Mechanical Properties of Phenol Formaldehyde Hybrid Composites Reinforced with Natural Cellulose Fibers

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This paper reports on the preparation and mechanical properties of hybrid polymer composites involving areca fine fibers (AFFs), sisal fibers (SFs), and roselle fibers (RFs) as reinforcing agents in a phenol formaldehyde (PF) resin-based polymer matrix. For comparative study, an AFF/glass fiber-reinforced PF hybrid composite and AFF/PF composite were also prepared. Hybrid composites were fabricated using a hand lay-up technique, where the weight fraction of fibers was kept at 40 wt% at a ratio of 1:1. Tensile and flexural properties of randomly oriented intimately mixed hybrid polymer composites were evaluated. The results revealed that the mechanical properties of the AFF/PF composite increased by a considerable amount when hybridized with the sisal fibers. Scanning electron microscopy (SEM) was used to analyze the fractured surface of the composite specimens after mechanical testing.

Keywords: Natural cellulose fibers; Hybrid composites; Mechanical properties; SEM

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INTRODUCTION

In recent years, the use of natural cellulose fiber-reinforced polymer composites has increased incessantly in various engineering applications. This can be attributed to their low density, low cost, environmental friendliness, highly specific mechanical properties (strength and stiffness), and ease of processing (Kurniawan*et al*. 2013; Mahjoub*et al*. 2014). Natural cellulose fibers obtained from various natural plants, such as flax, sisal, cotton, hemp, jute, kenaf, pineapple, ramie, bamboo, banana, roselle, and wood, are often applied as a potential reinforcement for polymer composites. The poor mechanical properties and certain manufacturing issues limit the use of natural cellulose fiber materials for non-structural and semi-structural applications. The hybridization of natural cellulose fiber with other natural cellulose fibers, synthetic fibers, or particulates (natural or synthetic) provides a method to improve the mechanical properties over natural cellulose fiber alone. Material researchers have done much research on natural cellulose fiber-reinforced polymer hybrid composites:jute/bagasse/epoxy (Saw and Datta 2009); jute/ oil palm /epoxy (Jawaid *et al.* 2010); kenaf/oil palm/poly(lactic acid) (Birnin-Yauri *et al*. 2016); jute/bamboo/low-density polyethylene (Liew *et al.* 2016). They are reported the mechanical, thermo-mechanical, dimensional stability, and morphological properties of the hybrid composites.

Among all the natural reinforcing fibers, areca, sisal, and roselle fibers are important fibers used in the polymer industry recently. Areca (*Areca catechu* L., family

Arecaceae) fibers are also recognized as a promising fiber material like the other natural fibers due to their low cost and high availability. It is a very high potential perennial crop. The areca tree is grown in India for use in medicine, Gutka, chocolate, paint, *etc*. Areca husk is abundantly available (6 lakh tonnes) as bio-waste materials in the South West-India (Sanjay *et al.* 2016). Roselle (*Hibiscus sabdariffa* L., family Malvaceae) fibers are one of the plant-based natural fibers used as a reinforcing agent in polymer matrix composites. These fibers have been found to be an important source of fiber and for other commercial applications in recent years. Therefore, they are cultivated for the production of fibers from the stem. Roselle also has been used as a diuretic and mild laxative in folk medicine, food colorings, syrup, and spicy version of spinach. Sisal (*Agave sisalana* Perr., under the family Agavaceae) fibers are a hard fiber extracted from the leaves of the sisal plant and traditionally used in making twine and rope. These fibers are used as a binding material, and are also used in the structural industry to reinforce plaster in ceilings and walls. Sisal fibers have been utilized as a reinforcing agent to replace synthetic fibers in polymer composites (Athijayamani *et al.* 2009). Thermoset polymers are directly responsible for the high physical and mechanical properties (high strength to support high stress or load, temperature, *etc*.) compared with thermoplastics polymers due to this highly cross linked structure produced by chemical bonds. This highly crosslinked structure provides a poor elasticity or elongation.

The present experimental study aims at obtaining the mechanical behaviors of hybrid natural cellulose fiber-reinforced polymer composites. Three different types of natural cellulose fibers were utilized to form the hybrid polymer composites. Roselle fiber (bast) and sisal fiber (leaf) were hybridized with areca fine fibers (fruit) in a phenol formaldehyde hybrid composite.

All types of composites were fabricated using a hand lay-up technique where the weight fraction of fiber was kept at 40 wt%. The fibers were hybridized at a ratio of 1:1. For the comparative study, an additional hybrid composite was prepared with areca fine fiber (AFF)/glass fiber at a ratio of 1:1. The mechanical properties of the hybrid composites were evaluated and also compared with 40 wt% AFF/phenol formaldehyde (PF) composite. The fracture surface of composite specimens was examined by scanning electron microscopy (SEM) to discover the mode of failure.

EXPERIMENTAL

Materials

The areca fine fibers were purchased from the Alphonse Fiber industry in Nagercoil, Tamilnadu, India. Roselle and sisal fibers were purchased from Global Agro Products in Coimbatore, Tamilnadu, India. General-purpose glass fibers (E-glass) in non-woven form are also used to prepare the hybrid composite. All fibers were used as reinforcement in the condition in which they were received.

The properties of Areca fine fiber, roselle fiber, sisal fiber, E-glass fiber are given in Table 1. A resole-type phenol formaldehyde resin (Windson Chemical Private Limited, Gujarat, India) was used as the resin matrix with a cross-linking agent (divinylbenzene) and acidic catalyst (hydrochloric acid). The typical properties of phenol formaldehyde resin are presented in Table 2.

Table 1. Properties of the Fibers Used in this Study

Properties	Areca	Roselle	Sisal	E-glass
Diameter (mm)	0.285-0.89	0.13-0.24	0.21-0.29	0.003-0.020
Length (m)	0.18-0.40	1-2	0.5-1	-
Density (g/cm^3)	1.05-1.25	1.45	1.51	2.54
Ultimate stress (MPa)	89.5-118.67	147-184	80-164	1950-2050
Elongation at break (%)	11-12.5	5-8	11-15	4.5-4.9

Table 2. Typical Properties of Phenol Formaldehyde Resin

Properties	
Specific gravity	1.12-1.16
Polar surface area (Å^2)	9.23
Flash point (°C)	72.5
Boiling point (°C)	181.8
Composition	Carbon-Carbon
Elongation at break (%)	2
Density (g/cm^3)	1.3

Preparation of Composites

Composites were prepared using a hand lay-up technique. Prior to process, a releasing agent (polyvinyl alcohol) was applied on the surface of the mould box. A mechanical stirrer mixed the fibers and PF resin for 30 min to obtain homogeneity. Then, the cross-linking agent and an acidic catalyst were added to the resin mixture and stirred mechanically for 15 min. Next, the mold box was allowed to cure at room temperature for 48 h. Composite plates were prepared with size of 150 × 150 × 3 mm and cut using an electrical, wood board cutter to get the composite specimens. For each combination, five specimens were tested, and the average values were recorded.

Mechanical Testing

The tensile tests were carried out according to ASTM D638-10 (2010) at a crosshead speed of 2 mm/min on a universal testing machine (make: Fuel Instruments & Engineers Pvt. Ltd.). The flexural tests were conducted on the same universal testing machine in accordance with ASTM D790-10 (2010) at a crosshead speed of 2 mm/min.

SEM

Scanning electron micrographs of the fractured surface of the composite specimens were acquired using an SEM (model: HITACHI S-3000N) to study the morphology.

RESULTS AND DISCUSSION

Tensile Properties

Figures 1a, b, c, and d show the tensile stress – strain curve of the hybrid composites (AFF/PF, AFF/RF/PF, AFF/SF/PF, and AFF/GF/PF). From these figures, it can be observed that all the curves were almost linear. This is depending upon the extension of the hybrid composite systems. The force and extension curves of composites are presented in Fig.2. It is clear from Fig.2a, b, c, and d that the specimens of the AFF/SF/PF hybrid composite could tolerate a load of 436.6 N at an extension of 1.9 mm; the specimens of the AFF/GF/PF hybrid composite could tolerate a load of 362.5 N at an extension of 2.2 mm; the specimens of the AFF/RF/PF hybrid composite could tolerate a load of 333.5 N at an extension of 2.5 mm; and the specimens of the AFF/PF composite could tolerate a load of 325.7 N at an extension of 2.6 mm.

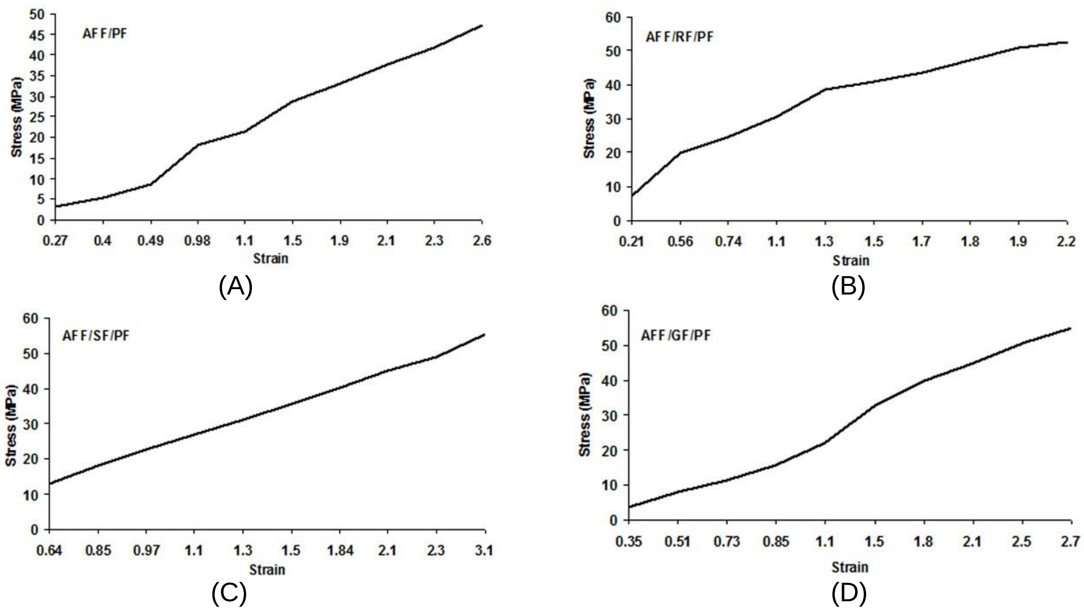

Fig. 1. Stress and strain curves of tensile property of composites

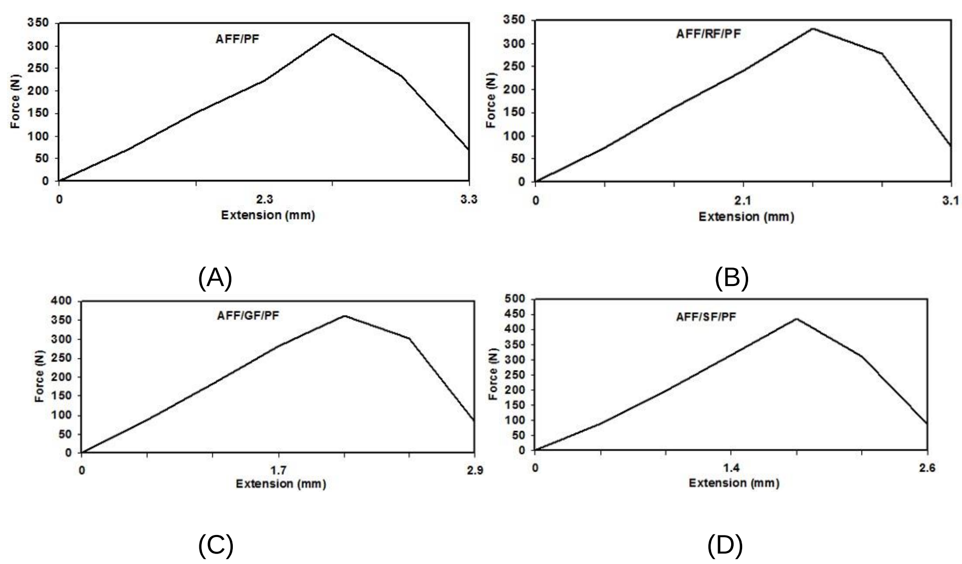

Fig. 2. Force and extension curves of tensile property of composites

The variation of the tensile properties of the composites is presented in Fig.3. It can be observed that the AFF/SF/PF hybrid composites showed the highest tensile strength, followed by the AFF/GF/PF and AFF/RF/PF composites. The lowest tensile strength was observed for the AFF/PF composite. It is evident that with the hybridization of sisal, roselle, and glass fibers, there was an increase in the tensile strength and tensile modulus of AFF/PF composites, and the value was highest for the AFF/SF/PF fiber hybrid composite. When compared with the AFF/PF composite, the AFF/SF/PF hybrid composite showed a 17.72% increase in tensile strength and a 17.66% increase in tensile modulus. When compared with the AFF/GF/PF hybrid composite, the AFF/SF/PF hybrid composite had a 1.64% increase in tensile strength and a 1.75% increase in tensile modulus. Though there were some minor differences in test results due to the hybridization of the other fibers, the tensile property values obtained were generally acceptable when compared with previous work (Athijayamani *et al.* 2009).

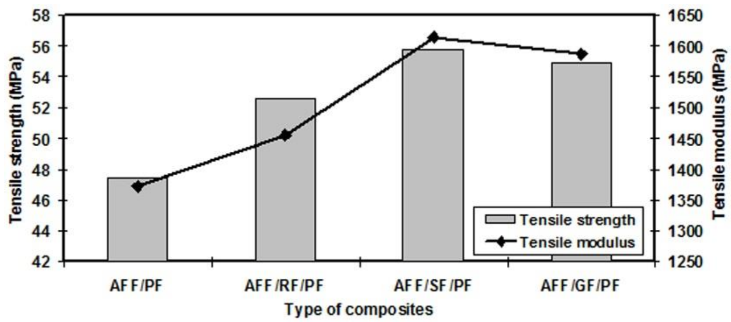

Fig. 3. Variation of the tensile properties of composites

Flexural Properties

The flexural properties of the AFF/PF composite were found to increase when hybridized with sisal, roselle, and glass fibers, and with the hybridization of sisal fibers, flexural strength gradually increased to a greater extent than in the AFF/GF/PF and AFF/RF/PF composites (Fig. 4a, b, c, and d). It is clear that specimens of the AFF/SF/PF hybrid composite could tolerate a load of 536.5 N with a deflection of 2.1 mm; the AFF/RF/PF hybrid composite specimens could tolerate a load of 435.5 N with a deflection of 2.6 mm; the AFF/GF/PF hybrid composite specimens could abide a load of 462.5 N with a deflection of 2.4 mm; and the AFF/PF composite could tolerate a load of 427.5 N with a deflection of 2.9 mm.

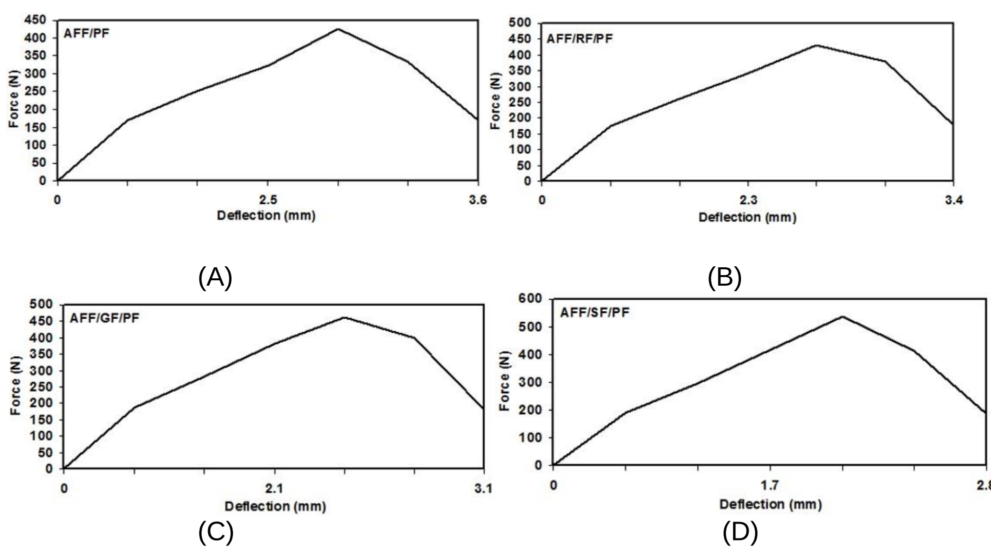

Fig. 4. Force and deflection curves of the flexural property of composites

The flexural properties of hybrid composites are shown in Fig. 5. It can be observed that with the hybridization of sisal, roselle, and glass fibers, there was an increase in the flexural strength and flexural modulus, which was highest for the AFF/SF/PF hybrid composite. When compared with the AFF/GF/PF composites, the AFF/SF/PF composite had a 2.47% increase in flexural strength and a 4.35% increase in flexural modulus. When compared with the AFF/PF composite, the AFF/SF/PF hybrid composite showed an improvement of 24.1% in flexural strength and 19.72% in flexural modulus. This increase in tensile and flexural properties of the AFF/SF/PF hybrid composites could be due to the increased interface bonding between the fibers and the matrix. The chemical reaction between the two natural fibers can change the fiber-to-fiber bonding. Therefore, the chemical reaction weakened the fiber-to-fiber bonds in the hybrid composites, but it strengthened the fiber-to-fiber bonds of the sisal fibers within the hybrid composites. The probable reason for this is that the polymer resin was easily impregnated into the surfaces of the sisal fibers because there were a large number of small voids that increased the compatibility and adhesion between the fiber and the matrix, which resulted in better mechanical properties. Generally, the natural cellulose fiber hybrid composite displays interesting properties that are not found in natural cellulose fiber-reinforced composites. The increase in mechanical properties of the AFF/SF/PF hybrid composites may have been due to the better mechanical interlocking and attractive forces between the fibers and the matrix. It is suitable to say that the AFF/SF/PF (1:1) hybrid composite was the best of the hybrid composites at 40%. When compared with the previous work (Athijayamani *et al.* 2009), the flexural property values are also acceptable. Hybridizing SFs with AFFs can produce a hybrid composite with better mechanical properties as compared with a single type of fiber reinforcement in a PF matrix.

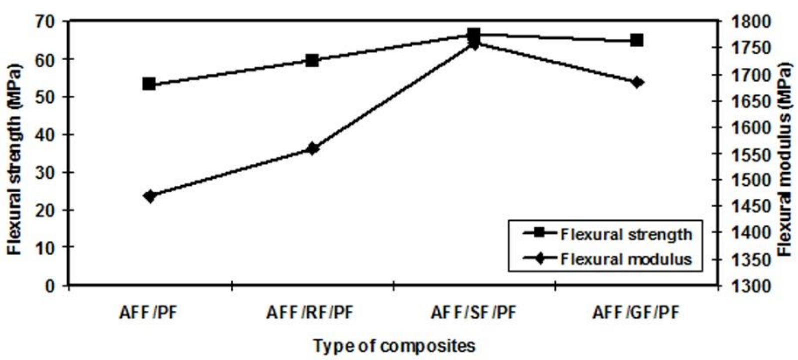

Fig. 5. Variation of the flexural properties of composites

Morphological Studies

Figures 6a and 6b show the SEM micrographs of the AFF/PF composite and the AFF/RF/PF hybrid composite, respectively. From Fig. 6a, it is clearly shown that the adhesion of the AFF fiber with polymer matrix in the interfacial area was poor. The mark out of fiber pullout and the hole left after the fiber pulled out because of poor impregnation are shown in Fig. 5a. The void spaces between the AFF fiber and the PF matrix are also shown, which indicate poor impregnation of fiber in the polymer matrix. This poor impregnation allowed the fiber to be pulled out, which was responsible for the lower strength of AFF/PF composite. Figure 6b shows an SEM micrograph of the AFF/RF/PF hybrid composite after the tensile test. It can be seen from Fig. 5b that the RFs were loosely embedded in the PF resin and that micro spaces exist at the interfacial province between the RFs and the PF matrix, which resulted in weak interconnection between RFs and the PF matrix. The void on the fractured surface was also more pronounced and gave weak bonding strength between the fibers and the matrix. The weak interfacial bonding strength reduced the strength of the composite.

From the above results, it can be seen that the AFF/SF hybrid composite showed overall better tensile and flexural properties as compared with the other hybrid composites. The mechanical interlocking between the fibers and the matrix appeared to be better, which could be attributed to the chemical bonding that resulted from optimum hybridization and occurred between the fibers and the matrix (Fig. 6c). It is possible that chemical reactions occurred between the chemical compounds of the AFFs and SFs. Therefore, it is possible to have a positive outcome through the bonding between fibers and the matrix (Fig. 6d), which would further lead to the improvement in mechanical properties in the hybrid composites.

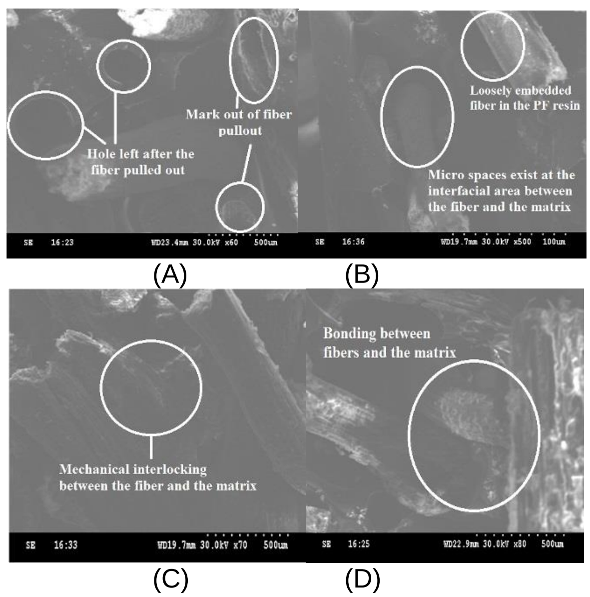

Fig. 6. SEM micrograph of a fractured surface of (A) the AFF/PF composite, (B) the AFF/RF/PF hybrid composite, and (C and D) the AFF/SF/PF hybrid composite after the tensile test

The SEM study clearly suggested that the addition of SFs into AFF/PF composites reduced fiber pull out as well as fiber protrusion from the surface of the composite specimens. Therefore, this type of hybrid composite may be used in high-end applications with a further addition of chemical agents.

CONCLUSIONS

1. The AFF/SF/PF hybrid composites showed the highest tensile strength, followed by the AFF/GF/PF and AFF/RF/PF composites.
2. The hybridization of sisal, roselle, and glass fibers with the AFFs increased the flexural strength and flexural modulus of the composites, which was highest for the AFF/SF/PF hybrid composite.
3. Hybrid composite AFF/SF (1:1) with 60 wt% of PF showed the best mechanical properties, followed by the AFF/GF/PF hybrid composite.

4. The hybrid composite prepared using AFFs (fruit fiber) and SFs (leaf fiber) managed to perform well without any additional process and additives.
5. The scanning electron microscopy analysis revealed that SFs strongly improved the adhesion and compatibility in the polymer matrix containing the AFFs.

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