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PROPERTIES OF COCONUT FIBRES REINFORCED CASHEW NUT SHELL RESIN

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ABSTRACT: Cashew Nut Shell Resin (CNSR) was reinforced with chemically modified coconut fibres. Modified fibres were treated with alkali solutions of NaOH (10%) and bleached with sodium hypochlorite. The bio-composites were fabricated using hand lay-up technique. The tensile tests showed that the mechanical properties (modulus, strength, and elongation at break for the tensile, compressive and bending) of composite improved significantly when compared with the fibre. The tensile strength improved by more than 200% while the Young modulus increased by over 305% in comparison with the fibre. Strain at break decreased by 61.76%. The compressive properties show that the strength increases by 169% while the Young Modulus increases by 53.05% and the compressive strain at break increases by 122.86%. SEM failure mode of all testing were observed to the same. Tensile failure was brittle in all nature with fibre pull-out in the same direction which is an indication of low speed failure. Specimens failed equally by matrix shear failure with constant debonding

KEYWORDS: Natural fibres; natural resin; cashew nut shell resin; coconut fibres, mechanical properties

INTRODUCTION

Natural fibres are those obtained directly from nature, usually from plants or animals. The common natural fibres are cotton, wool, silk, jute, banana stem, ramie, hemp, wood, bamboo, sisal, pineapple etc. Due to it renewability, biodegradability and environmental friendliness, properties of natural fibres as reinforcement have received tremendous attention. The reason for the attention are low cost, low energy content and recyclability, high strength to weight ratio, resistance to breakage during processing among others. In modern day, natural fibre is extensively used in the building industry. Research into natural fibres is as a result of environmental effect on the society due to pollution generated during the production and recycling of synthetic fibres. Properties of natural fibre based composite can be easily modified by the processing method (compression moulding, injection, extrusion moulding, and lay-up), fibre-matrix ratio, aspect ratio (L/D), and chemical modification. Irrespective of differences or similarities in the aspect ratio and volume fraction of fibres, the usage of more than two fibres can be applied.

Composites from natural fibre can be cost effective especially in construction industry; because of this it has received much attention from materials scientist over the past decades. This attention is due to its advantages over glass fibres. Other advantages alongside low cost are high resistance to corrosion, availability in large quantity, low density less abrasion to equipment and are not harmful to the human body [1,2]. The tensile performance of natural fibres has been investigated by numerous authors. The results coming out shows a large discrepancy of reported values for tensile strength and

Young's modulus [3-11]. Bearing in mind the variable and irregular cross-sections of natural fibres, their measurement can lead to enormous inaccuracies in the computation of stress. Some of the factors that can lead to discrepancies in the computation of the stress and Young modulus include, strain rate, gauge length, gripping, chemical treatment, the conditions of the fibre prior to characterisation, resolution of load cell and actuator precision and machine compliance among others.

Mechanical performance of sisal fibres at different gauge length (10 to 40mm with strain rate of 0.1 mm/min) was evaluated by Silva et al. [12]. he strain-to-failure decreased from 5.2 to 2.6% when the gauge length was increased from 10 to 40 mm. With an average tensile strength of 400 MPa it was observed that it is independent of the gauge length. While the average Young modulus was found to be 19 GPa, that of Weibull decreased from 4.6 to 3.0 when the gauge length was increased from 10 to 40mm.

Properties of alkalized and untreated coconut fibres with gauge lengths of 20 and 40 mm as reinforcement in cementitious composites were investigated by Li et al. [13]. Mortar made of cement, sand, water and super plasticizer at a ratio of 1:3:0.43:0.01 by weight respectively were mixed at a constant speed of 30 rpm. Coconut fibres were slowly added into the running mixer. The ensuing mortar was lighter than the conventional mortar, had higher ductility of up to1740% increase, an increase of up to 12% flexural strength and a higher energy absorption ability of up to 1680% increase.

Paramasivam et al. [14] carried out a research on coconut fibre reinforced corrugated slabs of 915 mm x 460 mm x 10 mm for low cost housing. A ratio of 1:0.5 cement-sand and 0.35 for water-cement ratio was used. Using third point loading, flexural strength of 22 MPa for volume fraction of 3%, fibre length of 2.5 cm and casting pressure of 0.15 MPa was adjoined to be the best. The thermal conductivity and absorption coefficient for low frequency sound were comparable with those of asbestos boards.

Slabs impact resistance falling with a weight of 0.475 kg from a height of 200 mm have been investigated by Ramakrishna and Sundararajan [15]. The slabs with dimension of 300 mm x 300 mm x 20 mm consist of 1:3 cement–sand mortar reinforced with coconut, sisal, jute and hibiscus cannabinus fibres. The slab with different fibre contents of 0.5%, 1.0%, 1.5% and 2% by weight of cement has three different fibre lengths of 20, 30 and 40 mm. Irrespective of the gauge length at a fibre weight of 2%, coconut fibres exhibited the best performance by absorbing 253.5, 231.14 and 210.3 J of impact energy at 40, 30 and 20 mm gauge length respectively. At ultimate failure, coconut fibres fail by fibre pull-out while sisal, jute and hibiscus cannabinus fibres fail by fracture.

One of the problems associated with Polyester and epoxy resins is pollution due to the fact that they are not biodegradable and also some volatiles are given off in the course of its usage. The problem of cost cannot be over looked. There is the need to find alternative resins that are pollution free. One of these resin is Cashew nut shell resin (CNSR). CNSR is a natural resin that can be obtained from cashew nut. CNSR is a rich source of phenol derivatives, examples are CNSL varnish (Reddish brown), Styrenated card phenol varnish (Pale yellow) and card phenol, Dehydrated Castor Oil varnish (light golden). It is a naturally occurring resin which can replace phenol in numerous applications with comparable if not better results. Like any other natural resin, it is a renewable and cheap substance which can be employed in the manufacture of variety of useful products. This paper discusses results from the development of natural composites using coconut fibre and Cashew nut

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shell resin. The mechanical properties are investigated and SEM used to study the failure mode of the composite.

Experimental

Materials

Coconut husks were locally obtained in Calabar market Cross River State Nigeria while cashew nuts were obtained from Obollo-Afor cashew plantation in Udenu Local Government Area of Enugu State, Nigeria. The silane solution constituents, catalyst (Methyl ethyl ketone peroxide (HY951)), accelerator (Cobalt naphthanate (CDA-4301)), PVA, wax and NaOH pellets were all of commercial grade.

Alkali Treatment of Fibres

Coconut fruit were obtained from local shops (Calabar, Nigeria), the fibres were extracted from the husk using mechanical extraction method and then dried at 36° C for 10 days (240 hours) in an incubator. The coir fibres were chopped and sieved into sizes between 2 and 10 mm. Alkaline treatment was carried out after sieving. The sieved fibres were placed in a stainless vessel containing 10% solution of NaOH, stirred for 2 hours and washed thoroughly with water to remove excess NaOH from the fibre. Coconut fibres obtained after alkaline treatment were dipped in NaClO 1% solution under heating (60° C - 75 $^{\circ}$ C) for 1 h, this is to present strong bleaching effect fibre. Final washing was carried out with distilled water and dried again at 60° C for 24 hours in an oven.

Extraction of CNSR

CNSR was extracted as reported elsewhere [16]. The cashew nuts were broken into two halves to remove the edible part. The shells were poured in a vat pan containing n-hexane, and allowed to stand for 24 hours. The filtered solution was heated to distil of n-hexane leaving behind CNSL. Oxalic acid and CNSL were mixed (ratio 1g: 32ml) in a three neck 500ml reactor equipped with stirrer and water cooled condenser. The solution was purge with nitrogen for 10 minutes and heated at 70°C. Formaldehyde was slowly added, while heating for about 2 hours at the same temperature. Heating was increased to 150°C to remove water. What is left behind is cardanol novolak resin. 1mole of cardanol novolak, 1mole of glycidylmethacrylate(GMA) and 0.8% of benzyltriethy ammonia chloride were mixed in a 500ml three neck reactor equipped with stirrer and water-cooled condenser. The mixture was purge with nitrogen for 10 minutes and heated to 105°C, GMA was added and allowed to heat for about ten hours obtaining the needed resin.

Composite Preparation

A PTFE mould of dimensions 302 mm x 22 mm x 7 mm for tensile and bending tests, 22 mm x 22 mm x 22 mm for compressive test were used to cast the composite sheets. A hand lay-up technique was used to prepare the samples. The treated fibres were thoroughly mixed by mechanical stirring with CNSR for 30 minutes. Prior to filling the mould with the resin and coir fibre, the inner surface of mould was coated with universal mould release wax to facilitate easy removal of the coconut fibre/CNSR composites. The mixture was then spread uniformly on the surface of the mould and hot pressed at 55°C for 30 minutes at a pressure range of 3-4 MPa. All specimens were post cured at 50°C for 12 h and machined into tensile, flexural and compressive specimen shapes of 300 mm x 20 mm x

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5 mm for tensile and bending tests, 20 mm x 20 mm x 20 mm for compressive test. The fibre content of the composite is 30%.

Mechanical Testing

Mechanical properties of samples were tested, in tension and compression using a Universal Testing Machine - UTM (Instron 1121). Bending was carried out according to ASTM D-790M standard. The sample dimensions were 300 mm x 20 mm x 5 mm for tensile and bending tests, 20 mm x 20 mm x 20 mm for compressive test. Five samples were tested for each mechanical test average result of the each was used. A crosshead speed of 5mm/min was used. All specimens were conditioned at a temperature of 23 ± 2 °C and 50 ± 5 % relative humidity for 48 hours before testing.

Scanning Electron Microscopy (SEM)

The morphologies and failure mode of the samples were investigated using a FEM-SEM XL30 scanning electron microscope. Samples were first carbon coated and imaged using a spot size of $3, \times 50$ magnifications and an acceleration voltage of 5 kV. Also back scattering image were taken.

RESULTS AND DISCUSSION

Tensile Properties

Coconut fibre is a multi-cellular plant fibre, the properties under tensile loading could be investigated through the cumulative outcome of a collection of individual cells in the plant fibre. As a multi-cellular plant, coconut fibre is expected to contain long-chain molecules comprising of a crystalline-cellulose region and a non-crystalline-lignin complex region as shown in Figure 1[17]. The non-crystalline region is expected to have long helical spiral crystals as shown in Figure 2 [17]. The deformation of a material with the above structure subjected to tension has been determined theoretically [18]. Coir with a spiral-like structure might deform by the elongation of micro-fibrils along with the non-crystalline regions or the uncoiling of the micro-fibrils with bending and twisting. For coir fibre it appears both mechanisms are involved during deformation, the dominant of the two is unclear.



Figure 1 Typical view of fringed fibril structure [17].

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Figure 2: Typical view of helical arrangement in a natural cellulose fibre [17].

The stress-strain curves of coconut fibre, cashew nut shells resin (CNSR), and the coconut fibre/CNSR composite are shown in Figure 3. The stress-strain curve for coir fibre did not show any sign of knuckle pattern as reported by some authors [17, 19, 20]. The presence of knuckle pattern was suggested to be the beginning of plastic deformation [17], others attribute it to the collapse of the weak primary cell walls and delamination between fibre cells [12]. It is well known that the properties of natural fibres depend among things on the internal structure of the fibres, the source, the age and the chemical treatment prior to characterization. The absence of knuckle pattern may be attributed to any of these factors. In this research, chemical treatment was carried out; the level and type of chemical treatment influence the properties. The coconut fibre curve is characterized by a linear stress-strain relation, the slope of which is taken as the modulus. The linear region shows a tendency to curve downward indicating some strain softening. With the progressive alignment of microfibrils (Fig. 6B) along the tensile axis work-hardening in tension is expected but this is not the case. The reason might be attributed to the age and or the chemical treatment of the fibre.



Figure 3: Tensile Stress-Strain curve of coconut fibre, CNSR and coconut fibre/CNSR composite

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The tensile strength for coconut fibre from table 1 is 48.08 MPa. This value are comparable with earlier reported value 15-327MPa [15], 69.3MPa [14], and 50.9MPa [21], but far below other reports 90MPa [20], 500MPa [22], 137MPa [23] and 142MPa [13]. From the same table the strain at break is 20.66 % while the Young Modulus is 588MPa. The strain at break is comparable with earlier reported result 18.% [20], 17.6% [21] and 24 % [13]. Similarly the Young Modulus obtained here differs from earlier reported values 2.0GPa [14], 2.0GPa, [13], 2.6GPa [20]. Figure 1 equally shows the stress strain curve of CNSR. The stress-strain curve of the cured neat resin is indicative of a typical elastomer. The tensile strength at break is 48 \pm 5.2 while the strain at break is 14 \pm 2.1 and the Young Modulus 380.33 \pm 4. The tensile strength values obtained are comparable with earlier reported values 50MPa [24] but differ with others 17MPa [25] and 18MPa [26]. The difference in properties could be attributed to gauge length, chemical modification, strain rate, condition of fibre prior to characterisation and nature of the fibre (species, location and maturity of the plant) among other factors.

Table 1: Tensile	properties of	of CNSR,	Coconut fibre and	CNSR/Coconut	fibre reinforced	composite
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	Tensile	Tensile	Tensile
	Stress (MPa)	Strain (%)	Modulus (MPa)
Coconut Fibre	48.08 <u>+</u> 3.2	20.66 <u>+</u> 1.8	588.56 <u>+</u> 5.5
Cashew nut shell Resin	48 <u>+</u> 5.2	14 <u>+</u> 2.1	380.33 <u>+</u> 4.3
Composite	163.08 <u>+</u> 4.7	7.9 <u>+</u> 1.2	2384.52 <u>+</u> 10.3

The tensile strength of coconut/CNSR composite is 163MPa more than 200% increase compare to the neat resin and coconut fibres. The linear mechanical behaviour of CNSR based composite was characterized by tensile tests performed at room temperature $(23\pm2 \,^{\circ}C \text{ and } 50\pm5 \,^{\circ}S \,^{\circ}$ relative humidity). Typical stress-strain curve obtained from tensile test for coconut/CNSR based composite is shown in Figure 3. The figure clearly shows the influence of the grafting of CNSR on the mechanical behaviour of the composite film. Young's modulus values were examined from the initial slope of the tensile curve. Table 1 show that the film display higher tensile modulus, strength, and strain at break compared to neat coconut fibre and CNSR. It clearly shows the positive impact of the surface chemical modification of coconut fibre on the mechanical behaviour of the composite film.

The composite film did not display a well-defined yield point, and do not strain-harden. From the graph no plastic deformation was observed. The ultimate and breaking strength are the same, one of the characteristics of brittle materials. The strength of a composite film may decrease or increase with the addition of natural lignocellulose fibres to a polymer matrix. Natural fibres like coconut are able to advance the strength of composite because lignocellulose fibres can support stress transfer from the polymer. The increase in tensile strength is due to the ability of the fibres to support stresses transfer from the polymer matrix.

Compressive Properties

The compressive test of CNSR/Coconut fibre was conducted according to KS F 4043/EN 12190. Five compressive strength samples with the dimensions 20 mm x 20 mm x 20 mm were prepared. The compressive strength test was performed using a universal testing machine (UTM). The compressive stress-strain curve of CNSR (figure 4) shows linearity from the onset. The same is applicable for the CNSR/Coconut composite. From table 2 the compressive strength of CNSR is 80 MPa while that of

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the composite is 215. 5 MPa. The compressive strains and elastic moduli for the resin and composite are respectively 11%, 25% 709 MPa and 1085.3 MPa. The result obtained here are comparable with those reported by Ugoamadi [24]. Most authors reported on the compressive properties of coconut fibre reinforced concrete [27, 28].



Figure 4: Compressive Stress-Strain curve of CNSR and coconut fibre/CNSR composite

Table 2: Compressive properties of CNSR, Coconut fibre and CNSR/Coconut fibre reinforced composite

	Compressive	Compressive	Compressive
	Stress (MPa)	Strain (%)	Modulus (MPa)
Cashew nut shell Resin	80 <u>+</u> 6.3	11.5 <u>+</u> 1.2	709.09 <u>+</u> 15.4
Composite	215.5 <u>+</u> 9.7	25.63 <u>+</u> 2.5	1085.3 <u>+</u> 20.23

Bending Properties

The flexural strength and modulus of elasticity of CNSR and CNSR/coconut fibre composite were evaluated in accordance to ASTM C580. Continuous measurements of the load applied and the corresponding deflection that occurred at the mid span were recorded. The maximum load was used to determine the flexural strength, and the tangent modulus was determined from the load versus deflection curve. The bending stress-strain curve for CNSR and CNSR reinforced coconut fibre (figure 5) shows some level of linearity from the onset. From table 3 the bending strength of CNSR is 35 MPa while that of the composite is 48.7 MPa. The Young modulus of resin is 331.7 MPa and that of the composite 892 MPa. There was a decrease in bending strain from 8.4 % to 5.8 %.

The literature review indicates that among other thing, surface treatments, fibre orientation, interfacial adhesion, chemical properties and physical are the major factors affecting mechanical properties of natural fibre reinforced composite. Other processing boundaries such as pressure, curing temperature

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and methodology could influence the mechanical properties and overall performance. The mechanical characterization of natural fibre reinforced composite is based on the tensile, compressive, impact and flexural strengths etc. These properties largely depend on the interfacial adhesion between the fibre and matrix, fibre strength, fibre orientation, physical properties of fibre, properties of the constituents' material and the fibre weight fraction. The higher interfacial adhesion between the matrix and the fibre smoothens, the stress transfer between them. This may have contributed to the enhanced flexural properties in this research.

Flexural strengths of natural composites are affected by the amount of reinforcement loading. It is a measure of the ability to resist the bending load. The flexural strengths of groundnut shell-epoxy composite was found to be maximum at 12.5 wt. % fibre loading while rice husks-epoxy composite was found to be maximum at 5 wt. % fibre loading[29]. Not with standing, groundnut shell-epoxy composite displayed higher flexural strength. At 30% fibre weight fraction, hemp fibre reinforced PLA composite exhibited maximum flexural strength [8]. 0.75 wt. % of cellulose nanofibre improves both the flexural modulus (3 GPa) and flexural strength (45 MPa) of epoxy resin [4]. Most authors report that maximum flexural strength can be achieved between 25-50% weight fraction of fibre [5-7, 11]. Sisal, bamboo, jute, banana and kenaf, fibre polymer composites give better flexural strengths (>100 MPa) [6,7, 11] as compared with coir [10, 11] fibre composites.



Figure 5: Bending Stress-Strain curve of CNSR and coconut fibre/CNSR composite

Table 3:	Bending properties of CNS	SR, Coconut fibre	and CNSR/Cocor	nut fibre reinforced composite
	Cashew nut shell Resin	35 <u>+</u> 2.4	8.4 <u>+</u> 1.2	331.68 <u>+</u> 21.2
	Composite	48.68 <u>+</u> 3.3	5.8 <u>+</u> 1.3	891.97±24.5

SEM Morphology

SEM images did not show any major difference between tensile, compressive and flexural failures. Figure 6 shows SEM images of (A) neat CNSR, (B) fibre cells showing lumen and middle lamellae, (B) coconut fibre showing pull-out of the resin cells and collapse of the cell walls and (D) Presence of voids due to filler detachment and crack due to resin failure. Fig. 6A shows that neat CNSR has a relatively smooth surface with brittle like tendencies. Fig. 6C shows that coconut fibre was not evenly disperse in the matrix this may have contributed to the low tensile strength of the composite. Inserted (Fig. 6C) is a very rough and highly fibrous surface of the fibre. The figure shows the tensile failure was brittle in all nature with fibre pull-out in the same direction an indication of low speed failure (fig. 6D). Fig. 6D equally shows that specimens failed by matrix shear failure with constant debonding. Debonding arises when the stress in the internal phase amid the matrix and the fibre surpasses the resident strength and so cracks are formed leading to failure. Fibres with low levels of chemical treatment tend to debond than fibre with high level of chemical treatment. In a region of high stress concentration, such as the tip of an advancing crack, fibres often fail and fracture. As the crack front continues to advance, these fibres are pulled out of the surrounding matrix leaving behind void (fig.6D). Again this occurs as a result of poor chemical treatment of the fibre surface.



Figure 6: SEM images of (A) neat CNSR, (B) fibre cells showing lumen and middle lamellae, (B) coconut fibre showing pull-out of the resin cells and collapse of the cell walls and (D) Presence of voids due to filler detachment and crack due to resin failure.

CONCLUSION

Cashew Nut Shell Resin (CNSR) was reinforced with chemically modified coconut fibres. The chemical treatment affected the mechanical properties (tensile, compressive and bending) and the adhesion between matrix and fibre, as observed in SEM images. The stress-strain curve for coir fibre did not show any sign of knuckle pattern as reported by many authors. The coconut fibre curve is characterized by a linear stress-strain relation with tendency of strain softening. Tensile properties of coir fibre obtained here are comparable with those in literature and some differences noted are attributed to gauge length, chemical modification, strain rate, condition of fibre prior to characterisation and nature of the fibre. The tensile strength of coconut fibre reinforced CNSR composite is 200% increase compare to the neat resin and coconut fibres. The composite film did not display a well-defined yield point, and do not strain-harden. The ultimate and breaking strength are the same, indicating some level of brittleness. Both compressive and bending strength-strain curves for matrix and composite shows linearity from the onset. SEM images did not show any major difference between tensile, compressive and flexural failures. All failures were characterise with fibre pull-out from resin cell, matrix shear failure with constant debonding. Voids were observed due to filler detachment and crack due to resin failure. In conclusion, the composites produced in this research, using matrix from a raw material derived from cashew nut shell liquid (CNSL) can be find useful application in the production of panels or ceilings as their tensile properties are stronger than gypsum.

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