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# Review article



# material in biocomposite production

Coir fiber as thermal insulator and its performance as reinforcing

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# ABSTRACT

Coir is a lignocellulosic natural fiber derived from the coconut's husk, an abundantly found fruit or nut worldwide. This fiber has some unique characteristics, such as its resistance to seawater, microbial attack, high impact, etc. But its low thermal conductivity or high thermal insulating property makes it suitable for being used as insulators in civil engineering sites. On the other hand, the sustainability of a material depends heavily on its environmental impact of the material. For making sustainable materials like biocomposite, there are no options other than using polymers derived from natural renewable sources. Polylactic acid(PLA) is an example of those types of material. And these materials are often being reinforced by fibers like coir for various reasons including improving mechanical properties, reducing the cost of the material, and improving the material's sustainability. Many coir-reinforced sustainable biopolymer composites have already been produced in many pieces of research, which will be discussed in this paper, along with the chemical and physical structure of coir fiber. In addition, this paper will try to focus on the insulating properties of coir and coir-reinforced composites while will also compare some properties of the composites with some commonly used materials based on different parameters to show the suitability of using the coir fiber in heat-insulating applications and to produce sustainable biocomposite materials.

# 1. Introduction

A Composite is a material made using two or more materials, and the final material would show different characteristics than the used materials [1]. Generally, two materials, along with other additives, are used to make a composite material [2]. One of those two materials can be a matrix or resin, and the other one can be a reinforcement for the matrix. These reinforcements are used to improve mechanical characteristics, lower the material's weight, improve the material's sustainability, etc [1,3]. There can be more than one

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matrix or reinforcing material while using two different matrix only can also be an option. The reinforcement can be a fiber or some particles. The matrix and reinforcing materials can be synthetic polymers or natural polymers, or either can be natural, and the other can be synthetic [4]. Composites made of synthetic resin and natural reinforcing material have become very popular nowadays [5–8]. These natural reinforcing materials are generally natural fibers. There are several reasons why natural fibers are selected as reinforcing material. Natural fibers are biodegradable and non-toxic in general. And in addition, the plants of those fibers also have the ability to suck carbon from the environment, which makes these fibers responsible for reducing carbon emission. On the other hand, these fibers are generally of very low weight and low cost compared to many synthetic polymers for which these natural fiber-reinforced composites are gaining popularity in automobiles, packaging, military, etc., various sectors [2,9-13]. There is another reason which has become very important these days, and that is the biodegradability of composite material. The earth has become endangered due to the use of non-biodegradable plastic materials, which are causing various problems when being dumped, such as clogging up drains, littering in the rivers and oceans, reducing soil strength, causing the greenhouse effect, and also doing harm to the environment in several other ways [14–17]. So, materials need to be biodegradable for the betterment of the environment. Actually, biodegradable means materials that are degraded by biological components such as bacteria, fungi, etc [18]. If a composite material contains one or more bio components or natural components, then that composite is called biocomposite [2,19-21]. Biocomposites can be fully biodegradable or partially biodegradable depending on the materials used for making it [4]. Partially biodegradable ones contain either fiber or matrix as the biodegradable material. Actually, these materials are also degradable, but they will take a very long time to degrade fully as only the portion containing biodegradable polymer would degrade biologically within a short period, while the rest will take a long time. And sometimes, the biodegradable part also tends to degrade more slowly than usual or does not degrade at all due to being covered with nondegradable contents, for which these types of composite materials become nondegradable or hard to be degraded, both of which are least desirable in terms of the environmental requirements [22]. Both matrix or resin and reinforcing material need to be biodegradable to be fully biodegradable. But these types of materials don't have all the attributes of synthetic materials, especially the low price, availability, easy processability, etc., of synthetic materials compared to biobased materials, which often limits their applications in many sectors. That's why natural fiber-reinforced synthetic polymer composites are preferred in all sectors these days. When choosing natural fiber, there are many options, such as cotton, jute, flax, ramie, kenaf, sisal, coir, etc [9,23]. Coir is a lignocellulosic fiber. It is obtained from the husk of coconut [24]. In this regard, coir fiber can be an excellent option, as it contains a low percentage of cellulose and a high percentage of lignin [25]. In terms of production, coir sits behind jute and well ahead of flax, ramie, etc., in 2017, according to the Food and Agriculture Organization of United Nations (FAO) [26]. The annual production actually was over a million tonnes in that year across the world. It used to be preferred by mariners for rope making because of its better resilience and resistance to seawater, as this fiber can tolerate sudden stress or pull, which breaks many other fibers [24]. Coir fibers are also known for their thermal insulating property, which makes it suitable for making composites that can act as heat insulators [27]. Heat insulators or thermal insulators are resistant to thermal currents. These types of materials help prevent external heat from entering or going out. Insulating materials are classified into three categories, namely organic, inorganic, and composite [28]. The coir fiber may fall into the category of organic materials which also include wood chips and similar materials. The composite materials can include all types of materials, and coir fiber-reinforced composite structures can also be a part of that. The heat insulation property is mainly affected by moisture content, temperature difference, and bulk density of a material which depend on the physical and chemical structure of the material [29]. The coir has the required attributes to act as an insulator, especially when being used in a composite structure which will be discussed in the latter part of this study. In fact, coir has been used in composite developments more than in other applications (e.g., in floor-matt and mattresses) nowadays, which includes the uses in many biocomposites. However, biocomposite production is not as straightforward as many may think. A sustainable, eco-friendly product is something that uses resources or raw materials in a manner that will not harm the ecological balance, but at the same time, the rate of production or growth will remain up to the required level [30–32]. So, in producing a sustainable product, both matrix and reinforcing fiber need to match that concept. To be selected as a sustainable biopolymer, the polymer needs to be efficient in production, emit the least amount of greenhouse gases during production and lifetime, be least costly, and have good enough properties for industrial applications [33]. In that case, petroleum-based synthetic polymers can't be selected because of the emission of greenhouse gases during the production and degradation process. Generally, polymers from renewable sources, or more precisely, polymers derived from plants, are the ones that can match the requirements of sustainable polymers. The most commonly used biopolymers are polylactide, starch, cellulose, cellulose derivatives, etc [33]. On the other hand, the unique characteristics of coir also make it suitable for developing composite materials for the thermal insulating purpose, which could be really useful in replacing currently existing synthetic materials, such as polyurethane, polystyrene, etc., that are harmful to the environment. This paper will discuss the thermal insulating property of coir fiber and its performance as a reinforcing material for some eco-friendly biocomposites.

Table 1	
Chemical Composition of Coir fiber	[2,9,35-38]

Content	Percentage
Colluloso	26 42

Somen	rereentage
Cellulose	36–43
Lignin	32–45
Hemicellulose	0.15 - 0.25
Ash	2.22
Pectin and related compounds	3
Water-soluble contents	5.25

# 2. Physical structure and chemical composition of coir fiber

Coconut contains a protective shell and husk around a nut containing white meat in the inner part called copra. Both green and ripe coconuts have water in that nut. The nut is not a quite visible or prominent figure in green coconut, but it gets to its shape after the fruit ripens. The white meat covers about 28% of the total weight of the whole thing. In the husk part, about 12–44% of its total weight is generally the fibers [24,34]. The fibers are lignocellulosic. It contains cellulose, lignin, hemicellulose, wax, and other materials. The chemical composition of coir fiber is given in Table 1.

The main component of cellulose is a polysaccharide in which the molecular units are linked by  $\beta$  1, 4 glycosidic bonds [39,40]. It contains three hydroxyl groups which produce inter and intramolecular hydrogen bonds with other cellulose chains between the fibers [41]. Hemicellulose is termed a matrix polysaccharide [42]. The major components of hemicellulose are xylose, galactose, arabino-furanose, and glucuronic acid residues. It has quite stable hydrogen bonds with cellulose and is insoluble in hot water [24]. Lignin is found in the middle lamella (a glue-like layer that adjoins plant cells) and cell walls of the fiber mostly along with some other places. It is a three-dimensional structure made by phenylpropanoid building units, and it is responsible for the stiffness of the fiber [43,44]. The fat and waxes are generally found at the surface of the fiber, while the water-solubles are water-soluble materials that can be removed by boiling [24,45].

Coir fiber has two types: green and brown fiber (Shown in Fig. 1(a-c)) [38]. Fibers collected from green coconut are called green coir (Fig. 1 (a and b)), while fibers collected from ripe coconut are called brown coir fiber (Fig. 1(c)). The chemical composition of both types of fibers is quite similar. But the green coir fibers are suitable for bleaching and dyeing [34]. In contrast, white fibers are much weaker and softer than brown fibers [3]. But the final fibrous materials are almost similar in appearance as the fibers from green coconut have a very slight whitish shade in comparison to the fibers from brown shells, and eventually, the shade actually diminishes, and both fibers show almost the same color by the time those are used in any application because of drying of the green fibers. Fig. 1 shows the two types of fibers (Fig. 1(a) and Fig. (b) show the green coconut contents, while Fig. 1(c) shows the brown fibers).

The fibers are extracted using retting [34,38]. In fact, the physics of coir has made the retting process essential for fiber extraction. The physical structure of coir fiber actually helps it to be resistant to microbes. The fibers consist of ultimate fiber cells. The ultimate cells of coir are held together by the middle lamella, termed the inner middle lamella. It is chemically linked with cellulose cell walls, which make this layer chemically inert, for which the fibers don't let microbes alter them chemically and show greater resistance to microbial attack as a result. But in the case of the unidirectional fibers, which are also held in bundles by another layer of the middle lamella, also known as the outer middle lamella, it is not involved in many vulnerable chemical bondings to microbial attack. That's why the fibers can be extracted by retting, depending on the bacterial attack during merging or submerging in water [24]. The bundle fibers are shown in Fig. 2.

Coir fibers are found in bundles, of which about 56–65% of fibers are 150 mm long, while the staple length can be up to 350 mm. About 5–8% of fibers (short fibers) are found below 50 mm in length. The fiber fineness varies from 50 to 300 µm, while the individual



(c)

Fig. 1. Images of different states of Coir fiber: (a) Green Coconut shell, (b) Green Coir fiber, (c) Brown Coir fiber. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

cells of fibers are about 1 mm in length and 5–8 µm diameter [38]. Generally, the length and diameter of the cells vary depending on species, maturity, location of the fiber within the plant, and extraction procedures [24].

The brown fibers are generally used in mattresses, automobile seats, brushes, brooms, nets, etc., while the green fiber can be finer and more flexible, which can be used in making twine, marine cordage, floor mats, etc. It is very often used in geotextiles for reinforcing embankments, preventing soil erosion, drainage, composite making, etc [24,34,46].

#### 3. Insulating property of coir fiber

The tensile properties of coir fiber are inferior to jute, flax, sisal, etc., but the specialty of coir fiber is its superior properties in salty water as it can resist salty water and any microbial attack while it can also be stretched beyond its elastic limit without rupture [38,47, 48]. But the one of the main advantage of coir is its thermal properties. It is often found that when coir is used as reinforcement in composites, those composites are known to have low thermal conductivity [49–51]. It is due to the low thermal conductivity of coir fiber which results from several factors like moisture absorbency, having empty spaces in the fiber structures (which are also responsible for high moisture absorbency), etc [52,53].

The thermal insulation property depends on three factors, the operating temperature, the density, and the thermal conductivity of the material, while the moisture content is also mentioned by some researchers [29]. The operating temperature cannot be controlled as this depends on the environment, and this will affect the conductivity naturally, either in a positive or negative way. The material's density also depends on the materials' morphological structure. In contrast, the material's thermal conductivity is controlled by the molecular arrangement in the structure. The conductivity in non-metallic materials depends heavily on the vibrational characteristics of the material. The highly oriented molecular arrangement helps transfer the vibrational movement in the material, leading to better heat conductivity. From that point of view, the amorphous regions offer more resistance to heat conductivity. Another factor can be the presence of an aromatic backbone in the structure, which is known to help the heat conductivity of a material. Meanwhile, cross-linked structures and long molecular chains help heat conductivity also. This reason is responsible for the comparatively high heat-conducting ability in polymers with a cross-linked structure rather than a linear structure [54].

The fiber being focused on here is coir, a cellulosic fiber with a significant amount of amorphous portion within the structure, which is relatively high in percentage (around 60%) [55]. It means the fiber should conduct a low amount of heat. Another factor is the presence of an aromatic backbone in the structure, which is not present in cellulose, for which all cellulosic materials are slightly poor conductors of heat [29,56]. And the cellulosic chains are not cross-linked either, which has been stated as another reason for increasing heat conductivity. So, as a result, coir fiber is expected to be quite a low conductor of heat.

Another important factor stated above is the morphological appearance of coir fiber. For example, the air is known as a poor heat conductor, which means if a material can entrap air in its structure, that will definitely show low heat conductivity than usual. Coir fiber has a physical structure that is quite porous as about one-third of the structure is filled with air due to having a lot of cavities inside the fiber structure, while at the same time, the fiber cells are quite narrow and hollow. It makes the fiber a poor conductor of heat [57,58]. And the water-absorbing property that makes the fibers take moisture from the atmosphere is also responsible for the poor heat conductivity of the material. These reasons are responsible for making the coir fiber one of the poorest heat conductors. These also play a similar role when the coir fibers are used in the composite structure, while the use of synthetic polymers as resins which do not have the ability to form chemical bonds with the fibers, suffers more because of the pores created by the fibers that entrap air within the composite structure which become responsible for reducing heat conductivity of the material even more.

The thermal conductivity properties of the material actually shape the other thermal properties. The thermal conductivity and thermal resistance of material are inversely proportional to each other as the unit of thermal conductivity is watt per meter per kelvin (W/mK) while the unit of thermal resistance is K.m/W. It means a material with low thermal conductivity is going to have high thermal resistance and vice versa. Since the thermal conductivity of coir fiber is quite low, its thermal resistance of it is definitely higher than



Fig. 2. Scanning Electron Microscopic image of Coir fibers (Bundle fibers comprising ultimate cells).

usual. This phenomenon also controls the performance of the coir fiber or coir fiber reinforced materials in typical thermal tests such as TGA (thermogravimetric analysis), which measures weight loss with applying temperature. The weight loss percentage shows similar trends with heat conductivity as the weight loss increases with the material's heat conductivity and vice versa [59]. So, in that sense, an increase in the weight loss of the material means the material would have less resistance to heat. But the thermal degradation temperature in TGA can show better results for materials with higher thermal conductivity (which is expected to have low thermal resistance due to absorbing more heat) if the material has a higher thermal degradation temperature. For example, polystyrene has higher thermal conductivity (0.13W/mK) while the degradation is higher than coconut contents (0.04–0.05 W/mK), but the degradation temperature is slightly higher (Up to 425 °C for polystyrene and around 400 °C for coir fiber) [29,60–62].

Researchers have already had a few applications of coir as an insulating material. Mintorogo et al. used coir in concrete slab roofings [63]. They found the temperature difference in the room air from outside to be 2.8°-3.1 °C while the temperature difference at the rooftop surface was about 13 °C which helped reduce the energy consumption to 3–9% on average. The experiment took place in Jakarta, Indonesia, where the average temperature varies from 24° to 34 °C. The comparison was made with a conventional concrete rooftop, and the coconut fibers were blended with the concrete materials to build the coconut fiber-reinforced rooftop. The researchers found that coconut fiber-reinforced rooftops delayed the release of radiated heat during the hot daytime, for which the temperature within the rooms was lesser than usual during the daytime. But the concrete could get cooled quickly, which resulted in slightly cooler room temperatures at nighttime, while the coconut fiber released the heat slowly, which resulted in slightly high temperatures during nighttime in the rooms. Iwaro and Mwasha developed coconut fiber-filled masonry walls to check the insulating property [64]. They compared the results with no insulation and fiberglass filler walls to determine the coconut fiber's effectiveness as the insulator. They found a similar trend as the previously stated research as the energy consumption rate and the temperature during day time was significantly lower than the nighttime as the fiberglass showed slightly better performance during nighttime.

On the other hand, the air relative humidity was found to be lesser all through the day in comparison to no-insulator model, but the fiberglass again showed slightly better performance. Meanwhile, Brose et al. developed a coir-cement composite as insulator and that resulted in 0.16–0.19 W/mK reduction of thermal conductivity. They also found that the compressive strength and modulus of rupture of the coir-cement insulator was better, which would help reduction of out-of-plane failure during seismic events like earthquakes [65]. So, the examples of using coir as an insulator have not been that high, but all the attempts have shown improvement in the insulation property to a considerable degree.

#### 3.1. Applications in biocomposite materials production

Coir fiber has also been used in many other composite materials made for different purposes other than insulation. Coir fiber has mainly been used with many synthetic polymers for producing composite materials [45,66–71]. But these polymers don't produce sustainable composites as the polymers like polyester, polyethylene, polypropylene, etc., are neither sustainable nor biodegradable in nature. These materials are derived from petroleum sources. Even though these polymers have been started to be derived from natural vegetable sources these days as for example bio polyesters and bio polyethylene are made from carbohydrates now, and the source of these carbohydrates is corn, epoxy, and nylon are derived from oils of sunflower and palm tree, etc [33]. But these polymers have not been started to be used in the production of composite materials at a large scale yet. Only a few polymers such as polylactic acid, starch, polycaprolactone, cellulose acetate, etc., are used mainly in making fully biodegradable sustainable composites. These polymers have problems, such as poor tensile strength, processability, absorbency, etc [72–75]. In fact, the biodegradability property is responsible for some of these problems; for example, PLA contains an acetal functional group for which PLA is susceptible to water degradation, but without this in the structure, the biodegradability nature would depend on enzymatic degradation only, which is not possible in landfills or ocean water [76]. So, it would not be called biodegradable then. Again, all bio-derived polymers are not biodegradable, while all petrochemicals are not non-biodegradable (for example, polycaprolactone) [33]. Composites of some common biodegradable polymers are discussed below.

# 3.2. Coir fiber reinforced starch biocomposites

Starch is one of the world's most abundantly produced natural polymers [4]. It is a type of polysaccharide which is consisted of two polysaccharides, amylose and amylopectin. Amylose and amylopectin are  $\alpha(1 \rightarrow 4)$  glucan-linked linear and  $\alpha(1 \rightarrow 4)$  and  $\alpha(1 \rightarrow 6)$  glucan linked branched polymers respectively [77]. Amylopectin is more soluble and hydrophilic in nature, while amylose is slightly hydrophobic. But starch contains more amylopectin, for which starch is very easily soluble in water [4]. The key component of coir is cellulose, which is also a polysaccharide. Both contain hydroxyl groups in the structure, which can produce inter and intra-hydrogen bonds between cellulose, starch, and cellulose. That's why starch can be used successfully with coir fiber to produce biocomposites. Lomelí-Ramírez et al. produced coir fiber-reinforced cassava starch composites in two different experiments [78,79]. They mixed cassava starch with 30% glycerol in a plastic bag. Glycerol was used for plasticizing starch because starch is quite brittle in the dry state; plasticizer makes the starch thermoplastic and increases the extensibility of the starch paste [72,80–82]. The fibers and starch were mixed in an industrial mixer. They varied the fiber percentage from 0 to 30%. The composites were made in a hydraulic press at 160 °C under 410.4 kgf/cm<sup>2</sup> pressure, and the process was continued for 50 min. The addition of fiber increased tensile strength and Youngs's modulus from 1.56 MPa to 14.56 MPa–3.24 MPa and 59.81 MPa, respectively (for 30% fiber content). While on the other hand, water uptake capacity also decreased from over 250%–59.83% in the case of 30% fiber addition in the 4-h absorbency test, which shows the significantly high absorbency property of starch compared to a natural cellulosic fiber which is generally known as quite good absorbent materials. This high absorbency property of starch has always been one of the major problems of starch, which

has restricted its use in producing materials for practical uses [83]. Significantly poor tensile properties in another huge drawback of starch which was improved to a quite higher degree by the addition of coir fiber (Coir fibers have superior tensile properties) but still within the lower range compared to other common composite materials [4]. An increase in tensile modulus was an indicator of the composite's increased stiffness or hardness property, which needed greater force to cause deformation of the material. This was down to the rigid nature of starch resin which is quite brittle at the same time though, but the brittleness was reduced and covered by the use of plasticizer and the introduction of coir fibers in the structure, which helped starch to gain a solid shape rather than being a stiff but brittle material. The strong and stiffer nature of coir fiber also played its part in the formation of a structure like this.

# 3.3. Coir fiber reinforced polylactic acid (PLA) biocomposites

Polylactide or polylactic acid (PLA) is a biodegradable thermoplastic polymer derived from bio sources, such as corn. It is also called biodegradable thermoplastic polyester [84]. It is produced by polycondensation or ring-opening polymerization of lactide monomer in the presence of a lewis acid [85,86]. In industrial production, PLA is produced from starch, as starch is converted to dextrose, from which lactic acid is produced via fermentation. Then lactide monomer is prepared from that already produced lactic acid, and this lactide then undergoes polymerization [84]. Generally, PLA is known for its very good tensile properties, film transparency, and biodegradability, but it is a bit brittle and may force plasticizer use when required [84]. Since it is a biopolymer, it works very well with natural fiber-like coir in composite materials. Nam et al. developed coir fiber reinforced PLA composites [87]. They varied the fiber percentage from 10 to 50%, and they also varied the pre-treatment time and concentration of sodium hydroxide for treating the fiber. They mixed the dried, alkali-treated, and untreated fiber with PLA resin in a mixer at 180 °C and then formed pallets of coir fiber-reinforced PLA, which were dried at 70 °C for 12 h. Then the composite was produced by using those pallets in an injection molding machine. Alkali treatment resulted in the removal of waxy and dust contents from the fibers, which helped better adhesion between the fiber and resin molecules and improved the composites' tensile properties. They found the best results for 72 h of pre-treatment with a 5% concentration of sodium hydroxide. Reinforcing with 20% fiber gave the best tensile properties, as the maximum tensile strength recorded was about 59 MPa. Adding more fiber showed a decrease in tensile properties because of poor wetting of fiber in a comparatively low amount of resin and the least amount of bonding between resin molecules due to too much fiber in between the resin molecules, which is called the agglomeration of fibers. The tensile modulus increased with an increasing amount of fiber, though which means the addition of fiber made the composite more rigid, just as it did to the starch composites, as stated in the previous section. It is a further sign of the strong and rigid nature of the coir fibers. The thermal stability of PLA composite was also improved because of the addition of fiber. It was down to the considerable thermal properties and low thermal conductivity of coir fiber. Coir fibers are generally known for low thermal conductivity like other natural fibers due to the absorbent nature and the presence of hollows in the physical structure of the fibers [52]. The thermal resistance is down to the cellulosic materials, which undergo thermal degradation from 200 °C to 370 °C but alkali-treated fibers show significantly better resistance as the degradation starts above 330 °C and lasts around 370 °C due to increased crystallinity or the removal of the contents other than cellulosic matters which have better crystallinity and thermal properties compared to other contents of the fibers [62,87].

Dong et al. also found that the addition of 20% pre-treated fiber with PLA resin gives the optimum tensile and flexural properties (about 15 MPa tensile strength and about 45 MPa flexural strength) [88]. The fiber percentage varied from 5 to 30%. They pre-treated the fibers with 2% sodium hydroxide solution for an hour at 70 °C. The composites were prepared in a hydraulic press under 1.5 MPa pressure for a 15 min treatment at 180 °C temperature in a more like sandwich structure. But they found a decrement in thermal stability because of poor bonding between fiber and matrix, while the biodegradability rate was found to be higher than pure PLA because coir fiber is more easily degraded because of having high water absorbency. Jang et al. tried to produce flame retardant composite using coir fiber and PLA [89]. They pre-treated the fibers with oxygen plasma (discharge power 100 W) to modify the surface of the fibers to improve fiber-matrix adhesion. They varied the fiber percentage from 5 to 10%. The composites were prepared using a commingled yarn structure (a hybrid structure containing two materials in the shape of fiber to produce a continuous filament-type structure by using the air-jet [90]) method at 200 °C). Fiber treatment improved the tensile properties, and the addition of fiber also improved the tensile strength of PLA, as about 75 MPa tensile strength was recorded for 10% fiber addition, whereas the tensile strength of pure PLA was 60 MPa. Plasma treatment reduced the thermal shrinkage of the composite, while the addition of fiber also showed the same trend because of having better bonding between fiber and matrix. The flame retardancy was measured by testing the LOI (limiting oxygen index value), which was found to be almost similar for the treated and untreated fiber-reinforced composites to pure PLA, which mean these treatments do not have any positive impact on this property but also no negative impact was found either, and the resulted material would always be flame retardant regardless of fiber addition or non-addition because of having LOI above 20 [91].

# 3.4. Coir fiber reinforced polycaprolactone biocomposites

Polycaprolactone (PCL) is a type of aliphatic polyester synthesized by ring-opening polymerization of caprolactone [92], and it is degradable by enzymatic activities [93,94]. It is easily degraded by bacteria and fungi in the environment [95–98]. It is derived from petroleum, one of just a few polymers available right now. It is very much hydrophobic in nature, has a semi-crystalline structure, is soluble at room temperature, and is very easily processable due to having a very low melting point (about 60 °C) [99]. Its properties heavily depend on its molecular weight. If the molecular weight is lower, then the PCL would be a waxy solid material, while if the molecular weight is above 20000, then the PCL would be a very strong solid material and these materials generally show similar mechanical properties to polyethylene [93]. Because of a very low melting temperature, it is difficult to use alone; instead, it is used in

blends or composites more successfully. Obasi et al. produced coir fiber reinforced PCL composite using solvent casting technique [100]. They used PCL of 8000 molecular weight and brown coir fiber husk. The PCL was dissolved in Dichloromethane at room temperature, and then the fibers were mixed in the solution. The mixture was stirred for 4 h and then sonicated for 20 min, and then the mixture was poured onto a glass plate for drying. All these processes took place at room temperature. The fiber percentage varied from 2 to 5%. Tensile strength of the PCL decreased after fiber addition, though it was comparatively higher at 2.5% addition (12.99 MPa) than 2% and 5% fiber loading. This happened because of poor wetting of fiber with matrix and no interfacial bonding between fiber and matrix. Wu also produced a coir fiber-reinforced PCL composite [101]. But he used white coconut fiber, which was collected from green coconut. He varied the fiber percentage from 5 to 20%. He also used acetic acid-grafted PCL to improve the thermal properties of the PCL composite. The fibers and matrix were mixed in a blender, and the composites were made in a hot press. Results showed that the thermal properties of PCL changed very little with the addition of fiber or the process of acetic acid grafting. The tensile strength decreased (from 37.6 MPa, the strength decreased to 13.9 MPa while 29.8 MPa strength was recorded for 5% fiber loading) with the addition of fiber for both types of the matrix but grafted PCL showed comparatively better tensile strength (35.8 MPa for pure grafted PCL and 34.5 MPa at 5% fiber loading). It was due to better adhesion between grafted PCL and the fibers compared to the pure PCL.

# 3.5. Coir fiber reinforced other biocomposites

There are just a few other examples of sustainable coir fiber-reinforced biocomposites; polybutylene succinate composite can be classed as one of those. Polybutylene succinate (PBS) is also derived from petroleum. A polycondensation reaction between succinic acid and butanediol synthesizes it. The melting temperature of PBS is slightly higher than 100 °C (about 115 °C), and the degradation temperature was above 300 °C, which was almost similar to polyethylene and polypropylene [93,102,103]. Nam et al. Used PBS and produced coir fiber-reinforced PBS composite [103]. They varied fiber percentages from 10 to 30%. The fibers were pre-treated in 3–7% sodium hydroxide solution at room temperature. The duration of pre-treatment varied from 24 to 96 h. The tensile strength of coir fiber increased from 139 MPa to 238 MPa after alkali treatment, just as in the previously stated studies where alkali treatment improved tensile properties. The maximum strength was recorded for 5% sodium hydroxide treatment for 72 h. The composites were prepared in a hot press under 10 MPa pressure at 150 °C temperature for 10 min. The addition of fiber increased the tensile strength of the composite from 33 MPa to about 51 MPa. Maximum strength was found for 25% fiber addition while further increment of fiber percentage decreased the strength of the composite. Flexural strength also showed a similar trend. Alkali treated fiber reinforced composite showed better properties due to having more interfacial bondings than untreated fibers, just as the previous studies.

Polyvinyl alcohol (PVA) is quite a commonly used biodegradable polymer derived from petroleum sources [93] which is extensively used in the sizing of yarn for fabric production in the textile sector [104]. It is synthesized by polymerizing polyvinyl acetate in a free radical polymerization mechanism [93]. It is odorless, colorless, non-toxic, highly water-soluble, and highly resistant to oil and grease [105]. Kong et al. used PVA and produced a coir fiber-reinforced PVA composite [105]. They pre-treated the fiber with a 5% alkaline solution (made using sodium hydroxide) for 2 h at room temperature. They varied the fiber percentage from 0 to 20% while they produced the composites in a compression molding machine at 185 °C within 10 min. They found better tensile properties for treated fiber and the tensile strength was found on the rise with fiber loading (Young's modulus of 4072.81 and 5207.90 MPa for 15% alkali treated fiber (5450 MPa for 20% untreated fiber loading and 3680.72 MPa for 20% treated fiber loading) because of the increment of treated soft fiber compared to untreated hard fibers as soft fibers tend to take lesser space and allowed more amount of fibers because treated fibers are comparatively lighter than untreated fibers. It caused aggregation of fibers and reduced adhesion between fiber and matrix. On the other hand, alkali treated fiber loading resulted in poor thermal properties than pure PVA due to possible alkaline contamination in the fibers, while untreated fibers showed some improvement compared to pure PVA due to the naturally high thermal resistance of coir fiber [62].

Another natural biodegradable polymer is tannin. Tannin is a type of polyphenol. It has many phenolic groups in its structure [106, 107]. It is synthesized from various plants, barks of plants, etc [108]. Its molecular weight varies from 500 to 20000 [109]. Tannins can be of two types on the basis of the chemical structure and physical properties, hydrolyzable tannins and condensed tannins [110,111]. Hydrolyzable tannins can be hydrolyzed by acid, alkali, and enzymes, while condensed tannins are quite resistant to hydrolysis. Barbosa Jr. et al. produced a coir fiber-reinforced condensed tannin-phenolic composite [108]. Coir fiber of 3–6 cm length was used in random orientation in the composite. The fiber percentage varied from 30 to 70%. The composite was prepared using a compression molder. The addition of fiber increased the impact strength of the composite form from 0.016 KN to 0.096 KN. Further increment in fiber percentage decreased impact strength because interfacial bonding between fiber and matrix was found to be quite higher in these composites, just like the previous research.

# 4. Performance of coir fiber in reinforcing the composites based on different parameters

From the above examples, some common features can be noted, which are.

# 4.1. Effect of the type of fiber (surface treated or untreated)

Alkali treatment of fiber increases the tensile strength of the composite. This trend can be seen in Fig. 3(a and b) where Fig. 3(a) is showing the smooth surface of raw coir fiber while Fig. 3(b) is showing the roughened surface of the same fibers after being treated with alkali [87,88,103]. This is due to the increment in interfacial bonding between fiber and matrix molecules [71,74,87,103].

Actually, alkali treatment removes the impurities from fiber and exposes lignin to the fiber surface shown in Fig. 3, which makes the surface rougher, and it helps to create mechanical bonding between fiber and polymer [103].

But too much exposure to alkali can cause too much removal of cuticles and globular parts from the fiber, which makes the surface comparatively smoother, and results in comparatively weak mechanical bonding between fiber and matrix. That's why alkali treatment with 5% alkaline solution for three days at room temperature gave the best results among some different concentrations and different duration of treatment in different research. It gave the best mechanical properties of the composites. Other than this, an increment of tensile strength may have also happened because the fibers become softer and free of dust and waxy content. It also increases the roughness of the surface of the fibers, which helps the formation of mechanical bonds between fiber and matrix. It enables the matrix molecules to adhere to the fiber surface even without forming any chemical bonds, especially when high pressure is applied during composite production. And also, the improved crystallinity of the composite overall when the treated fibers are used due to the compact and highly oriented structure that has been formed by the improved adherence between fiber and matrix molecules also helps the tensile properties of the composites [62,87].

Fig. 4 contains two examples of PLA composites where the tensile properties varied even though similar products were used. It was down to the production process of the materials. The tensile properties of the pure PLA sheet were already lesser for the study of Dong et al. It happened because they used a multi-layered sandwich system instead of single-layer method, which tends to reduce the mechanical properties of the material due to slippage or easy debonding between the layers in the composite [112]. But it is not always the case, and exceptions have been seen very often. Fiber length can also be a reason which was shortest for the study of Dong et al. But the other studies did not use too long fibers either to establish this concept. Another difference found in these studies was the alkali treatment process which was of only an hour for the study of Dong et al. whereas the other two experiments used 72 h of treatment with 5% sodium hydroxide, which has very often been specified as a very fruitful ratio for alkali treatment. Meanwhile, the tensile strength of two PLA composites and Polypropylene composites has shown a different trend; it may be due to many reasons, including different processes of composite production, different fiber length, etc. The scanning electron microscope images showed that two composites of PLA have different types of cross-sections, as the amount of void in the composite made by Dong et al. was comparatively higher [88].

The alkali treatment has also increased moisture absorption of the composite due to improved absorbency properties of the fibers.



(a) Untreated raw fiber



(b) Alkali treated fiber

Fig. 3. Scanning Electron Microscopic images of untreated and alkali-treated fiber.



Fig. 4. Bar diagram of comparison of tensile strength in case of fiber treatment in different composites.

The introduction of fiber always creates voids in the structure where the water molecules can penetrate. It also affected the thermal characteristics of the composite, which is described later.

On the other hand, fiber reinforcement has not always improved tensile properties in comparison to pure polymer sheets. In fact, sometimes the tensile strength decreases when the fiber is added, as this trend is seen in Fig. 5, where it is clear that sometimes the tensile strength has been found to be less than the tensile strength of pure polymer sheet after being reinforced with fiber [69,79,87,88, 100,103,113]. However, some other characteristics like impact strength and biodegradability properties of the composite improved, which can be important in some cases according to the desired application of the products [87,108]. Decrement of tensile and other mechanical properties by loading small fibers that do not take part in the improvement of tensile or flexural properties but helped increase the stiffness of the composites, which affected the mechanical properties like tensile, and flexural, negatively [112]. Poor interfacial bonding between the fibers and matrix is another big reason, as found in several pieces of research. Fiber agglomeration can also be a big issue if the fiber percentage is not maintained properly.

Another important point can be the pre-treatment of the fibers. All the studies specified in Fig. 5, which had lesser success or lesser improvement or deterioration in some cases in terms of the tensile properties, had one thing in common, neither experiment used 5%



Fig. 5. Bar diagram of comparison of tensile strength increment percentage in different composites.

sodium hydroxide for pre-treatment or did not perform the process long enough or did not perform the pre-treatment at all. This matches the previous claim made in Fig. 4 also. The chemical bonding between fiber and resin is always difficult because both materials are generally found as chemically inactive materials and are considered good in terms of thermal resistance to any chemical changes. This makes the materials very unlikely to form any chemical bonding between the fibers and matrix. The Fourier-transform infrared spectroscopy of a few of these studies showed that no specific bonds were created between fiber and matrix. This led to the deterioration of mechanical properties apart from the cases where proper pre-treatment was used, which helped the formation of mechanical bonds between the fibers and matrix polymers. Also, it has been reported in previous studies that the incorporation of a similar type of fiber and matrix can give better results due to better bonding between fiber and matrix molecules [45]. It helped in some cases only when proper pre-treatment was performed. This problem has also been sorted in some research where the fibers have been treated for 72 h with 5% sodium hydroxide solution [62].

# 4.2. Effect of the type of resin

Natural biodegradable polymer composites have shown better mechanical properties than synthetic polymer composites, as shown in Figs. 6 and 7 [45,67,74,87,88,103,113,114]. This may be due to better adhesion between natural, biodegradable polymers and fibers than synthetic (non-biodegradable) ones, as it is already known that natural fibers don't generally work that well in association with synthetic polymers unless being undergone surface treatment for reducing the hydrophilic nature of the fiber [45]. Also, the tensile strength of pure PLA and PBS is higher than pure polyethylene and polypropylene, as shown in Fig. 5. So better strength of PLA, PBS, etc., composites was expected.

So, according to this figure, the tensile properties and improvement of any mechanical properties depend heavily on the properties of the resin material. In fact, it is always expected to get comparatively poor mechanical properties when natural fibers are used with synthetic materials because the anisotropic and non-homogeneous composite materials produced from these combinations due to totally different types of strength, shrinkage, and surface properties of the resin and reinforcement are destined to fail to improve mechanical properties [115]. These combinations generally result in better biodegradability, lesser cost of the product, etc [112]. But still, better properties are found very often whenever surface treatments or any other means are used, in addition to improving the bonding between the fiber and resin polymers.

# 4.3. Effect of fiber on the thermal properties of the composite

The thermal properties of coir fibers are quite unique, and the composites made using coir fibers have also shown the properties associated with coir fibers. The fibers have very good thermal resistance, low thermal conductivity, and better resistance to weight loss at heating. These properties are also found in the composites made with coir fiber in considerable amounts. Hasan et al. used long coir



Fig. 6. Bar diagram of comparison of tensile strength in different coir fiber-reinforced composites.



Fig. 7. Bar diagram of comparison of flexural strength in different coir fiber-reinforced composites.

fiber and short fibrous chips collected from coconut husk with melamine-urea-formaldehyde, and the resulting composites showed a gradual increase in heat conductivity when the percentage of melamine-urea-formaldehyde was increased (from 8% to 12%) [62]. The results were down to decrement in the amount of coir fiber in the structure, but at the same time, some other important factors played vital roles. For example, the density of the composites increased with the increasing amount of resin (due to better bonding between the fibers), which reduced the voids within the structure and the percentage of air within the structure. Since air is a bad conductor of heat, its presence reduces the heat conductivity of the material, while the absence of air increases the conductivity of the composite [116].

Besides, the TGA (thermogravimetric analysis) results showed the percentage of weight loss of the material with increasing temperature. The weight loss percentage was found in lower amounts when the composites contained higher amounts of coir contents which were also evident in previous research reports [117]. The results actually revealed several important points. The degradation occurred in three steps [118,119]. The first step of the degradation took place around 100 °C due to the loss of moisture from the structure. The second step took place between 230 and 300 °C due to the degradation of hemicellulose while the cellulose content degraded between 300 and 400 °C. The percentage of char or residue depended on the presence of lignin in the composite as a higher percentage of coir content presented more lignin in the structure, which resulted in more residue percentage (around 40% for only coir contents which was reduced to around 30% when 8–12% melamine-urea-formaldehyde was introduced) after the completion of the heating cycle. They also reported the composites showed a slightly higher percentage of weight loss during the first step due to the high absorbency of the coir contents, especially after alkali treatment which removed the impurities from the coir as reported before [120]. However, the final properties of the material would depend on both the structure's resin and coir fiber contents. In the case of a resin with better thermal resistance or low heat conductivity than coir fiber (e.g., PLA, wool, etc.), the thermal conductivity of the composite would be reduced with the addition of coir fiber content in the structure.

In terms of composites made of biopolymers and coir have not been many in previous years where the thermal properties have been studied. A few pieces of research were done on coir-reinforced PLA composites, which showed the same trend as stated above. Sun et al. developed PLA composites using 3–7% coir fiber (hydrogen peroxide and alkali treated) and found that the addition of coir fiber increased the production of residue or char in TGA results which matches the explanation given above [121]. The same trend was followed by Nam et al. [87]. But they also showed that PLA has slightly better thermal resistance than coir, especially at a lower temperature when the coir fiber goes through two degradation steps. But at high temperatures, the PLA degrades about 97% of its weight while the presence of coir fiber increases the residue percentage, which improves the thermal property of the material slightly. They also reported that the alkali-treated coir fiber produced a higher amount of residue in the end because of having a lower amount of impurities in the structure, as most of the impurities are removed in the alkali treatment stage from the fibers. In that sense, the treated coir fibers are expected to show better performance in terms of flammability as the formation of a high percentage of char or residue generally increases the thermal or flame resistance of the material because the flame needs heat to continue burning, but the presence of char or residue absorbs heat which reduces the intensity of the flame and eventually reduces the flame [2]. Jang et al. showed exactly that as they found that the LOI (limiting oxygen index: the required amount of oxygen for a material to burn [2]) of alkali-treated coir fiber composite (PLA composite) was slightly higher than the untreated coir fiber composite [89].

So, from the above discussions, a few points can be found.

- Coir fiber has poor thermal conductivity properties, which can help these being used as insulating material.
- Coir fiber undergoes three-step degradation during heat treatment in TGA, which reduces its ability to withstand lower temperatures compared to other materials due to its first two steps of degradation generally occurring by 300 °C. But at high temperatures, the temperature resistance of coir is slightly better because of its ability to produce a high amount of residue or char during heating.
- Due to having better char or residue-forming ability, the coir fiber can be a good option for resisting flame.
- Alkali treatment reduces the percentage of impurities in the structure, which can lead to a slightly high percentage of weight loss in the first step of degradation but increase the formation of the residue.

#### 4.4. Reasons behind low production of biocomposites

Even though the above-mentioned points have some proof to show here, these still cannot be taken as guaranteed because these are very small sample sizes from which a final result can be given. But these points may be helpful for further research. These assessments may give a wrong concept as it is clear here that polymers like PLA, PCL, and PBS can be very good alternatives to synthetic polymers, but still, it may seem baffling that's why these polymers are not being used in all sectors instead of synthetic polymers. It is mainly due to the cost of natural polymers, as shown in Table 2 below.

As it can be seen here, natural polymers are way costlier than synthetic polymers. Reinforcing these polymers with cheap fibers can be a very good option; in that case, fibers with a huge production rate will be the best option. That's why coir fiber has been selected for many studies, as it is produced in large quantities worldwide. This fiber is derived from coconut waste, making it cheaper than other fibers. This makes it perfect for reinforcing the natural polymers to produce sustainable biocomposites. In fact, the price is always expected to be higher for composite materials because of the added options like composite making process, labor cost, etc. So, it is essential to reduce costs and the number of processes. And it is clear that coir fibers are generally added to reinforce or to reduce cost as it is an inexpensive product due to very easy retting with only water, but a high percentage of fiber is recommended to achieve these principles properly. Since coir is one of the lighter fibers among the natural fibers, its use as reinforcement and to reduce the cost of the composite material is well justified. But due to poor mechanical properties compared to other commonly found natural fibers, the composites made of coir are not expected to be used in many industrial applications where mechanical properties are of the greatest interest [103]. Still, due to its very low cost, it can be very useful in products of daily use.

Another issue to be discussed is the sustainability of the composites discussed in this study. The term 'sustainable biocomposites' was used at the beginning of this review, which needs quite a few specifications to be fulfilled. As stated earlier, sustainable biocomposites must be fully biodegradable, easily produced, cheaper, and environmentally friendly. The cost has already been mentioned, and, from that point of view, the bio-polymers fall quite far back from the synthetic polymers. In that sense, adding fiber and reducing the material cost is a very good option. Another point would be the bio-degradability or the environment-friendliness of the materials. Table 3 below shows the degradability nature of the common bio-polymers.

Table 3 above shows that all the bio-polymers discussed in this study are biodegradable. The duration may vary for different polymers, but all those are biodegradable. In that sense, the bio-polymers tick the box for being considered for sustainable production, as all other synthetic polymers are non-biodegradable and may take several hundred or even thousand years to degrade. But the mechanical properties of all the bio-materials are not up to the mark, though, as shown in the table, for which these can not be used in just any type of application if desired.

The next point is the production or abundant production of the material, and from that point of view, starch is the most easily and abundantly found polymer among all the bio-polymers discussed in this study [83]. But starch-based resins have significant problems in terms of mechanical properties. PLA is a new concept compared to other common polymers produced from starch sources. It is also quite easily found in that sense, but since the production process is not fully established yet in terms of the sustainable system, just like the other bio-polymers of this study [128]. But in comparison with the common synthetic polymers, the bio-polymers lack in terms of quantity and a well-established sustainable production process. But still, the bio-polymers offer a far greater advantage of sustainable production in comparison to synthetic polymers. The greenhouse gas emission data can give the clearest idea of any polymer's environmental impact. Table 4 below will show the greenhouse gas (GHG) emission data for different polymers.

The GHG emission data shows that the specified bio-polymers are more environmentally friendly in terms of GHG emissions. Starch-based resin is the best among these, while PLA is also at the lower end of the chart. It is said that the other synthetic materials are

Table 2	
Cost of different polymers	[122].

Polymer Resin	Cost Range (\$/Pound)
Polylactic Acid	2–2.5
Starch based Resin	1–1.25
High Density Polyethylene	0.2-0.5
Low Density Polyethylene	
Polypropylene	
Polyester	
Poly Carbonate	1.2–1.5
Polyvinyl Alcohol (PVA)	

#### Table 3

Bio-degradation nature of different polymers [123-127].

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Polymer Resin	Duration for Bio- degradation	Nature of Degradation	Feature
Polylactic Acid	Several months or more	Not easily bio-degradable, needs specific condition.	Good mechanical properties but brittle in nature
Starch-based Resin	About a month or more	Degrades by biological means	Brittle and poor mechanically, but very cheap.
Poly Caprolactone (PCL)	Several months or more	Degrades very slowly	Poor mechanical properties but good resistance against chemicals.
Poly Butylene Succinate (PBS)	A few months	Degrades by biological means	Thermally resistant but stiffness is insufficient.

already being synthesized using the most environmentally friendly and sustainable system, while the PLA synthesis is still under modification to find the optimum level [128]. So, from this point of view, the bio-polymers pass the sustainability mark, while adding coir fiber in these resins further improves the GHG emission.

The last point to be considered would be the properties of the biopolymers. The tensile properties of different polymers are stated in Table 5 below.

Table 5 shows that, among the bio-polymers, PLA is the best one that can match or even go beyond the tensile properties of any common synthetic polymers. The other option would be PBS. Then PCL and starch would follow afterward. But by comparing all the previously mentioned properties and other features, PLA seems to be the most suitable option from all points of view except the high price. The price can be compromised by using coir fiber as reinforcement. These two can match quite well as both have a similar density (about 1.25 gm/cc [103,124]) while the tensile properties of coir fiber are quite good too, even though not as good as other fibers like jute, flax, etc [103]. And with some further optimization in the production procedure, both the price and GHG emission of the PLA can be taken up to the required level, and the addition of coir fiber would help the process as it would reduce the cost and GHG emission further and at the same time, the possible deterioration of by the addition of the fiber can be compromised as PLA generally has better tensile properties than other common polymers, so a bit deterioration would not be too detrimental for the final product. And thus, the production of sustainable biocomposite can be justified. Other than that, the starch-based resin can also be used when biodegradation is far more important than tensile properties, especially for our daily-use products, which are used once or twice in the product's lifetime. And the addition of cheap coir fibers would help produce cheaper materials, and thus sustainable biocomposite can also be produced.

# 5. Conclusion

This study has been done to learn about the thermal insulating property and the performance as reinforcing material in composite structures of coir fiber, a fiber that is produced from coconut waste and found in large quantities all over the world. It is a lignocellulosic fiber, but it doesn't have all the best properties that are found among natural fibers. Actually, its low cost, thermal insulating property, and relatively fair mechanical properties make it suitable for composite material. It has been used successfully in quite a few biopolymer composites discussed earlier. The results of those research again showed that it is possible to produce better and more sustainable materials from natural, biodegradable polymers. A few composites under discussion have shown better thermal resistance and insulating properties which have been used in civil engineering sites. The description of the coir structure also indicated the coir's ability as an effective, low-cost heat insulator that can be used in practical life. But the major drawback is the price of biopolymers, which can be solved if fibers like coir, found as a waste material, are used with those polymers in composites. Some factors like alkali treatment, use of suitable treatment time, use of suitable resin, etc., that impact the mechanical properties such as GHG emission, tensile properties, nature of the degradation process, thermal properties, etc., the use of bio-polymers to produce coir fiber-reinforced sustainable biocomposites has been justified in this study. The results are not up to the optimum level yet, but with some further improvement, the level can be reached in the near future, and those modifications will have to be the production of the bio-polymers oriented, which will help to reduce the cost further as well as the GHG emission to a far more tolerable level.

# Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

#### Data availability statement

Data included in article/supp. material/referenced in article.

# **Financial information**

There is no financial information to share regarding this study.

#### Table 4

GHG Emission	for	different Pol	vmers	[128-131	1.
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Polymer Resin	GHG Emission (kg $CO_2$ equivalent/kg material)
Polylactic Acid	1.7
Starch based Resin	0.6
Poly Butylne Succinate (PBS)	2–2.3
High Density Polyethylene	2.6
Low Density Polyethylene	2.9
Unsaturated Polyester	7.6
Epoxy Resin	6.7

#### Table 5

Tensile Properties of different polymers [4,83,103,124,132-137].

Polymer Resin	Tensile Strength (MPa)	Tensile Modulus (MPa)
Polylactic Acid	21–60	270–1600
Starch based Resin	1–5	25–35
Poly Butylne Succinate (PBS)	15–35	400–500
Polycaprolactone (PCL)	16	400
High Density Polyethylene	15–26	600–900
Low Density Polyethylene	10-20	300-400
Unsaturated Polyester	25-40	900–920

#### Statement of the conflict of interests

The authors can clarify that there is no relation of the authors with any subject discussed in this paper which can cause any conflict of interests.

#### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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