

# Reliable Natural-Fibre Augmented Biodegraded Polymer Composites



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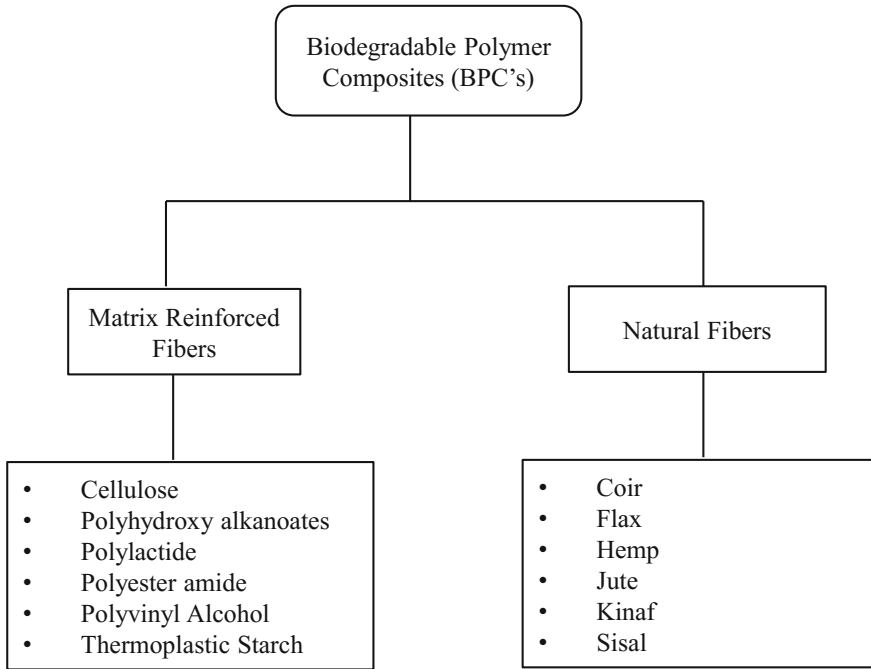
## 1 Introduction

Since the early 19th century investigations on natural fibre composites have been going on but get an acknowledgement in 1980s. Fibres are hair like materials which are similar to thread pieces, continuous filaments or can exist as distinct extend pieces. They may be twisted into filaments, yarn or cord, as a component of composite materials, can also be matted into sheets to yield a wide variety of products. Biodegradable polymer composites obtained by incorporating natural resources have witnessed a tremendous research interest nowadays. The biocomposite polymers are widely emergent areas in polymer science which perceived huge attention for their use in various applications such as automobiles, fossil plastic materials, building industries, railway coach interiors, packaging, storage devices and aerospace [1, 2].

Compared with traditional fibres (glass, aramid and carbon fibres), natural fibres (e.g. banana, coir, flax, hemp, henequen, kenaf, jute, sisal, kapok, and many more— Fig. 1) offers many advantages, for instance, abundance and lower cost of materials, decrease in density, biodegradability, nominal health hazards, sustainability, flexibility and less machine wear during processing, comparatively high tensile and stretch modulus, extraordinary stiffness and strength [1]. Additionally, biodegradable, eco-friendly and renewable properties of natural fibres assist their disposal by incineration or composting and alleviate the use of non-biodegradable polymers. In addition, these fibres are environmentally benign as their dimensional structure encompass confiscated atmospheric carbon dioxide and emit lower energy comparative to industrially manufactured synthetic fibres [3].

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**Fig. 1** Classification of biodegradable polymer composites

BPC's also showed fairly good mechanical properties, has high specific strength, and a good amount of tensile strength as an outcome of interfacial adhesion amid the matrix and fibres. Generally, the tensile strengths of BPC's increase on increasing the fibre content reached an optimum value and later a drop in value is witnessed. Also, the tensile properties of composites are prominently boosted by adding fibres to a polymer matrix as fibres have the much greater strength and stiffness relative to the matrices. Furthermore, it was established that the composites having fibres in the perpendicular direction deliver inferior tensile strength compared to the composites having fibres in the parallel direction [3–5]. Hence, it is required to alter the surface of fibre by means of chemical modifications to enhance the adhesion amid fibre and matrix by employing suitable processing methods and parameters to produce optimum composite products.

This chapter briefly deals with the reported works on the characterization of natural fibres, along with the comparative properties of natural and synthetic fibres (Table 1), advantages and disadvantages of BPC's in addition to manufacturing techniques, chemical and physical treatments and their applications in several areas.

**Table 1** Properties of various natural-and synthetic fibres [1, 7–9]

Fibre	Density (g/cm <sup>3</sup> )	Elongation	Elastic modulus (GPa)	Specific elastic modulus (GPa)	Tensile strength (MPa)
Aramid	1.4	3.3–3.7	63–67	33–36	3000–3150
Carbon	1.4	1.4–1.8	230–240	164–171	4000
Cotton	1.5–1.6	7.0–8.0	5.5–12.6	3.1–5.8	400
Coir	1.2	30	40	20.4	593
E-glass	2.5	0.5	70	28	2000–3500
S-glass	2.5	2.8	86	41.2	4570
Flax	1.5	2.7–3.2	27.6	26–46	500–1500
Hemp	1.47	2–40	70	47	690
Jute	1.3	1.5–1.8	26.5	7–21	393–773
Kenaf	1.45	1.6	53	36.5	930
Polyester	1.2–1.5	2.0–4.5	2	–	40–90
Polyhydroxy alkonates	1.1–1.4	1–6	3–6	–	35–100
Sisal	1.5	2.0–2.5	9.4–22	6.3–14.7	511–635

## 2 Classification of Fibres

Mostly, polymers may be categorized into two diverse classes: thermoplastics and thermosetting. Both types of materials were widely used as matrices for fabrication of bio-fibres. Polyethylene (PE), polyvinyl chloride (PVC), and polypropylene (PP) are frequently used thermoplastics; whereas epoxy, phenolic, and polyester resins are generally used thermosetting polymers for the manufacture of composites. Now a day, fibres obtained from natural sources like jute, flax, coir, hemp, kenaf, sisal etc. are extensively used in the fabrication of composites.

### 2.1 Drawbacks of Natural Fibres

The natural fibres suffer limitations, namely: (1) strength, (2) water absorption, (3) thermal stability

1. **Strength:** The tensile strength of BPC's formed using natural fibres is very low comparative to glass, aramid and carbon fibres. This is due to the incompatibility amid the fibre-resin matrix, which in turn reduces the wettability of the fibres and creates a challenge in their productions. Although, by comparing the specific strength of both natural and synthetic fibres, it is observed that there is barely any difference.

2. **Water absorption:** Another limiting factor for the assembly of BPC's is the absorption of water from the atmosphere or direct contact with the surroundings. The difference in the polarity of hydrophilic natural fibres and hydrophobic polymer matrix provides minimal interactions and results in the formation of aggregates. The absorption allows the distortion of surfaces of resulting composites via swelling and forming voids, thereby alleviating the interfacial adhesion and weakens the strength in addition to proliferation in the mass of composites. The wettability also permits the growth of fungi on/in the composites resulting in decay of their structure [6]. The high water absorption indicates inadequate resistant to moisture, which provides reduced tensile properties to reinforced natural fibre composites (Table 1).
3. **Thermal stability:** Natural fibres are of restricted thermal stability and lead to microcracking and, consequently, thermal degradation may possibly occur in the course of BPC's processing at an elevated temperature, specifically in the cases of hot compression and thermal extrusion processes.

## 2.2 *Advantages of Natural Fibres*

The biggest advantage of using natural fibres for the fabrication of BPC's is the low cost of materials, reduced density, and sustainability. Natural fibres are easy to cultivate and can be grown within few months and are cost effective. They also have the perspective as a cash crop for the agriculturalists. Natural fibres are eco-friendly, lightweight, consumes lesser energy, non-abrasive, non-carcinogens, strong, renewable, less health risk, non-irritation to the skin, recyclable and biodegradable, and have small processing time [2].

The fabrication of biocomposites uses various techniques which include: compression moulding, extrusion (extensively used for green biocomposite), filament winding, injection moulding, machine press, pultrusion, resin transfer moulding, sheet moulding compound. Thermoplastic bio-composites can be mainly processed through compounding, compression moulding, extrusion, injection, and vacuum consolidation. Alternatively, thermosetting biocomposites are manufactured by compression moulding, vacuum assisted resin transfer moulding, hand lay-up, pultrusion, resin transfer moulding, and vacuum bagging [10–12].

## 2.3 *Strategies for Surface Modification in Natural Fibres*

Surface modification is one of the crucial processes in the fabrication of biocomposites since natural fibres possess hydrophilic characteristic and with the aim of improving the compatibility of a hydrophobic polymer matrix with the natural fibres, this surface modification is needed. Surface modification is categorized into

types, namely chemical and physical methods. The surface modification eases fibre dispersion within polymer matrix along with enhancement in their interaction. Various techniques described in the literature surveys for altering fibre-matrix adhesion consist of acetylation [13], acrylic acid treatment [14], bleaching [15], esterification [16], grafting of monomers using maleic anhydride [17], and using bi-functional molecules [18]. The usage of coupling agents such as isocyanates [19], chitosan [20], maleated polypropylene [21], silanes [22], titanates [23], zir-conates [24], etc. have been testified in previous studies for the improvement of conventional polymer composites.

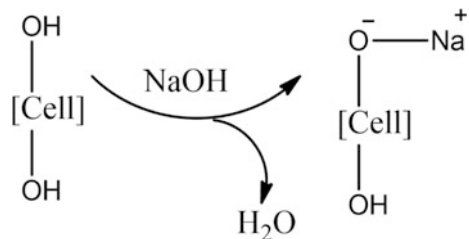
### 2.3.1 Chemical Techniques

A wide range of chemical techniques is reported in the literature but alkaline-, silane-, esterification-, and isocyanate treatments are some of the frequently used chemical techniques, which are described as follows:

1. **Alkaline treatment:** Alkaline treatment is a unique approach to be used as a chemical treatment of BPC's. This method is also known as mercerization and is usually used to reinforce thermoplastics and thermosets with natural fibers by removing lignin, wax and oil from the external surface of the fibers. This methodology disrupts the hydrogen bonding in the structural framework of BPC's, providing surplus positions for mechanical interlocking, henceforth support surface irregularities and boost fibre and matrix diffusion at the boundaries. In alkaline treatment, the fibres are dipped in sodium hydroxide (NaOH) solution for a fixed time period depending upon the interaction between the two. The resulting interaction facilitates the ionization of the hydroxyl group to the alkoxide group (Fig. 2), which is then stirred continuously in a binary solvent such as water-ethanol solution (80:20) for 1 h and the solution is kept undisturbed for ~3 h. Consequently, the fibers are washed repeatedly with distilled water followed by drying in a hot air oven at 80 °C for ~5 h [25, 26].

The literature surveys reported that the natural fibers such as coir, ramie, sisal, jute were utilized to fabricate BPC's using alkaline treatment and it was established that these composites possess high storage as well as significant flexural modulus [27].

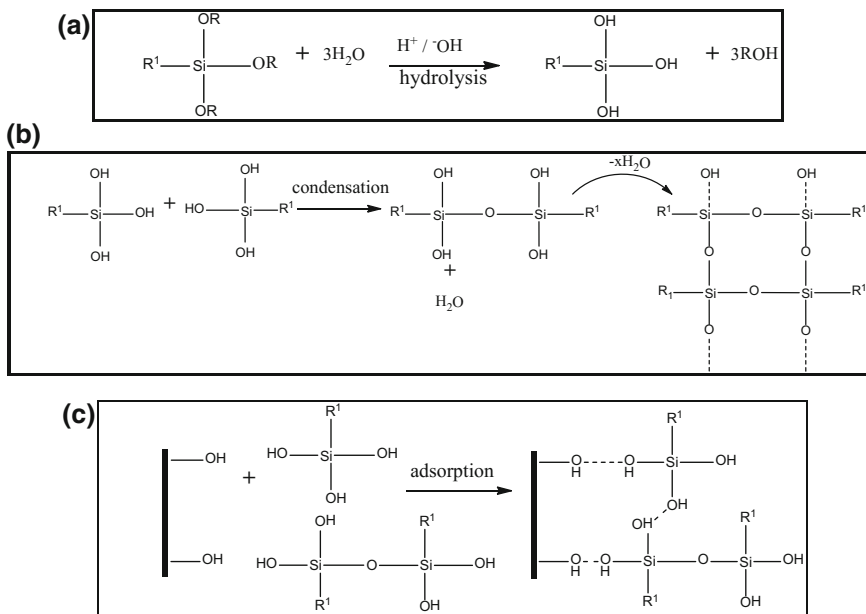
**Fig. 2** Ionization of the hydroxide group used in alkali treatment of the fibers



2. **Silane treatment:** Earlier silanes (with chemical formula  $\text{SiH}_4$ ) were reinforced with glass fibres for the augmentation of polymer composites. Silanes reduce the prevalence of hydrogen bonding in the matrix-fiber structural complex. Presence of moisture allows hydrophilic alkoxy groups to form silanols, which in turn responds to the hydroxyl groups of the fibre and form a stable, cross-linked network due to the covalently bonded structure with the cell wall (Fig. 3a, b).

As a consequence, hydrophobic fibre surface is generated; consecutively intensify the compatibility with the polymer matrix (Fig. 3c). A significant enhancement in tensile strength was discovered as a consequence of strong interfacial bond produced by the acid and water conditions during the course of fibre pretreatment [22]. The modification of kenaf and polyester fibre composites finds mention in the literature using silane treatment method [28, 29]. The fiber modification offers higher storage modulus and lowers  $\tan \delta$  values than those with untreated fiber indicating a greater interfacial interaction between the matrix resin and the fiber.

3. **Esterification:** Esterification involves the modification of the surface of fibres especially wood composites with organic acid anhydrides (Fig. 4). Acetylation



**Fig. 3** Interaction of silanes with natural fibers via different processes: **a** hydrolysis, **b** condensation, **c** adsorption [30]

with acetic anhydride (non-cyclic anhydrides) and maleic anhydride (cyclic anhydrides) is broadly defined in the literature [17]. In Acetylation process, a hydroxyl group is converted into an ester group via association of the free hydroxyl groups present in wood composites with the carboxylic group of the anhydrides.

4. **Isocyanate treatment:** Isocyanates are organic compounds having isocyanate group  $-N=C=O$  in their structure. These are highly reactive with polar groups such as hydroxyl ( $-OH$ ), amino ( $-NH_2$ ) and others (Fig. 5). Isocyanates are not considered feasible treatment method for natural fibres but testified a meek improvement in strength of polymer matrix relative to the unaided matrix.

### 2.3.2 Physical Techniques

Physical approaches [29, 31] mentioned in the literature involves the corona or plasma treatments for amending conventional polymers. In recent years, a “greener” alternative is available for the expansion of polymer composites, which comprises plasma treatment for reinforcement of natural fibres. Some of the chemical techniques mentioned previously proved to be harmful, e.g. isocyanates are hazardous, and as a result, such agents are not viable for the augmentation of BPC’s. Therefore, physical methods involving plasma treatment provides a better option for treating natural fibres. Plasma has the tendency to modify the properties of surface/interface of natural fibres via formation of free radicals (*for instance* electrons, ions, etc.) on their surfaces by the bombardment with high energy particles inside the stream of plasma (Fig. 6) [32].

Using physical techniques various surface properties, for example, chemistry, wettability, and surface irregularities of composites can be improved without using

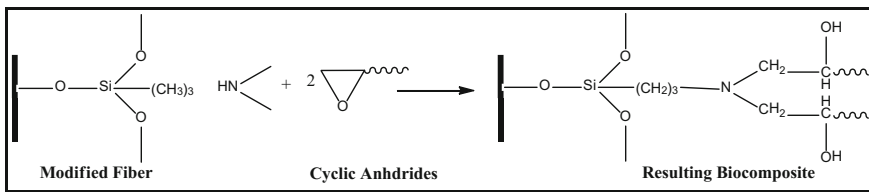


Fig. 4 Coupling reaction of modified natural fibers with anhydrides

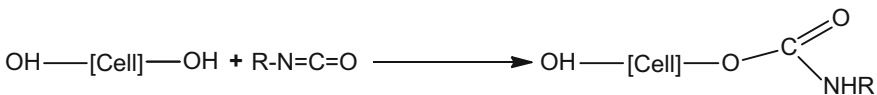
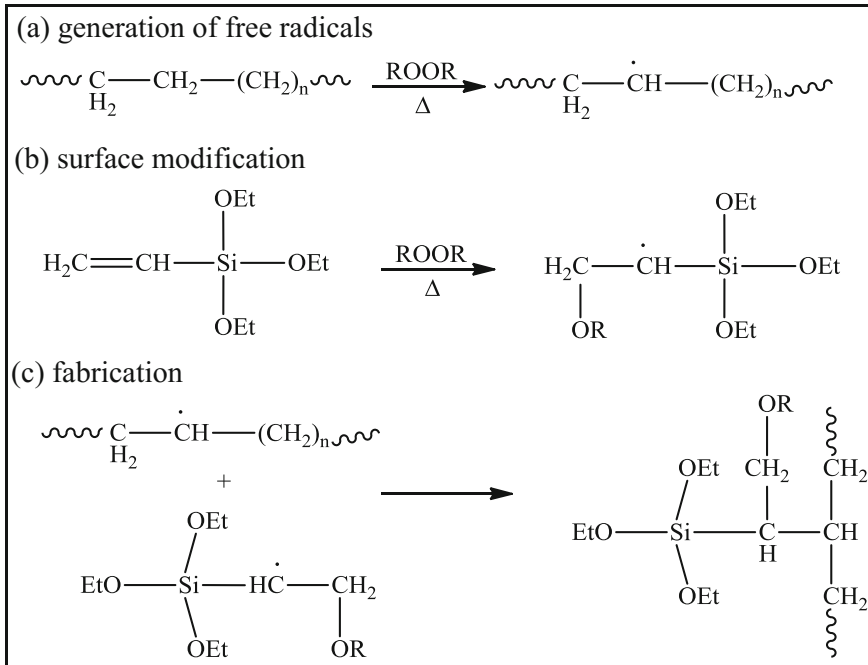


Fig. 5 Isocyanation of fibers



**Fig. 6** Fabrication of silanes onto polymer surface by free radicals (where Et = ethyl group)

solvents or employing any other harmful materials. An alternate approach for surface modification by plasmas is to alter the carrier gas or by depositing free radicals or other reactive species on the shells of natural fibres [33]. Furthermore, this can be stimulated by embedding monomers or polymers on the surface of the reactive natural fibre, which accelerates its aptness for the polymer matrix.

### 3 Types of Biodegradable Polymer Composites (BPC'S)

Over the decades with the accelerated progress of biocomposites, a considerable interest has been developed for polymer matrices that are reinforced with natural fibres. Owing to difficulties in discarding the non-biodegradable polymers, researches are continuously carried out to fortified new biodegradable polymer composite materials from natural resources.

Conversely, natural fibre reinforced composites (BPC's) are manufactured from biodegradable polymers to overcome the shortcomings of non-biodegradable polymers composites. Biodegradable matrices were enclaved with natural fibres to amplify the properties of BPC's and they are scientifically sound, lightweight, high mechanical properties and cost-effective. Some of the BPC's along with their synthetic method and properties are explained as follows.



### 3.1 Coir Fibre Reinforced Composite

Coir is generated by the husk of coconut fruit fibre. The life expectancy of coir is more comparative to other natural fibres as a result of high lignin content. Coir fibre reinforced polyester matrix was tested for their interfacial adhesion characteristics against different ageing solutions and was found to display excellent interfacial adhesion under arid conditions [34]. Coir fibre reinforced polymer composites are developed for industrial and various household applications such as automotive interior, helmets and post boxes, packing material, paneling and roofing as building materials, mirror casing, projector cover, storage tank, paperweights, voltage stabilizer covers.

The efficiency of coir fibre fabricated epoxy composites is dependent on alkali treatment in addition length of the fibre. Coir fibres having lengths 10, 20 and 30 mm were cured with NaOH for 10 days. Fibre length was Alkali treated composite along with increased fibre had Coir fibre having length of 30 mm and 8% alkali concentrations showed better impact strength ( $27 \text{ kJ/m}^2$ ) [35, 36]. Pretreated coir based composite have far better mechanical and flexural properties than the untreated one. On increasing fibre content the flexural strength of composite decreases as the matrix is inadequate to shield the complete surface of the coir fibre (Fig. 7).

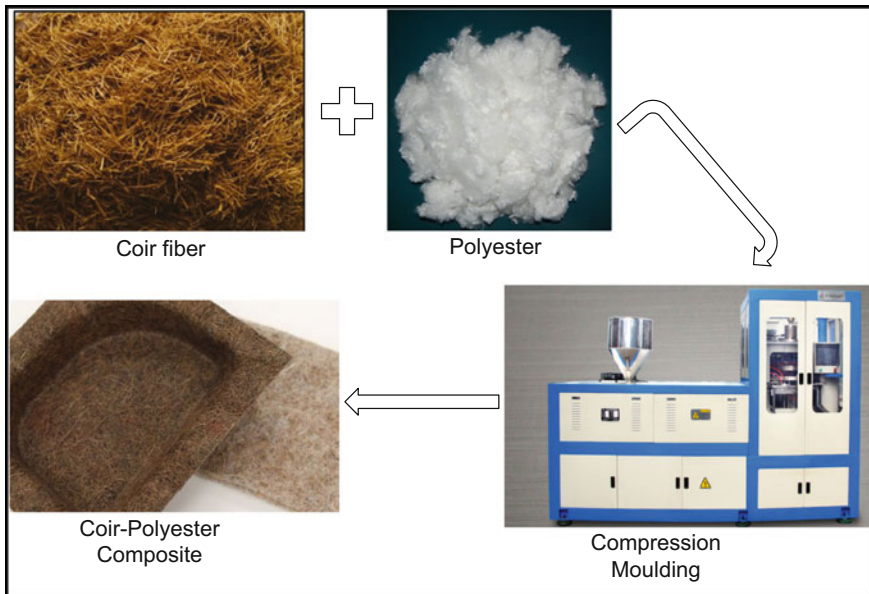


Fig. 7 Coir reinforced polyester composite formation via compression moulding

### ***3.2 Cellulose Fibre Reinforced Composite***

Cellulose and its derivatives (lignin, hemicellulose, pectin etc.) are semi-crystalline polysaccharides that impart hydrophilic nature as well as tendency to hold the fibre. These materials have found wide use in the potential matrices-composites as strong molecular interactions exist at the interface of cellulose fibres-polymer matrix composites, which in turn results in strong interfacial adhesion. The composites may be treated via blow and rotation moulding, extrusion, injection moulding, etc. to form fundamental components [18, 33].

### ***3.3 Jute Fibre Reinforced Composite***

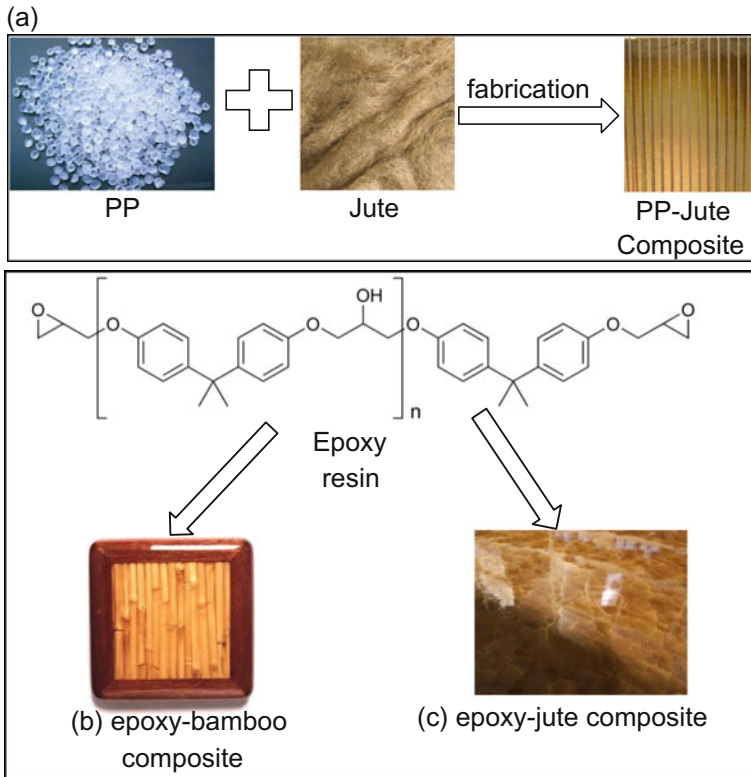
Jute has wood like characteristics, has a high aspect ratio, good insulation properties, strength to weight ratio. In view of above-mentioned properties, jute fibre reinforced polymer composite has tested for grooved sheet, door, furniture, roofing, floor tiles, I-shaped beam, recovery of underground drain pipes, window, and water pipes [37].

The jute fibre reinforced PP composites were analyzed for their mechanical properties. The washing of fibres preceded by alkaline treatment and bleaching revealed intensification in tensile strength and tensile modulus with an increase in % weight fraction and NaOH % of fibres in the PP matrix (Fig. 8a).

Jute fibres stiffen epoxy composites were examined and results were also compared with bamboo fibre supported epoxy composites and formers were found to have a higher strength. Additionally, upon alkaline treatment jute fibre reinforced epoxy composites encompass enhanced mechanical properties of bamboo fibre reinforced epoxy composites (Fig. 8b, c).

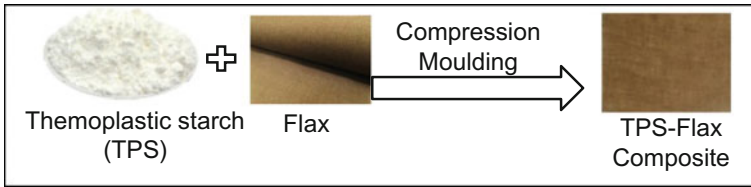
### ***3.4 Poly Lactide (PLA) Fibre Reinforced Composite***

The blend of natural fibre and PLA bids an excellent response to preserve the viable economic and ecological development. Ochi [38] carried out a study on PLA composite and explored that PLA fabricated unidirectional biodegradable composite materials showed tensile strengths of 223 MPa. He also evaluated the biodegradability of same via composting and the conducting tests revealed a 38% decrease in composites weight after a time period of four weeks. Oksman et al. [39] reported the fabrication of PLA-Flax composites and matched them with frequently used polypropylene (PP) flax fibre composites (PP-Flax). The comparative study marked 50% higher mechanical properties of PLA-Flax fibre composites over PP-Flax fibre composites.



**Fig. 8** a Jute enforced PP composites, b epoxy-bamboo composites, c epoxy-jute composite

PLA-Natural fibre composites containing >30% weight fibre showed an improved tensile modulus with lesser tensile strength on comparing with untreated PLA. This was ascribed as a result of poor interfacial interaction flanked by hydrophilic cellulose fibres and hydrophobic PLA matrix, as well as an insufficient fibre dispersion caused by the elevated amount of fibre agglomeration [40]. Hu and Lim [41] inspected that tensile properties of hemp fibre enforced PLA composite significantly improved upon alkali treatment than that of untreated composites. The composites produced using 40% treated fibre has approximately twice tensile modulus comparative to neat PLA (35 GPa). Fabrication of PLA with natural fibres is most likely done by conventional methods such as blow- and injection moulding, extrusion, as well as film-forming operations.



**Fig. 9** Thermoplastic starch-flax composite using compression moulding

### 3.5 *Polyhydroxyalkanoates Fibre Reinforced Composite*

Polyhydroxyalkanoates (PHAs) are found to be renewable and biodegradable and represent a class of polyesters that are manufactured by bacterial action [42]. The fabrication of the green composites was carried out by Singh and Mohanty [43] by means of injection moulding succeeding the extrusion compounding of bacterial polyester (PHBV) i.e., poly(hydroxybutyrate-co-valerate) with 30–40 weight percentage of bamboo fibre. They also examined morphological, mechanical, and thermomechanical properties and corroborate that the tensile modulus and storage modulus of PHBV composites amplify progressively with increasing fibre loading. Moreover, the tensile strength of bacterial polyester was reduced by adding bamboo fibre due to insufficient interfacial interaction amid fibre and matrix.

### 3.6 *Thermoplastic Starch (TPS)*

One of the utmost popular eco-friendly biodegradable polymer-thermoplastic starch can be used as a matrix in fabricated of biocomposites [44, 45]. In the quest for improved performance of biodegradable and environmental acceptable TPS polymer, natural clays can be accumulated on to its surface to produce nanocomposites. Ecologically acceptable filler (such as clay) improve the properties of TPS so that it can be used in various applications. It has been established that the tensile strength of TPS showed an increase from 2.6 to 3.3 MPa on treatment with 5 wt% sodium montmorillonite (Fig. 9).

## 4 Conclusions

Natural fibre fabricated biodegradable polymer composites (BPC's) are becoming scientifically sound for a plethora of applications as they are lightweight, environmentally benign, and possess good mechanical properties. The modification of the surface of fibres is presently an area of research. The mechanical properties of biodegradable fibres such as sisal, kenaf, hemp, coir, jute, reinforced composites

have been discussed. It was established from the various studies that the tensile modulus and strength raises with an increase in fibre content. It was also accepted that the specific properties of natural fibre composites were far superior relative to synthetic fibre reinforced composites (glass, carbon and aramid). This advocates that the natural fibre composites would prove to be a potential candidate to substitute glass and alike materials in various applications. However, hydrophilic characteristics of biodegradable polymers challenge them to be a good candidate for outdoor applications due to reduced adhesion between natural fibres and matrix resins. However, BPC's are in demand these days and open their possibilities as an excellent candidate in a wide range of applications, such as an automobile, constructional and household applications.

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