing wood strips.

Modern composites are of three types (based on matrix). (i) Metal matrix composites. (ii) Ceramic matrix composites. (iii) Polymer matrix composites.

Metal matrix composites are mostly used in aeroplanes, rockets and space vehicles due to their high-temperature resistance and low coefficient of thermal expansion. In making metal matrix composites inorganic or ceramic fiber or particulate phase is embedded into light-weight metals. Addition of SiC fibers to metal matrix like aluminum produces a composite which has coefficient of thermal expansion lower than the metal matrix itself. Addition of carbon fiber to aluminum provides a composite with much higher modulus than the metal. Carbon reinforced copper, SiC reinforced copper, Al_2O_3 reinforced aluminum and SiC reinforced aluminum are some of the metal matrix composites that are used in manufacturing various parts of spacecrafts. The antenna boom on the Hubble Space Telescope is made of a graphite-aluminum composite.

Ceramic matrix composites consist of ceramic fibers like SiC embedded in ceramic matrix. Ceramic matrix composites are lighter than metal matrix composites, can withstand much higher temperatures and can also be made as strong as metal. Usually ceramic matrix composites are made by combining oxide fibers with oxide matrices and non-oxide fibers with non-oxide matrices. Thus, the main types of ceramic matrix composites are carbon fibers in carbon matrix (C/C), carbon fibers in SiC matrix (C/SiC), SiC fibers in SiC matrix (SiC/SiC) and oxide fibers in oxide matrix (Ox/Ox). Some of oxides used as matrices and also for making ceramic fibers are alumina, zirconia and silica. Ceramic matrix composites are used in many high temperature processes. For example, hot gas valves used to control gas flow in gas fired high temperature furnaces are manufactured from Ox/Ox composites. Due to high wear resistance and the favorable friction properties of ceramic matrix composites, they find applications as brakes and clutch-plates of passenger cars.

Both metal matrix and ceramic matrix composites are heavier and their production is prohibitively expensive. Higher weight of materials increases the overall weight of aircrafts and vehicles causing higher consumption of fuel. Heavy aircraft can carry less cargo. Therefore, manufacturers always search for high strength, light weight and less expensive materials. Synthesis of polymers by chemists during later half of eighteenth century opened up a new area of research in the field of material science.

In a polymer matrix composite, the polymer matrix provides the platform and supports the formation of the composite by holding the reinforcing component. Numerous synthetic polymers are being employed all over the globe for this purpose. When the reinforcing component (e.g., fiber) is added to the base matrix, it plays a vital role in binding the fibers and when the load is applied to the composite material it helps in transferring the load to the fibers. The matrix presents itself as a barrier in the event of adversity and to protect the fiber and surface from mechanical abrasion by trying to hold the fibers in place so as to prevent further abrasion and new surface cracks and damages. The credit of the matrix lies in its ability to deform easily under the applied stress, transfer the stress to the fibers thereby distributing the stress evenly throughout the entire portion of the composite. The two types of polymers used as matrices in the fabrication of composites are thermoplastics and thermosets. Thermoplastics are often formed by addition polymerization reaction leading to long linear chain polymers with no cross-links. Thermoplastics soften on heating readily and hence can be reshaped and reused. Thermoplastics are generally soft, less brittle and soluble in suitable solvents. Polystyrene (PS) is an important example of commercial thermoplastic. Other major examples are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polycarbonate (PC), polymethyl methacrylate (PMMA), styrene acrylonitrile (SAN), etc. Thermosets are formed, on the other hand, by condensation polymerization reaction leading to three dimensional network structures. Thermosets do not soften on heating and hence cannot be reshaped and reused. Thermosets are usually hard, strong, more brittle and insoluble in almost all organic solvents. Principal examples of thermosets include epoxy, phenol formaldehyde resin (Bakelite), melamine resin and unsaturated polyesters.

The early polymer matrix composites were prepared by using synthetic fibers. The first synthetic fiber used for reinforcement was glass fiber. Fiberglass is actually glass fiber reinforced plastic (GFRP) or simply glass reinforced plastic (GRP). Fiberglass is a strong lightweight material and is used for many products. Its bulk strength and weight are better than many metals, and it can be more readily molded into complex shapes. Applications of fiberglass include aircraft, boat hulls, automobiles, sports items, etc. The other synthetic fibers used for reinforcement are carbon and aramid. Some advanced composites are now made using carbon fibers instead of glass. These materials are lighter and stronger than fiberglass but more expensive to produce. They are used in aircraft structures. More than 20 % of Airbus A380 is made of composite materials, mainly plastic reinforced with carbon fibers.

Polymer composites were superior materials in terms of specific strength due to comparatively lower weight than the traditional metallic counterpart and attention has been paid always to synthesize newer composite materials which could meet the demand of the industry keeping in view the economic aspect. Despite having several superior properties, these materials are now facing challenges due to issues related with health hazard and biodegradability [1]. Synthetic fibers like glass and carbon can cause acute irritation of the skin and upper respiratory tract. Also it was suspected that the exposure to these fibers for longer period of time might cause lung scarring and cancer. These fibers are not easily degradable and may be responsible for serious environmental hazards. Thus the charm of using synthetic fibers in polymer composites started fading because they are expensive, non-biodegradable and pollute the environment.

The issues of health hazard, environmental pollution and high cost associated with synthetic fibers led the scientists and researchers to focus more on the application of natural fibers and plant based resources towards

the development of natural fiber reinforced polymer composites. Natural fiber reinforced polymer composites find increasing applications in many manufacturing sectors like vehicle parts, sports devices, electrical parts, and packing materials. The primary advantages of natural fibers over synthetic fibers have been their low cost, light weight, high specific strength, renewability, and biodegradability [2]. Depending upon the type of reinforcing fiber, composites are classified as (i) fiber composites, (ii) particle composites and (iii) laminate composites. In fiber composites, fibers of varying lengths are used for reinforcement. Moreover, fibers are laid in different orientations including random orientation. In particle composites, fibers are powdered and sieved, and then particles of different size are used for reinforcement. In laminate composites, fiber mats are prepared and these mats are laid layer by layer for reinforcement with, of course, the polymer matrix being filled in between the mat layers. Fibers may also be laid layer by layer without making prior mats.

Nature has abundant natural fibers and only a few of them have been harnessed for the purpose of reinforcement. Jute, hemp, kenaf, flax, sisal, coir, cotton, wood, and bamboo are the commonly used natural fibers for polymer reinforcement [3-9]. Besides these, several other lignocellulosic natural fibers such as curaua, henequen, olive husk, rice husk, pineapple, bagasse, banana, wheat straw, oil palm, abaca, aloe vera, and pine needle have also been employed to prepare polymer composites [10-16]. The progress of research on natural fiber–reinforced polymer composites is being reviewed continuously [17-26].

In this paper, our focus is on the use of natural fiber obtained from a plant whose scientific name is *Schumannianthus dichotomus* (Roxb.) Gagnep. (one of its vernacular names is murta) for reinforcement of polymer matrix. Murta plants are grown in Assam and West Bengal states of India and in north-eastern parts of Bangladesh. According to Wikipedia, murta plants are also found in Myanmar, Thailand, Cambodia, Vietnam, Malaysia and Philippines. The picture of murta plants is shown in Figure 1. Several handicraft items like mat (commonly known as sitalpati in local language), hand bag, hat, hand fan, etc. are produced from murta fiber thereby providing livelihood to many common people in India and Bangladesh. The details of murta plant and murta fiber based small-scale enterprises have been documented [27-29]. Recently, we prepared polymer composites reinforced by murta fibers (both core and bast) and reported their characteristics [30-32].

Figure 1: Murta plants (figure copied from Wikipedia).

The reported studies [30-32] revealed that murta core and bast fibers can be profitably used for manufacturing natural fiber reinforced composites of non-hybrid (containing one type fiber only) and hybrid types. The polymer matrix used in the previous studies contains 2, 2-bis (4-glycidyloxyphenyl) propane (also known as bisphenol A diglycidylether or BADGE) and hardener *N*-(3-dimethyaminopropyl)-1,3-propylenediamine. Since the characteristics of a composite depends on the properties of both polymer matrix and the reinforcing fibers, an attempt has been made in this paper to prepare and analyze hybrid composites by changing the polymer matrix and using mixtures of murta bast fiber with different other natural fibers as reinforcing natural material. The polymer matrix is prepared from BADGE and the hardener known by the trade name PPA-7040. The different natural fibers mixed with murta fiber are jute fiber, luffa fiber, coconut fiber and betel nut fiber.

2. Materials and Methods:

Materials. The polymer matrix is prepared from the epoxy resin BADGE (density 1.15 g cm^{-3}) and the hardener PPA-7040 (Phenalkamine epoxy hardener (density 1.01 g cm^{-3})). The structures of BADGE and PPA – 7040 are shown in Figs. 2 and 3, respectively. The resin was procured from (TCI chemicals) and the hardener was procured from Paladin Paints and Chemicals, Pvt. Ltd. and used as supplied. Laboratory reagent grade NaOH (S. D. Fine Chemicals, India) was used for treating the fibers.

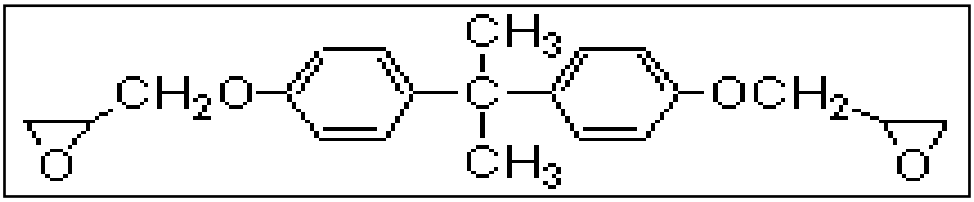

Figure 2: Structure of Bisphenol A diglycidyl ether (BADGE).

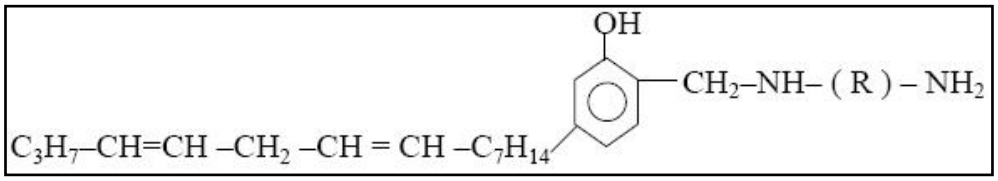

Figure 3: Structure of the hardener (PPA-7040).

Extraction and Processing of Fibers. The mature stems of the murta plants were collected and their outer portions are separated (Figure 4) and soaked in water for about one month which made the manual extraction of the fibers easier. The extracted fibers were then washed with water and then dried under the sun.

Figure 4: Outer part of murta stems.

Betal nuts or areca nuts are grown abundantly in India (coastal regions of Karnataka and Kerala, and in north-eastern states) and it belongs to the Areca catechu Linnaeus species. Sufficient quantity of ripe betel nut fruits was bought from local market. The outer covers of these fruits obtained after removing the betel nuts from the central portion are soaked in water for nearly 30 days. The retting process loosens the fibers and makes manual extraction of fibers easier. The extracted fibers were then washed with water and then dried under the sun.

The jute fibers were directly bought from local market. The fibers were washed thoroughly with water and then dried under sun.

Dried luffa fruits *(Luffa Cylindrica)* were also bought from local market. After removing the outer cover of these dry fruits sponge type luffa fibers were collected. These fibers were washed with water and then sun dried.

The above fibers were then boiled separately for two hours in 2 % (w/w) NaOH solution. The fibers, after removing from the NaOH solution, were washed with ordinary water and finally with distilled water. These fibers were sun dried first, and then kept in an oven at about 60 °C for 24 hours and finally at 100 °C for 2 hours. The fibers treated thus were cut into required lengths.

Fabrication of Hybrid Composites. All the composites were made by using hand lay-up method followed by compression molding. For the synthesis of hybrid composites the amount of fiber mixture was kept fixed at 30 weight %, 70 weight % being the polymer matrix. In the fiber mixture the weight ratio of the two types of fibers was also kept fixed at 50:50 and fibers of 30 mm length were used. The epoxy resin and hardener were taken in a beaker in 1:1 volume ratio and mixed thoroughly with the help of a thick glass rod. At first silicon oil was used in the inner side of the metallic mold measuring 140 mm x 140 mm x 5mm. Then one thin layer of polymer was applied on the bottom of the mold. On this polymer layer, the betel nut fibers were spread randomly and evenly. Above this fiber layer, another layer of polymer was placed. Now above this second layer of polymer, murta fibers were spread randomly and evenly. Above the murta fiber so arranged the third layer of resin was placed. Each of the fiber and resin layers was pressed with the help of a metallic roller to ensure the removal of air bubbles. Thus the hybrid composite prepared in this study contains total of three resin layers one at the top and other at the bottom including one layer in between the layers of betel nut and murta fibers. The mould was closed and kept under constant pressure for 24 hours at room temperature. Finally, the sheet was removed from the mold and kept in hot air oven for another 24 hours. The composites so prepared were cut into equal pieces of dimension 140 mm x 25 mm x 5mm for carrying out various measurements. In a similar fashion the other hybrid composites were also prepared. The sample codes used for the different samples are listed in Table 1.

Table 1: Description of the [30 weight % (fiber mixture) + 70 weight % polymer matrix] hybrid composite samples.

Sample Code	Description of fiber mixture
NP	Neat Polymer (0 % Fiber Mixture + 100 % Polymer Matrix)
MB	(15% Murta + 15% Betel Nut) Fibers
MC	(15% Murta + 15% Coconut) Fibers
MJ	(15% Murta + 15% Jute) Fibers
ML	(15% Murta + 15% luffa) fibers

Measurements. The densities of the polymer, fiber and the composites were measured at room temperature (25 ± 0.5 ^{o}C) by using the Archimedes' principle. For water absorption test of the polymer and composites, the samples of dimension 75 mm x 25 mm x 5 mm were prepared and then kept in an oven for 24 hours by maintaining the temperature at 80 $\pm$ 3 ºC. The samples were then cooled in desiccator and immediately their weights were noted. Thereafter the samples were immersed in distilled water for 24 hours. The samples were removed from water one at a time and the surface water was wiped off with a dry cloth and the weights were again noted. From the increase in weight of the soaked sample the % of water absorption was then calculated.

Thermo gravimetric analysis (TGA) was done by using Perkin Elmer STA 6000 Simultaneous Thermal Analyzer. The sample was taken in alumina crucible and the analysis was carried out under nitrogen atmosphere (maintained with a continuous flow rate of 100.0 mL min^{-1}) at a heating rate of 10 ºC min^{-1}.

Tensile, flexural and compressive strengths of the samples were measured in a Universal Testing Machine (INSTRON Model 8801). The impact strengths of the samples were measured in a Drop Weight Impact Testing Machine (SPRANKTRONICS) with a drop height of 0.5 meter and drop weight of 3.072 kg having an impact velocity of 3.13m s^{-1}.

The Rockwell hardness of the samples were measured in HRL scale in a Digital Rockwell Hardness Tester (FIE Model RASNE-1) by using 1/4 inch steel ball indentor and 60 kg load. The measurements of strengths and

hardness were made in the Mechanical Engineering Department of Indian Institute of Technology, Guwahati, India at room temperature (25 ± 1 °C). Sample specimens for the different measurements were prepared according to the ASTM standards.

3. Results and Discussion:

For the sake of comparison, some of the properties of the fibers used here for making hybrid composites are collected from the literature [33-35] and listed in Table 2. The pictures of the neat polymer and hybrid composites are shown in Figure 5.

Table 2: Properties of the fibers used for reinforcement.

Properties	Fiber				
	Betel nut[33]	Coconut[34]	Luffa[35]	Murta[32]	Jute[34]
Density (g cm^{-3})	1.05-1.25	1.2 - 1.4	1.4	1.10	1.3 - 1.5
Cellulose (%)	53.2	32.1 - 34.5	60	56	60 - 70
Hemi Cellulose (%)	33	1.3 - 4.1	22	19	12 - 20
Lignin (%)	7.2	46.0 - 47.1	10.6	16	11 - 15
Diameter (mm)	0.05-0.10	0.19 - 0.21	0.27- 0.40	0.135-0.153	0.017 - 0.020
Tensile Strength (MPa)	183	145	375	365 - 391	400 - 800

Figure 5: Pictures of the neat polymer and hybrid composites. N_o, O_1, O_2, O_3, and O_4 samples refer to the sample codes NP, MB, MC, MJ and ML, respectively.

Table 3: Properties of the hybrid composites.

Sample	Density (g cm^{-3})	Tensile Strength (MPa)	Flexural Strength (MPa)	Compres-sive Strength (MPa)	Impact Strength (J m^{-1})	Rockwell Hardness Number (HRL)	Tensile Modulus (GPa)
NP	1.10	48.9	103.1	72.7	49.5	42.5	1.6
ML	1.07	83.4	148.8	99.5	62.9	83.5	5.4
MC	1.05	89.9	129.2	79.1	54.2	72.6	9.4
MB	1.09	90.5	147.1	109.7	80.7	79.8	7.4
MJ	1.04	110.3	168.1	108.3	97.3	88.4	6.9

The experimental values of the density of the hybrid composites are given in Table 3. All the four hybrid composites have density less than that of the neat polymer indicating presence of voids in the composite. The water absorption features of the hybrid composites are shown in Figure 6. Out of the four hybrid composites studied here, MJ absorbs least weight % of water and ML absorbs highest weight % of water. No correlation could be drawn between the water absorbing capacity of the composite and the cellulose / hemi cellulose / lignin contents of the fibers.

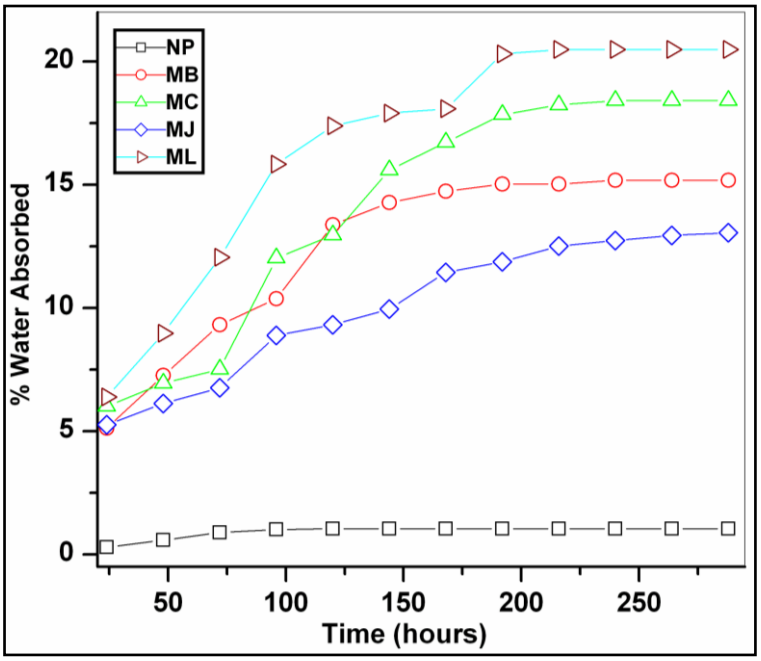

Figure 6: Water absorbed by the neat polymer and the hybrid composites at different time intervals.

The thermograms of the composites are shown in Figure 7 and all the four hybrid composites have thermal stability up to about 245 ºC. From Figure 7 it is evident that all the composites have similar thermal behaviour. Thermal stability of hybrid composites is lower than the composites reinforced by single type fiber. In the case of hybrid composites the fiber appears to control the thermal stability of composite, whereas in the case of non-hybrid composites the polymer matrix appears to control the thermal stability of the composite [30-32]. This inference however needs to be examined further by studying more number of hybrid systems.

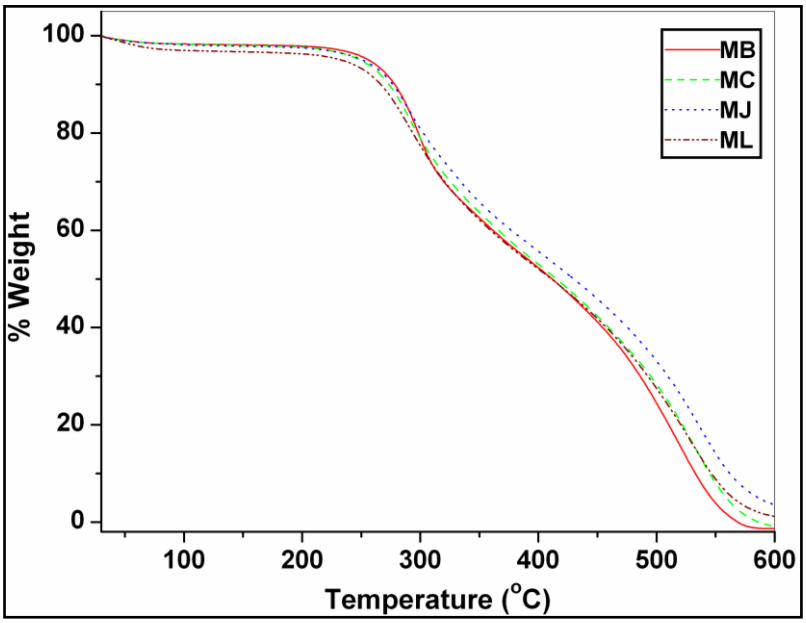

Figure 7: Thermal gravimetric analysis curves of the hybrid composites.

The tensile strength (TS), flexural strength (FS), compressive strength (CS), impact strength (IS) and hardness of the hybrid composites are given in Table 3 and Figs. 8 and 9. Among the four hybrid composites studied here, the composite MJ containing murta and jute fibers has the highest values for strengths and hardness. From the tensile stress versus tensile strain plots shown in Figure 10, it is also clear that MJ hybrid has the highest

toughness. Better homogeneity between murta and jute fibers, optimum fiber-matrix adhesion and least water absorption are probably responsible for the optimum strength, hardness and toughness of MJ hybrid composite.

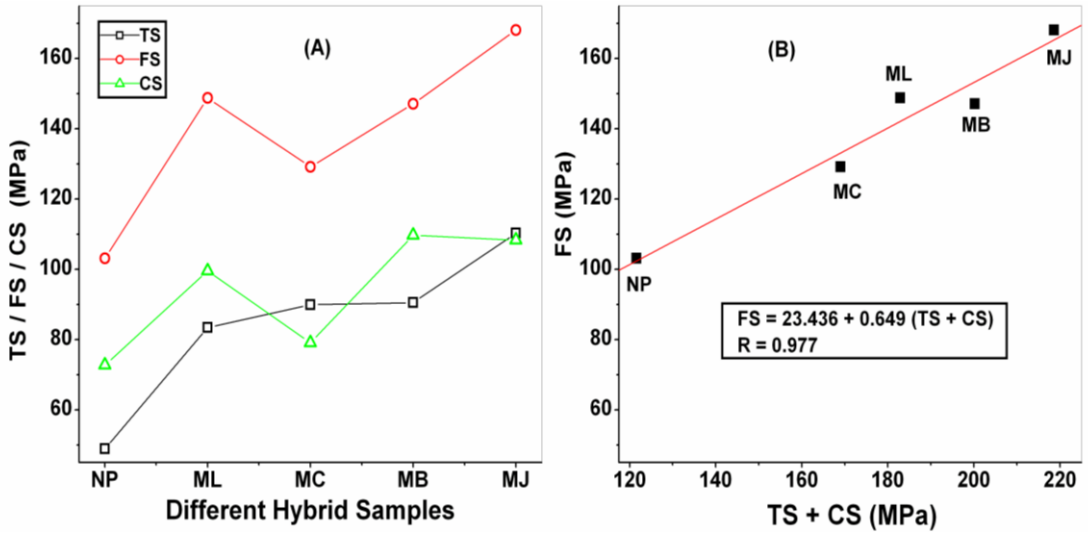

Figure 8: (A) Tensile (TS), flexural (FS) and compressive (CS) strengths of the neat polymer and hybrid composites, and (B) correlation between flexural, tensile and compressive strengths.

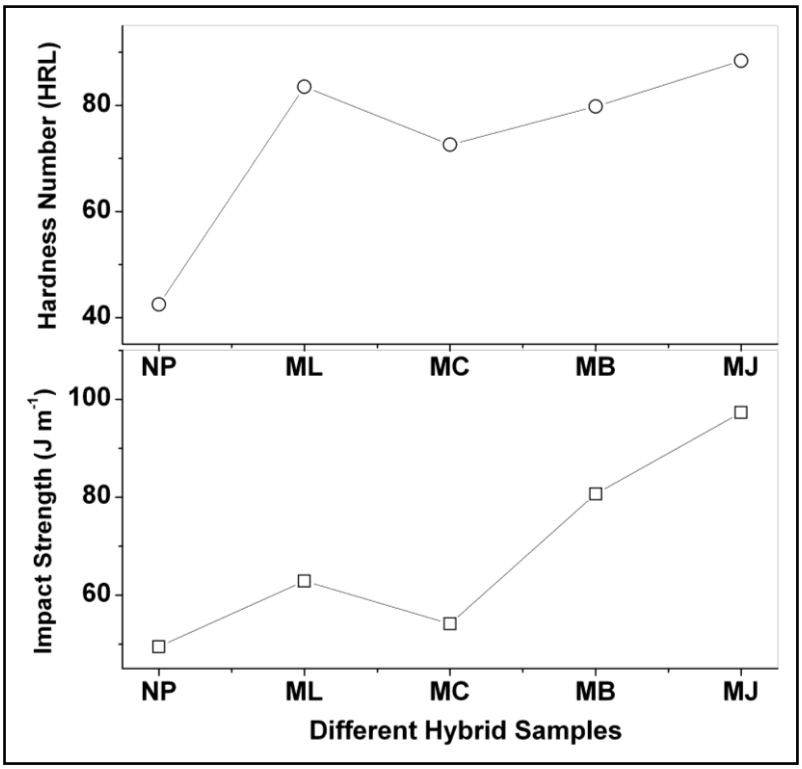

Figure 9: Impact strength and hardness of the neat polymer and hybrid composites.

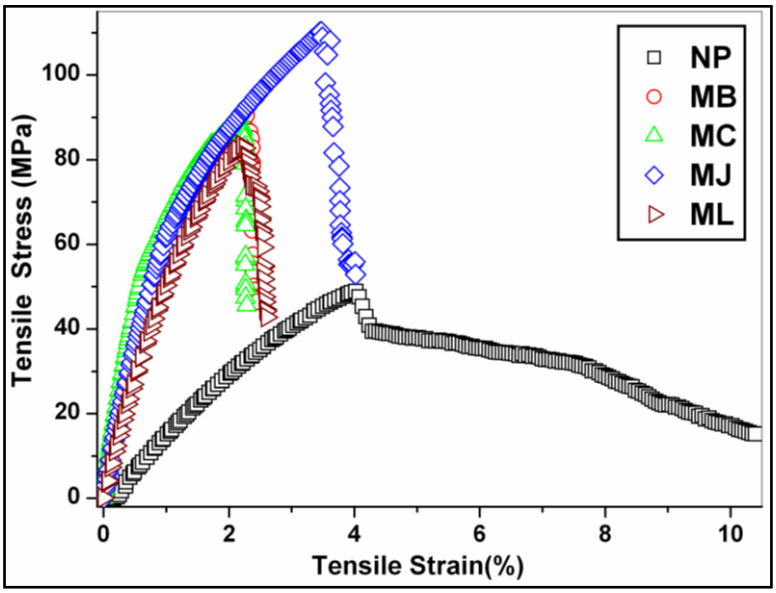

Figure 10: Tensile strain versus % strain plots for the neat polymer and the hybrid composites.

FS and sum of TS and CS of the neat polymer and the hybrid composites of different fiber mixtures with fixed total weight % of fibers (30 %) have been correlated through an empirical correlation of the type

$$FS = A + B(TS + CS) \quad (1)$$

where A and B are the intercept and slope, respectively. It is interesting to see from Figure 8B that a fairly good linear correlation exists between FS and TS + CS irrespective of different types of fiber mixtures. A material during bending experiences internally compressive as well as tensional stresses and this may be responsible for the correlation between FS, TS and CS shown by Eq. (1).

4. Conclusions:

Murta + jute form the best mixture for making hybrid composite as this composite possesses superior physical and mechanical properties compared to hybrid composites containing murta + luffa, murta + coconut and murta + betel nut fibers. The amount of water absorbed by a hybrid composite does not appear to have any correlation with the chemical composition of the fibers used for reinforcement. In hybrid composites, fibers appear to control the thermal stability of the composites, and this observation however needs to be examined further by studying more such type of hybrid composites.

Acknowledgment:

The authors are thankful to the Head, Department of Mechanical Engineering, Indian Institute of Technology, Guwahati for providing instrumental facilities to test the composite samples.

References:

[1] P. Wambua, J. Ivens and I. Verpoest, *Compos. Sci. Technol*., 63 (2003) 1259.
[2] A. K. Mohanty, M. Misra and L. T. Drzal, *J. Polymer. Environ*., 10 (2002) 19.
[3] W. Nuthong, P. Uawongsuwan, W. Pivsa-Arta and H. Hamada, *Energ. Proced*., 34 (2013) 839.
[4] R. Naveenkumar, V. Sharun, B. Dhanasakkaravarthi and I. P. T. Rajakumar, *Int. J. Appl. Sci. Eng. Res*., 4 (2015) 250.
[5] W. L. Ngo, M. M. Pang, L. C. Yong and K. Y. Tshai, *Adv. Environ. Biol*., 8 (2014) 2742.
[6] M. Ramesh, K. Palanikumar and K. H. Reddy, *J. Appl. Polym. Sci*., 133 (2016), DOI: 10.1002/APP.42968.
[7] S. B. Koradiya, J. P. Patel and P. H. Parsania, *Polym.-Plast. Technol. Eng*., 49 (2010) 1445.
[8] K. Begum, M. A. Islam and M. M. Huque, *J. Sci. Res*., 7 (2015) 97.
[9] G. Kalaprasad, K. Joseph, S. Thomas and C. Pavithran, *J. Mater.Sci*. 32 (1997) 4261.
[10] K. O. Reddy, C. U. Maheswari, K. R. Reddy, M. Shukla, E. Muzenda and A. V. Rajulu, *Int. J. Polym. Anal. Charact*., 20 (2015) 612.

[11] G. Xia, K. O. Reddy, C. U. Maheswari, J. Jayaramudu, J. Zhang, J. Zhang and A. V. Rajulu, *Int. J. Polym. Anal. Charact*. 20 (2015) 377.
[12] M. Shibata, K.-I. Takachiyo, K. Ozawa, R. Yosomiya and H. Takeishi, *J. Appl. Polym. Sci*., 85 (2002) 129.
[13] M. F. V. Marques, J. N. Lunz, V. O. Aguiar, I. Grafova, M. Kemell, F. Visentin, A. Sartori and A. Grafov, 23 (2015) 251.
[14] S. N. Monteiro, V. Calado, R. J. S. Rodriguez and F. M. Margem, *J. Mater. Res. Tecnol.*, 1 (2012) 117.
[15] A S. J. Sekaran, K. P. Kumar and K. Pitchandi, *Bull. Mater. Sci*., 38 (2015) 1183.
[16] S. B. Hazarika, S. U. Choudhury, S. S. Panja, S. K. Dolui and B. C. Ray, *J. Sci. Indust. Res.*, 74 (2015) 589.
[17] V. K. Thakur and M. K. Thakur, *Carbohyd. Polym*., 109 (2014) 102.
[18] M. Jawaid and H. P. S. A. Khalil, *Carbohyd. Polym.*, 86 (2011) 1.
[19] K. P. Ashik and R. S. Sharma, *J. Miner. Mater. Charact. Eng*., 3 (2015) 420.
[20] Y. R. Loh, D. Sujan, M. E. Rahman and C. A. Das, *Resour. Conser. Recyc*., 75 (2013) 14.
[21] C. W. Nguong, S. N. B. Lee and D. Sujan, 2013. *Int. J. Chem. Mol. Nucl. Mater. Metall. Eng*., 7 (2013) 33.
[22] H. Ku, H. Wang, N. Pattarachaiyakoop and M. Trada, *Compos. B*, 42 (2011) 856.
[23] D. N. Saheb and J. P. Jog, *Adv. Polym. Technol.*, 18 (1999) 351.
[24] A. G. Facca, M. T. Kortschot and N. Yan, *Compos. Sci. Technol*., 67 (2007) 2454.
[25] K. Begum and M. A. Islam, *Res. J. Eng. Sci*., 2 (2013) 46.
[26] L. Mohammed, M. N. M. Ansari, G. Pua, M. Jawaid and M. S. Islam, *Int. J. Polym. Sci.*, 2015: Article ID 243947, dx.doi.org/10.1155/2015/243947.
[27] D. Chowdhury and B. K. Konwar, *Curr. Sci*., 91 (2006) 648.
[28] R. N. Mandal, R. Bar and P. P. Chakrabarti, *Indian J. Nat. Prod. Resour*., 5 (2014) 365.
[29] R. Ahmed, A. N. M. F. Islam, M. Rahman and M. A. Halim, *Int. J. Biodiv. Sci. Manag.*, 3 (2007) 252.
[30] A. H. Barbhuiya and K. Ismail. *Int. J. Polym. Anal. Charact.*, 21 (2016) 221.
[31] A. H. Barbhuiya and K. Ismail. *Int. J. Polym. Anal. Charact.*, 21 (2016) 563.
[32] A. H. Barbhuiya, S. U. Choudhury and K. Ismail, *J. Appl. Polym. Sci*., 133 (2016) DOI: 10.1002/APP.44142.
[33] S. B. Hazarika, S.U Choudhury, S. K. Saw, G. Sarkhel and A. Choudhury, *J. Appl. Polym. Sci*., 125 (2012) 3038.
[34] S. K. Saw, G. Sarkhel and A. Choudhury, *J. Appl. Polym. Sci*., 125 (2012) 3038.
[35] Y. Laidani, S. Hanini, G. Mortha and G. Haninia, *Iran. J. Chem. Chem. Engg.*, 31 (2012) 119.