



Review

## Environment friendly, renewable and sustainable poly lactic acid (PLA) based natural fiber reinforced composites – A comprehensive review



G. Rajeshkumar<sup>a, \*\*</sup>, S. Arvindh Seshadri<sup>a</sup>, G.L. Devnani<sup>b</sup>, M.R. Sanjay<sup>c\*</sup>, Suchart Siengchin<sup>c</sup>, J. Prakash Maran<sup>d</sup>, Naif Abdullah Al-Dhabi<sup>e</sup>, Ponnurugan Karuppiah<sup>e</sup>, Valan Arasu Mariadhas<sup>e</sup>, N. Sivarajasekar<sup>f</sup>, A. Ronaldo Anuf<sup>g</sup>

<sup>a</sup> Department of Mechanical Engineering, PSG Institute of Technology and Applied Research, Coimbatore, Tamil Nadu, India

<sup>b</sup> Department of Chemical Engineering, Harcourt Butler Technical University, Kanpur, Uttar Pradesh, India

<sup>c</sup> Natural Composites Research Group Lab, Department of Materials and Production Engineering, The Sirindhorn International Thai-German Graduate School of Engineering, King Mongkut's University of Technology North Bangkok, Bangkok, Thailand

<sup>d</sup> Department of Food Science and Nutrition, Periyar University, Salem, Tamil Nadu, India

<sup>e</sup> Department of Botany and Microbiology, College of Science, King Saud University, P.O. Box 2455, Riyadh, 11451, Saudi Arabia

<sup>f</sup> Laboratory for Bioremediation Research, Unit Operations Laboratory, Department of Biotechnology, Kumaraguru College of Technology, Coimbatore, Tamil Nadu, India

<sup>g</sup> Department of Biotechnology, Kamaraj College of Engineering and Technology, Virudhunagar, Tamil Nadu, India

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### ABSTRACT

The Environmental concern and awareness around the globe have led to the development of sustainable bio composites which are derived from renewable resources. Biodegradable polymers and natural fibers derived from different renewable resources have played a vital role in the manufacture of bio composites. Poly lactic acid or polylactide (PLA) is one of the versatile aliphatic linear thermoplastic biodegradable polymers obtained from fully renewable sources such as wheat, corn, rice and sweet potato, and it has unique characteristics like renewable, sustainable, biocompatible and compostable. PLA has distinct advantages like low energy consumption and emission of low greenhouse gas during production and suitable for 3D printing applications. It also has some demerits such as low gas and water barrier properties, poor toughness, low glass transition temperature and is hydrophilic in nature, which limit its use in commercial applications. To overcome this, PLA is blended with various natural fibers in order to improve the thermal, water barrier, crystallization, mechanical, antimicrobial and degradability properties. Moreover, inclusion of natural fibers not only decreases the cost of PLA products but also helps in producing good competitive commercial products which are used in different industries. Hence, this review focuses on the synthesis and degradation of PLA, its applications in various sectors and manufacturing methods involved in PLA composites. Moreover, this review discusses about the different types of natural fibers and their influence on the unique properties of PLA based natural fiber reinforced composites. The overall aim of this paper is to provide a holistic idea about PLA based bio composites to academicians, industry personnel and researchers.

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### 1. Introduction

The continuous advancements in science and technology have resulted in the increase in demand for natural resources all over the world (Rangappa et al., 2020). This development has led to concerns such as scarcity of materials and conservation of environment (Thyavihalli Girijappa et al., 2019). In addition, the rapid depletion of oil reserves, greenhouse emissions due to the extensive usage of

petroleum-based products and their non-biodegradability, have triggered researchers to explore novel materials that are biodegradable, renewable and recyclable (Scaffaro et al., 2018a). Biodegradable composites are one such material that can be employed to address these issues and simultaneously ensure a sustainable environment. These composites contain matrix materials that are derived from agricultural and forestry feedstock and they are reinforced with cellulose fibers (Campilho, 2015). End-of-life disposal is not an issue when bio

\* Corresponding author.

\*\* Corresponding author.

E-mail addresses: [grajeshkumar.me@gmail.com](mailto:grajeshkumar.me@gmail.com) (G. Rajeshkumar), [mavinkere.r.s@op.kmutnb.ac.th](mailto:mavinkere.r.s@op.kmutnb.ac.th), [mcemrs@gmail.com](mailto:mcemrs@gmail.com) (M.R. Sanjay).

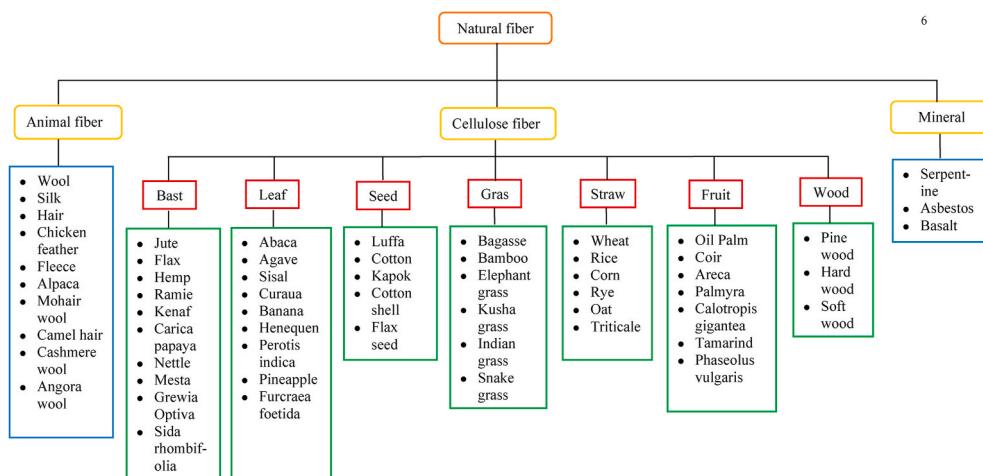


Fig. 1. Classification of natural fibers.

composites are used as they compost naturally without affecting the ecology (Gurunathan et al., 2015; Sanjay et al., 2018). In comparison with conventional petroleum-based synthetic composites, the usage of bio composites has unique benefits such as recyclability, renewability and biodegradability, low density and less expensive (Gholampour and Ozbaakkaloglu, 2020), which are essential characteristics for commercial applications. Moreover, they do provide adequate acoustic and thermal insulation and are flexible in nature. In addition, bio composites are non-toxic which means they do not cause any ailments and hence, are safer to use when compared to the synthetic composites (Hemath et al., 2020). Due to these advantages, they have a wide range of applications in various industries such as automobile, aerospace, construction, defense, biomedical, packaging, sports goods, etc. (Gurunathan et al., 2015; Vinod et al., 2020).

There are different naturally occurring and synthetic biopolymers such as starch, soy resin, gelatin, poly-hydroxy butyrate (PHB), poly-hydroxy butyrate-co-valerate (PHBV), poly lactic acid (PLA), etc. that can act as a matrix material for a bio composite (Cheung et al., 2008a, 2008b; George et al., 2020). Among these, poly-lactic acid has cited to be a feasible option to substitute synthetic plastics that are processed from petroleum by-products. PLA is renewable and biodegradable, and is obtained from agricultural crops like sugar beet, corn, etc. It possesses good mechanical strength, considerable stiffness and its composites can be fabricated by traditional manufacturing methods (Scaffaro et al., 2018b, 2020; Bher et al., 2019). In addition, life cycle assessment of PLA states that it requires nearly one tenth of the resin fossil energy only when compared to synthetic polymers (Gowda et al., 2018; Madival et al., 2009). Hence, these factors make PLA to be an ideal choice for fabricating biodegradable composites.

This review discusses about various recent developments in the area of biodegradable composites, specifically PLA reinforced with cellulose fibers. It focuses on the influence of natural fiber reinforcements on the properties of PLA namely; static and dynamic mechanical, thermal, tribology and vibration. This review also discusses about the number of cellulose fibers that are available to be used as reinforcements in PLA based composites. In addition, the manufacturing methods that are currently used to fabricate PLA bio composites are also detailed. The overall aim of this paper is to provide a holistic idea about PLA based green composites to academicians, industry personnel and researchers.

## 2. Cellulose fibers

Generally, a composite consists of two major constituents; matrix and reinforcement. Polymers are generally lightweight, but they lack the stiffness, strength, and dimensional stability needed for load-bearing and structural applications. Therefore, they are reinforced with fibers

Table 1

Producers and amount of production of different natural fibers all over the world (Balaji et al., 2014; Biswas et al., 2001; Faruk et al., 2012; Foulk and McAlister, 2002; Jayavani et al., 2016; Li et al., 2000; Mamun et al., 2013; Pandey, 2007; Shahzad, 2012).

Fibers	Producers	Amount of production ( $\times 10^3$ ton)
Wheat	China, France, India, Russia, USA	772,000
Rice	Bangladesh, Brazil, China, India, Indonesia, Myanmar, Pakistan, Philippines, Thailand, Vietnam	668,000
Bagasse	Australia, Brazil, China, India, Thailand, USA	75,000
Bamboo	Chile, China, Ecuador, India, Indonesia, Japan, Myanmar, Nigeria, Pakistan, Philippines, Sri Lanka	30,000
Oil palm	Indonesia, Malaysia, Nigeria, Thailand	30,000
Cotton	Brazil, China, India, Pakistan, Turkey, USA, Uzbekistan	25,000
Chicken feather	Worldwide	5000
Jute	Bangladesh, India, Myanmar, Nepal	3450
Flax seeds	Canada, China, India, Kazakhstan, Russia, USA	2930
Wool	Argentina, Australia, China, Iran, New Zealand, Russia, UK, Uruguay	2100
Coir	Brazil, India, Indonesia, Philippines, Sri Lanka, Thailand, Vietnam	1200
Kenaf	China, India, Malaysia, Mexico, Thailand, USA, Vietnam	970
Agave	India, Mexico	950
Flax	Belgium, China, France, Netherlands, Poland, Russia	830
Sisal	Brazil, China, Cuba, Haiti, India, Kenya, Madagascar, Mexico, Sri Lanka, Tanzania	378
Ramie	Brazil, China, India, Laos, Philippines	280
Hemp	Chile, China, France, Germany, UK	214
Silk	Brazil, Bulgaria, China, Egypt, India, Madagascar, Thailand, Turkmenistan, Uzbekistan, Vietnam	150
Kapok	China, Indonesia, Malaysia, Philippines, South America, Thailand	101
Abaca	Ecuador, Philippines	70
Cashmere wool	Australia, China, India, Iran, Mongolia, New Zealand, Pakistan, Turkey, USA	20
Alpaca	Bolivia, Chile, Peru	7
Mohair wool	South Africa, USA	5
Angora wool	Argentina, Chile, China, Czech Republic, France, Hungary	3

**Table 2**

Natural fibers' merits and demerits (Jawaaid and Khalil, 2011; Sanjay et al., 2016, 2019).

S. No.	Merits	Demerits
1.	Low density results in high specific properties	It has lower mechanical properties compared to synthetic fibers
2.	Energy requirement and greenhouse gas emissions are low during production It has good electrical and acoustic insulation properties Low cost and abundant in availability	Properties are dependent on different environmental factors It has low thermal resistance It has low durability
5.	It is biodegradable in nature	It is hydrophilic and absorption of moisture deteriorates its properties
6.	Eco-friendly processing is possible	There is poor interfacial bonding between matrix and fibers. Fibers require surface treatment to enhance matrix/fiber adhesion.
7.	Thermal recycling of fiber is possible	It lacks flame retardant properties

which possess high strength and stiffness (Mohanty et al., 2002; Sanjay et al., 2021).

Natural fibers are broadly divided into three different groups: animal fibers, cellulose fibers and mineral fibers as shown in Fig. 1 (Bajpai et al., 2014; Madhu et al., 2019a, 2019b). The amount of natural fiber produced all over the world is presented in Table 1. Among all the natural fibers, cellulose fibers are predominantly used as reinforcement for polymer matrix composites (Dwivedi and Chand, 2009; Hammajam, 2019; John and Anandjiwala, 2009; Ray et al., 2009; Scarponi et al., 2009). The cellulose fibers are extracted from different parts of a plant like leaves, stems, fruits, seeds, etc. The properties of the fibers depend on location of the crop, type of plant species, climatic conditions, soil type, availability of water, age of fibers, mode of transportation of fibers and storage conditions (Dittenber and Ganga Rao, 2012; Rajeshkumar et al., 2016; Thakur and Thakur, 2014).

Plant fibers contain three important chemical constituents: cellulose, hemi-cellulose and lignin (Nagarajan et al., 2021). In addition, waxes and other water-soluble compounds are also present. Cellulose is a linear 1,4- $\beta$ -glucan polymer that has repeating units of D-anhydroglucose ( $C_6H_{11}O_5$ ), which contains hydroxyl groups. It is the strongest and stiffest component of the fiber and it determines the mechanical properties of the fiber. Cellulose is hydrophilic in nature due to the presence of hydroxyl groups that form inter- and intra-hydrogen bonding (Ramamoorthy et al., 2015). On the other hand, thermal degradation, biodegradation and moisture absorption of the fibers are taken care by hemi-cellulose (Saheb and Jog, 1999). Lignin is a phenolic compound and acts as a structural support material in plants. It is attributed to high carbon content and low hydrogen content. Even though, majority of functional groups that make up the lignin molecules are identified, the chemical structure of lignin remains obscure. Lignin is hydrophobic and has an amorphous structure in the solid state (Hatakeyama and Hatakeyama, 2009; Mohanty et al., 2000).

For the past few decades, there is an ever-increasing interest towards natural fibers as reinforcement among academia as well as industries. This is due to their peculiar properties such as low cost, availability, lightweight, biodegradability, renewability, and good electrical insulation and acoustic properties (Rajeshkumar et al., 2017; Rajeshkumar, 2020a; Sanjay et al., 2019). In addition, replacing synthetic fibers with natural fibers provides significant environmental benefits (Sumrith et al., 2020; Vinod et al., 2021). It was shown that replacing 50% of glass fiber reinforced composites with natural fiber reinforced composites in North American automobiles led to a saving of 3.07 million-tonnes of CO<sub>2</sub> emissions and 1.19 million m<sup>3</sup> of crude oil consumption (Pervaiz and Sain, 2003). The natural fibers' merits and demerits are described in Table 2.

Polymer materials have several advantages such as low weight, chemical resistance, better mechanical properties, good thermal electrical insulation, etc. However, these properties are not enough to use polymers in structural and semi-structural applications. Therefore, there is a need to reinforce it with fibers to enhance its properties. In this context, natural fibers are an eco-friendly option. The physio-chemical and mechanical properties of different natural fibers are presented in Table 3.

### 3. Poly-lactic acid

Scientists and industry personnel are continuously working to replace synthetic polymers with biodegradable ones due to the rapid rise in the ecological, environmental, and safety challenges (Jamshidian et al., 2010a, 2010b). A bio-polymer or a bio-plastic is a polymer which is manufactured from a renewable or natural source (Rudin and Choi, 2012). A classification of bio-polymers is given in Fig. 2. A leading candidate among these bio-polymers is polylactic acid (PLA).

PLA is a thermoplastic polymer with high strength and modulus that can be made from renewable resources such as corn, sugar beets, etc. It belongs to the family of aliphatic polyesters which are generally made from  $\alpha$ -hydroxy acids like polymandelic or polyglycolic acid and are considered compostable and biodegradable (Garlotta, 2001). Its chemical structure is shown in Fig. 3. Lactic acid, which is optically active, has two stereo-isomers, L- and D-isomers. The L-lactic acid rotates the plane polarized light in clockwise direction whereas D-lactic acid rotates in counterclockwise direction (Rasal et al., 2010). The ratio of L-isomer to the D-isomer of lactic acid plays a crucial role in the crystallinity, degradation and processing behavior of PLA. The stereo-chemical structure of PLA can be altered by polymerization of a controlled mixture of L- and D-isomers to obtain a high molecular weight semi-crystalline or amorphous polymer whose melting point ranges from 130 to 180 °C (Grijpma and Pennings, 1994; Kolstad, 1996; Witzke, 1997). Pure PLA is in semi-crystalline form but polymers prepared from meso or racemic-lactide are usually amorphous. In addition, crystalline polymers can also be obtained by using stereo-selective catalysts (Baillie, 2005).

#### 3.1. Synthesis of PLA

There are a variety of methods to produce PLA. These methods include ring opening polymerization, poly-condensation, azeotropic dehydration and enzymatic polymerization (Enomoto et al., 1995; Garlotta, 2001). Out of these methods, direct condensation of lactic acid and ring opening polymerization are the most common processes.

Cargill Dow LLC, which is the largest manufacturer of PLA in the world, developed a patented, low cost continuous process for the production of lactic acid-based polymers (Drumright et al., 2000). In this process, the synthesis of PLA takes place in melt rather than in solution, thus providing substantial environmental and economic benefits. Initially, natural or renewable resources like corn, sugar beet, potatoes, etc., are transformed into dextrose. The dextrose undergoes a fermentation process to yield lactic acid. The lactic acid is converted into lactide in the presence of a catalyst. The molten lactide is further purified with the help of vacuum distillation. It is then converted into PLA by polymerization reaction. The remaining lactide monomer is then recycled within the process. The life cycle of PLA is shown in Fig. 4.

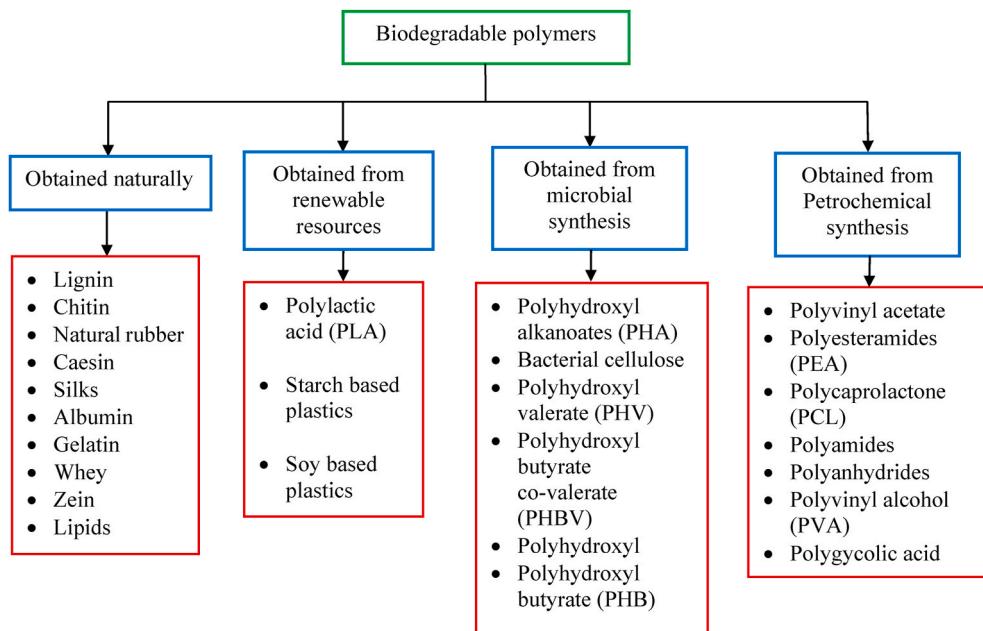
#### 3.2. Sustainability of PLA

As detailed in the aforementioned section, PLA is an eco-friendly polymer due to its biodegradability and recyclability. In addition to these merits, the production and usage of PLA results in significant energy savings and reduced greenhouse gas emissions. These factors are pivotal to ensure a sustainable environment (Scaffaro et al., 2019). The gross energy use for the production of PLA is 75.4 MJ/kg. This includes the complete process i.e., from the growth of corn to the shipment of PLA

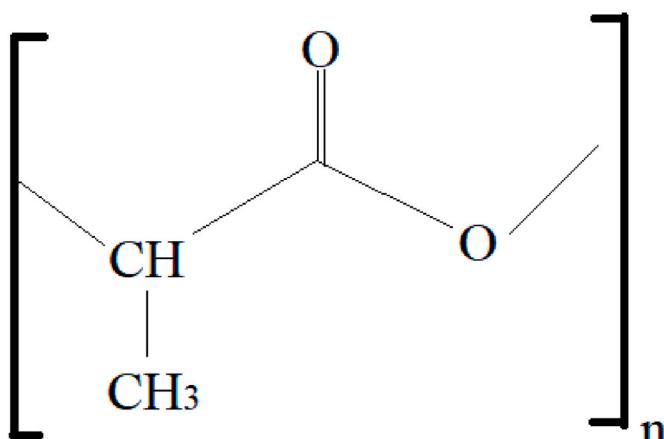
**Table 3**

Physical, chemical and mechanical properties of natural fibers.

Fibers	Density (kg/m <sup>3</sup> )	Diameter (μm)	Cellulose (wt.%)	Hemicellulose (wt.%)	Lignin (wt.%)	Wax (wt.%)	Ash (wt. %)	Moisture (wt.%)	Tensile strength (MPa)	Young's modulus (GPa)	Elongation (%)	References
<i>Furcraea foetida</i>	778	12.8	68.35	11.46	12.32	0.24	6.53	5.43	623.52 ± 45	6.52 ± 1.9	10.32 ± 1.6	Manimaran et al. (2018a, 2018b, 2018c, 2018d)
<i>Coccinia grandis</i>	1243 ± 22.64	27.33 ± 0.3789	62.35	13.42	15.61	0.79	4.388	5.6	273 ± 27.74	10.17 ± 1.261	2.703 ± 0.2736	Senthamarikannan and Kathiresan (2018)
<i>Acacia leucophloea</i>	1385	–	68.09	13.6	17.73	0.55	0.08	8.83	317–1608	8.41–69.61	1.38–4.24	Arthanarieswaran et al. (2015)
<i>Cissus quadrangularis</i>	1220	770–870	82.73	7.96	11.27	0.18	–	6.6	4157 ± 1224	130 ± 44	6.46 ± 2.88	Indran and Raj (2015)
<i>Heteropogon contortus</i>	602	–	64.87	19.34	13.56	0.22	–	7.4	476 ± 11.6	48 ± 2.8	1.63 ± 0.06	Hyness et al. (2018)
<i>Dichrostachys cinerea</i>	1240	–	72.4	13.08	16.89	0.57	3.97	9.82	873 ± 14	–	–	Baskaran et al. (2018)
<i>Ipomoea staphylina</i> plant	1401	–	72.26	13.6	19.56	1.51	1.40	8.28	173–658	–	2.03–6.63	Santhanam et al. (2016)
Kusha grass	1102.5	70–100	70.58	–	14.35	1.52	2.46	8.01	–	–	–	Balaji et al. (2016)
Cotton shell	–	–	53.86	11.43	16.97	0.59	7.22	9.93	–	–	–	Rajkumar et al. (2016)
<i>Carica papaya</i>	943	–	58.71	11.8	14.26	0.81	4.7	9.73	530 ± 11.2	–	1.62 ± 0.02	Saravana Kumaar et al. (2019)
<i>Pithecellobium dulce</i>	865 ± 26	144.3 ± 46.63	75.15 ± 0.26	10.23 ± 3.45	12.14 ± 0.22 ± 1.56	0.22 ± 0.08	2.13 ± 0.56	6.24 ± 1.26	654.28 ± 36	6.81 ± 1.7	10.52 ± 1.4	Manimaran et al. (2018a, 2018b, 2018c, 2018d)
<i>Lagenaria siceraria</i>	1216	–	79.91	12.69	7.62	0.31	0.92	9.96	257–717	7–42	1.38–4.67	Saravanan et al. (2016)
<i>Azadirachta indica</i>	740	–	68.42	13.72	13.58	0.43	–	–	–	–	–	Manimaran et al. (2018a, 2018b, 2018c, 2018d)
<i>Phaseolus vulgaris</i>	–	–	62.17	7.04	9.13	0.36	9.02	6.1	–	–	–	Gurukarthik Babu et al. (2019)
<i>Sida cordifolia</i>	1330 ± 20	–	69.52 ± 2.42	17.63 ± 3.35	18.02 ± 4.34	0.42 ± 0.14	2.62 ± 0.04	8.51 ± 1.2	703.95 ± 23.73	42.84 ± 2.1	2.89 ± 0.24	Manimaran et al. (2018a, 2018b, 2018c, 2018d)
<i>Tridax procumbens</i>	1160 ± 12	233.1 ± 9.9	32	6.8	3	0.71	–	11.2	25.75 ± 2.45	0.94 ± 0.09	2.77 ± 0.27	Vijay et al. (2019)
<i>Chloris barbata</i>	634	–	65.37	10.23	9.32	0.26	2.52	7.29	–	–	–	Balasundar et al. (2018)
<i>Hierochloe odorata</i>	1160 ± 20	136.71 ± 4.34	70.4	21.5	8.1	–	–	–	105.73 ± 35.42	2.56 ± 0.98	2.37 ± 0.95	Dalmis et al. (2020)
<i>Ficus racemosa</i>	895	–	72.36	11.21	10.45	0.25	6.59	6.13	270.83	67.45	–	Manimaran et al. (2019)
<i>Perotis indica</i>	785	–	68.4	15.7	8.35	0.32	4.32	9.54	317–1608	8.41–69.61	1.38–4.24	Prithiviraj et al. (2016)
<i>Acacia planifrons</i>	660	–	73.1	9.41	12.04	0.57	4.06	8.21	–	–	–	Senthamarikannan et al. (2016)
<i>Cyperus pangorei</i>	1102	1331 ± 17	68.5	–	17.88	0.17	3.56	9.19	196 ± 56	11.6 ± 2.6	1.69	Mayandi et al. (2016)
<i>Sida rhombifolia</i>	1320	–	75.09	15.43	7.48	0.49	4.07	12.02	673 ± 14	–	–	Gopinath et al. (2016)
<i>Lygeum spartum L</i>	1499.7 ± 3.1	180–433	–	–	–	–	–	–	64.63–280	4.47–13.27	1.49–3.74	Belouadah et al. (2015)
<i>Acacia arabica</i>	1028	–	68.10	9.36	16.86	0.49	–	–	–	–	–	Manimaran et al. (2016)
<i>Artisida hystrix</i>	540	–	59.54	11.35	8.42	–	–	–	440 ± 13.4	–	1.57 ± 0.04	Kathiresan et al. (2016)
Saharan aloe vera cactus	1325.1	80.61	60.2	14.2	13.7	1.5	–	7.6	621.8	40.03	2.47	Balaji and Nagarajan (2017)



**Fig. 2.** Classification of bio-degradable polymers.



**Fig. 3.** Chemical structure of PLA.

pellets. The gross energy use can be further accounted for the use of fossil energy and renewable energy sources. The renewable energy part accounts for 24.6 MJ/kg. On the other hand, the fossil energy use is 50.8 MJ/kg and the major contributors to this fossil energy use are coal, oil, gas and nuclear energy (Vink et al., 2007).

In comparison with various traditional plastics that are derived from petroleum-based reserves, the production of PLA consumes nearly 25–55% less fossil energy. Furthermore, around 90% reduction in the use of fossil energy can be achieved by using renewable energy sources like wind energy during different stages of production (Vink et al., 2003). Moreover, the greenhouse gas emission associated with PLA is low as the CO<sub>2</sub> emission during the bio-degradation of PLA is balanced by the intake of CO<sub>2</sub> from the atmosphere during the growth of agricultural feed-stocks. The CO<sub>2</sub> emission rate of PLA is around 1600 kg/metric ton, while PP, PS, PET and nylon have an emission of 1850, 2740, 4140, and 7150 kg/metric ton, respectively. In a long term, if PLA is produced from biomass or agricultural residues, it will start acting as a CO<sub>2</sub> sink and contribute to the reduction of greenhouse gases (Mohanty et al., 2005).

### 3.3. Degradation of PLA

Bio-degradation of any polymer takes place through the scission of the main chains or the side chains of macro-molecules. Polymer degradation is induced by different factors like hydrolysis, thermal activation, photolysis, biological activity or oxidation (Wan et al., 2019; Farah et al., 2016). PLA is becoming more popular due to its degradation behavior. The degradation of PLA depends on various factors like crystallinity, purity, molecular weight, temperature, pH, etc. (Park and Xanthos, 2009). In a compost atmosphere (i.e., high temperature and high humidity), the degradation of PLA is quick and it takes place within weeks.

The degradation of PLA is a two-step process beginning with hydrolysis, which is then followed by bacterial attack on the fragmented residues. Initially, the high molecular weight polyester chains are hydrolyzed to form low molecular weight lactic acid oligomers. When the average molecular weight reaches around 10,000 Da, the micro-organisms present in the soil begin to digest the low molecular weight oligomers, thus producing CO<sub>2</sub> and H<sub>2</sub>O (Farah et al., 2016).

### 3.4. Applications of PLA

The rise in oil price, depletion of oil reserves, global awareness of sustainable development and the enforcement of different legislations like the End of Vehicle Directive have paved the way for the use of biopolymers on a large scale. With this background, PLA has emerged to be one such biopolymer which has excellent potential to replace synthetic polymers. The properties of PLA can be controlled, allowing it to be used in a variety of applications. Some of the sectors in which PLA is used are given in Table 4 (Ahmed et al., 2018; Nagarjun et al., 2021).

### 4. Manufacturing methods

The final properties of a composite are heavily influenced by the manufacturing process. During fabrication, the different parameters can be controlled in order to tailor the output properties of the composites (Rajak et al., 2019). The manufacturing methods used to produce a composite depend on a number of factors. For instance, injection moulding and hot-pressing can be used to produce medium sized components whereas the pultrusion process is used to produce long,

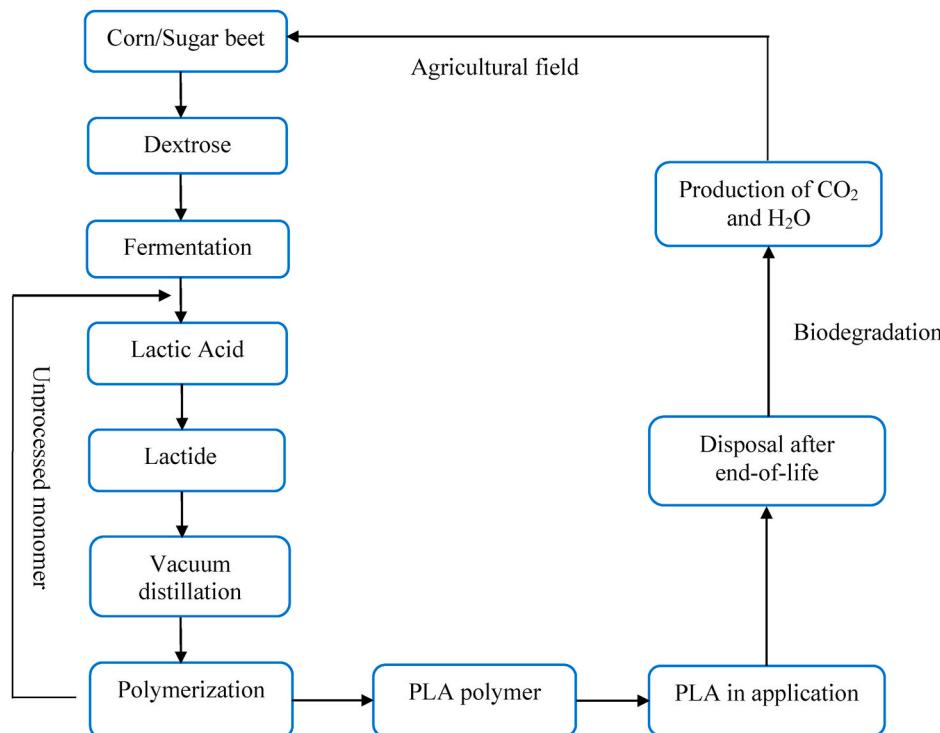


Fig. 4. Life cycle of PLA.

constant cross-sectional members. In addition, the cost, size of the final product, the number of required components, etc., dictate the selection of the appropriate process (Ho et al., 2012). PLA based green composites can be manufactured by a number of methods and some of these are discussed below in detail. Fig. 5 represents the different fabrication methods discussed in this paper.

#### 4.1. Injection moulding

Injection moulding is a typical process in which a measured quantity of fiber-filled resin in the liquid form is forced into mould cavities (Mazumdar, 2001). In order to manufacture cellulose fiber-based PLA composites by the injection moulding process, the required raw materials are PLA pellets or granules and chopped fibers. The injection moulding machine is shown in Fig. 6.

First, the PLA pellets and the chopped cellulose fibers are added to a hopper that is then fed into a cylindrical barrel, which has heaters placed on the top. The heaters are kept at a temperature equal to or greater than the melting point of PLA and are used to melt the resin. The barrel also contains a rotating screw inside it with which the liquid PLA and the chopped cellulose fibers are mixed uniformly in order to obtain a homogeneous mixture. The friction between the screw, barrel, and pellets dissipates some amount of heat, which is also used to melt the polymer pellets (Birgitha, 2007). This action converts the solid pellets into a viscous form, which further flows through the sprue nozzle and is forced into the mould cavity to produce part of the required dimensions.

A number of research studies have been reported using injection moulded PLA bio composites (Huda et al., 2005; Pääkkönen et al., 2017; Serizawa et al., 2006). An interesting study was performed using PLA and banana fibers. Here, the PLA pellets and banana fibers (20 wt.%) were fed into the hopper of an injection moulding machine. The barrel was set at temperature values of 160–165–165–170 °C from hopper to the nozzle. The injection, holding and back pressure were kept at 60, 40 and 10 MPa respectively whereas the holding and cooling time were kept at 15 and 25 s, respectively (Komal et al., 2020). Injection moulding is capable of producing complex parts at a high production rate. This

makes it a cost-effective process and is widely used in industries such as automobile, consumer goods and sporting.

#### 4.2. Extrusion injection moulding

Extrusion is a manufacturing process in which pellets of thermoplastic polymer are melted and then mixed with fibers using single or two rotating screws and then forced out of the chamber through a die. Twin screws are predominantly employed as they provide better dispersion of fiber and matrix. The output of the extrusion process is used as a precursor for the injection moulding process (Pickering et al., 2016). A substantial number of experiments on the production of PLA/cellulose fiber composites using extrusion method followed by injection moulding have been reported (Chaitanya and Singh, 2017; Jaszkiewicz et al., 2013; Komal et al., 2020). An illustration of extrusion injection moulding is depicted in Fig. 7.

Initially, the polymer pellets and the short fibers are added to the hopper of a single or twin-screw extruder. As the mixture passes along the barrel, the resin melts due to the application of heat and the screw action causes the polymer melt to uniformly blend with the fibers. The mixture is then forced out of a die. The composite is then cooled and then formed into pellets with the help of a pelletizer.

These composite pellets are then supplied to the injection moulding machine and are fabricated to the required dimensions. The drawback of PLA composites is that there is poor interfacial bonding between the cellulose fibers and the PLA matrix. Therefore, in order to improve the adhesion between the matrix and fiber, this two-step process of extrusion followed by injection moulding is carried out.

#### 4.3. Extrusion compression moulding

Another important method through which we can produce PLA based green composites is Extrusion compression moulding. This is a two-step process as in the case of extrusion injection moulding and is shown in Fig. 8. Initially, the PLA pellets and cellulose fibers are fed into a single/twin-screw extruder and then converted to PLA/fiber melt. This

**Table 4**  
Applications of PLA.

Sector	Application	Remarks	References
Biomedical	Surgical sutures	–	Maharana et al. (2009)
	Bio-absorbable stent	Stacks and his colleagues from Duke University introduced PLA stent for reduction of post-PTCA experimental restenosis.	Erne et al. (2006); Stack et al. (1988)
	Drug delivery system	PLA has been used to deliver different medical agents like vaccines, proteins, anesthetics etc.	Tan et al. (2010); Valantin et al. (2003)
	Biodegradable screws and fixation pins	These PLA based fixation devices are used in areas of hand, wrist, ankle, knee, foot etc.	Bucholz et al., 1994; Casteleyn et al. (1994); Stähelin et al. (1997)
	Injectable micro-spheres	PLA micro-spheres are used as temporary fillers in facial reconstructive surgery.	Lasprilla et al. (2012)
	Dermal fillers	These fillers stimulate the production of collagen in our body and aids in facial improvement.	Castro-Aguirre et al. (2016)
	Bone Scaffolds	Osteogenic stem cells are placed on PLA scaffolds and are used as implants to help the bone development process.	Ikada (2011)
	3-D porous scaffold	These scaffolds are used in the treatment of cardiovascular, orthopedic and neurological based diseases.	Coutu et al. (2009); Papenburg et al. (2009)
	Spinal cages	PLA has been used as inter-body cages for spinal infusion surgery.	Wuisman and Smit (2006)
	Inflatable balloon implants	These PLA based implantable balloons are used to reduce negative effects of radiation in prostate cancer by prostate-rectum separation.	Levy et al. (2009)
Textile	Sports apparel	Properties such as buffering capacity to sweat and thermal insulation have enabled the use of PLA in sports apparel.	Farrington et al. (2005)
	Ski jackets	PLA fibers are used in garments like Ski jackets to give an effective wadded layer due to low specific gravity and high resilient properties.	Farrington et al. (2005)
	Curtains, rugs and pillowcases	PLA fabrics are used as blankets, pillows, carpets, curtains etc. due to their good retention and crimp properties.	Castro-Aguirre et al. (2016)
	Non-woven textiles	PLA is used in hygiene products, upholstery, nappies, and disposable garments	Ahmed et al. (2018)
Automotive	Carpets	Companies such as Ford Motor Company are considering PLA as options for car interiors.	Ghosh and Krishnan (2007)
	Floor mats		
	Trims		
	Seat fabric		
Packaging	Food packaging films	These films are used for packaging purpose due to their supreme twist retention property.	Jamshidian et al. (2010a, 2010b); Mohanty et al. (2005)
	Food containers	Food packed in PLA containers is sold by Walmart which is the world's largest retailer.	Nampoothiri et al. (2010)
	Food-contact articles	PLA can be used to manufacture cups, cutlery, plates, straws, lids etc.	Farah et al. (2016); Rajak et al. (2019)
	Bottle labels	PLA is used in bottle labels of soft drink products by S&B foods, Nisshin OilliO and Asahi Soft Drinks.	Obuchi and Ogawa (2010)
	Grocery bags	Shopping bags are produced with 45% of PLA.	Castro-Aguirre et al. (2016)
	Tea bags	PLA exhibits high infusion capability due to which it is used in tea bags and coffee pouches.	Foss and Turra (2014)
	Coffee Pouches		
Agriculture	Mulch films	PLA can be blended with other bio-polymers to yield commercialized mulch films.	Jandas et al. (2013)
			Hayes et al. (2012)
Other applications	Cigarette filters	PLA fibers have been considered to make degradable cigarette filters.	Sebastian et al. (2012)

melt is transferred to a mould and it is pre-heated to the desired temperature. The mould is then placed in a compression moulding machine and hot pressed to obtain the final composite.

Composites of PLA and banana fibers were fabricated using this process by a research group. The PLA pellets and the fibers were compounded in an extruder and the melt was collected in a pre-heated mould. The mould was then hot pressed at 170 °C and a pressure of 40 bar. The resultant composite was then removed from the mould when the temperature reached 50 °C. In order to easily remove the composites, polyester sheets were used (Komal et al., 2020).

#### 4.4. Hot pressing

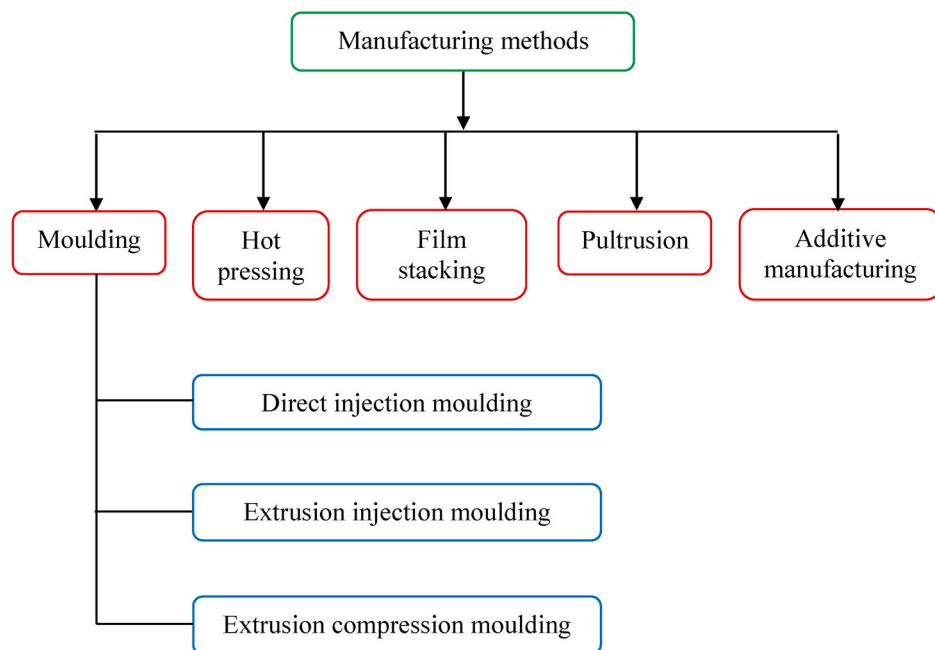
Hot pressing, also known as compression moulding, is a manufacturing technique similar to sheet forming process and is presented in Fig. 9. The raw materials required for the process are polymer (PLA) sheets or pellets and fibers, either in the form of loose chopped fibers or as fiber mats. Generally, the fibers are stacked alternatively with the polymer sheets in a closed mould followed by the application of heat and pressure. It is important to control the viscosity of the resin during pressing in order to obtain an optimum fiber impregnation (Pickering et al., 2016).

Initially, the mould is preheated to the desired temperature. Then, it is cleaned and a suitable release agent is applied in the mould to facilitate easy removal of the final formed composite panels. The fibers and the polymer precursors are then placed on the mould and then it is closed. The desired pressure is applied on the mould and the

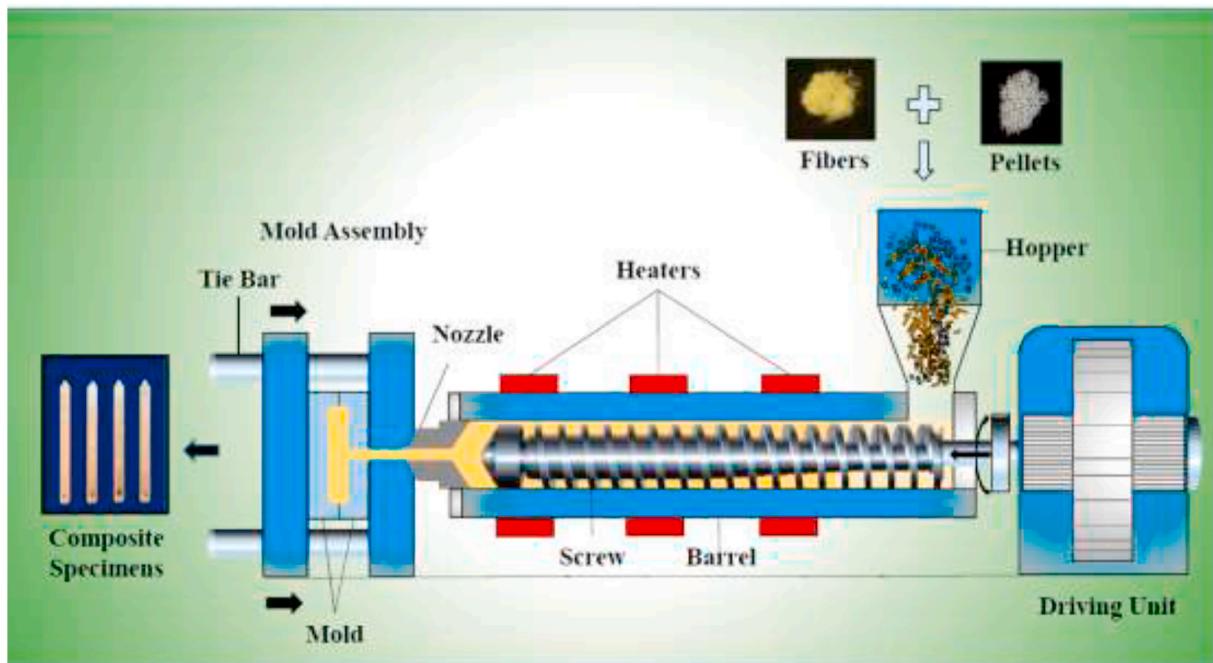
temperature is raised closer to the melt temperature of PLA. The process time depends on design requirements but typical process time is between 4 and 8 min. After completion of the process, the mould is cooled and the pressure is released. Subsequently, the mould is removed and the final product is taken out of the mould using ejector pins. The fabrication of PLA - bamboo fiber composite using compression moulding is explained here. Initially, the fibers were impregnated with PLA resin and then dried at 105 °C for 120 min in an oven. Simultaneously, the metallic mould was heated to a temperature of 180–200 °C. The mould was kept at the same temperature for 5 min and then resin impregnated fibers were hot pressed at 10 MPa. By this way, specimens with different volume fractions of bamboo fibers were fabricated (Ochi, 2015).

#### 4.5. Film stacking

Film stacking is a type of hot-pressing technique in which films of the thermoplastic matrix are stacked together along with the fiber mats. It is then consolidated by the application of heat and pressure. This process is clean but it requires special care during fabrication in order to impregnate the fibers completely. There are only a few studies that have used the film stacking method to produce PLA/cellulose fiber composites (Bajpai et al., 2013; Plackett et al., 2003). One important study uses this method to manufacture biodegradable composites based on PLA and sisal fibers. Initially, the PLA pellets are converted into a film using a compression moulding machine. Then, the fiber mats were cut into appropriate sections as per the mould dimensions. This was followed by



**Fig. 5.** Manufacturing processes used to fabricate PLA based bio-composites.



**Fig. 6.** Direct injection moulding process (reproduced with permission from Elsevier, license number: 4965130202761) (Komal et al., 2020).

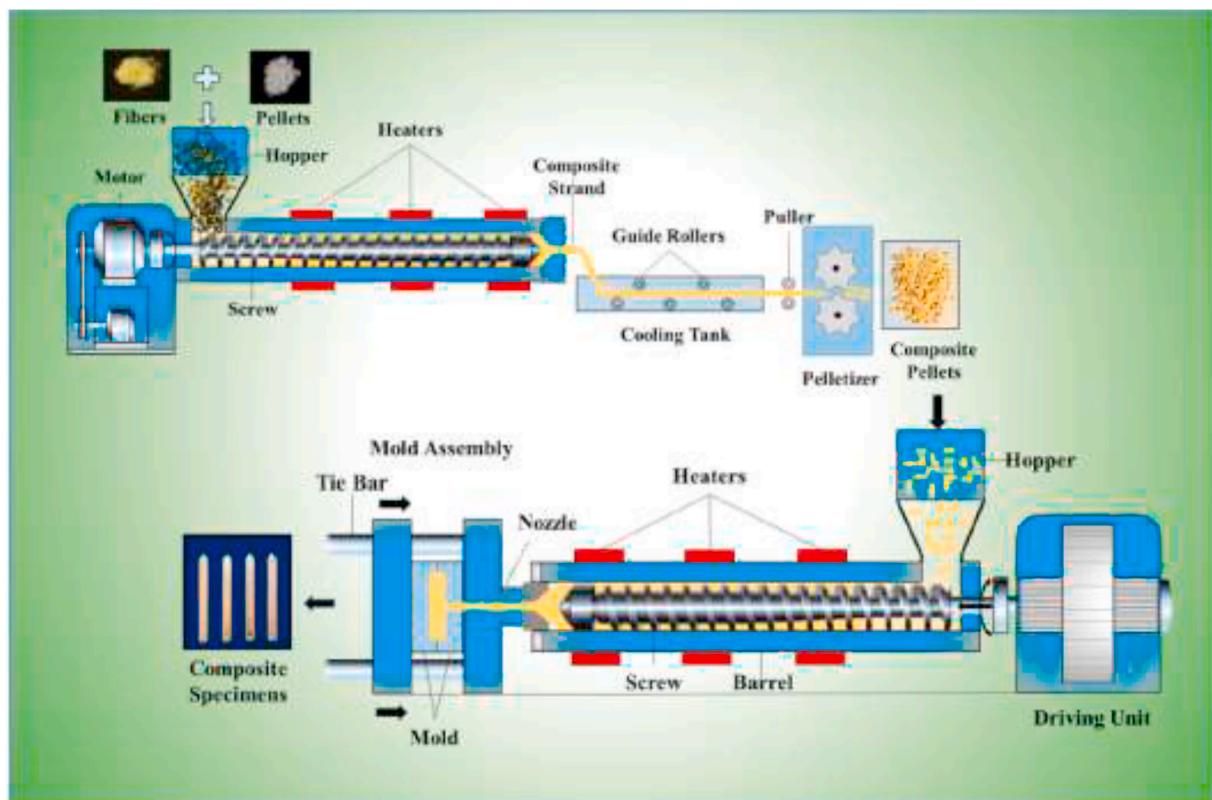
the alternative stacking of PLA films and fiber mats as illustrated in Fig. 10.

Stacking was done in such a way that the fiber mats were always placed in between two polymer films. Teflon sheets, which has a higher melting point than PLA was present at the top and bottom of the mould to prevent the polymer films from sticking to the metallic mould. The entire assembly was hot pressed at a temperature of 180 °C and at a pressure of 4 MPa for a period of 8 min. The pressure was then raised to 6 MPa for another 2 min and the assembly was subsequently cooled under pressure. When the mould reached a temperature of 80 °C, the composite was removed (Bajpai et al., 2013).

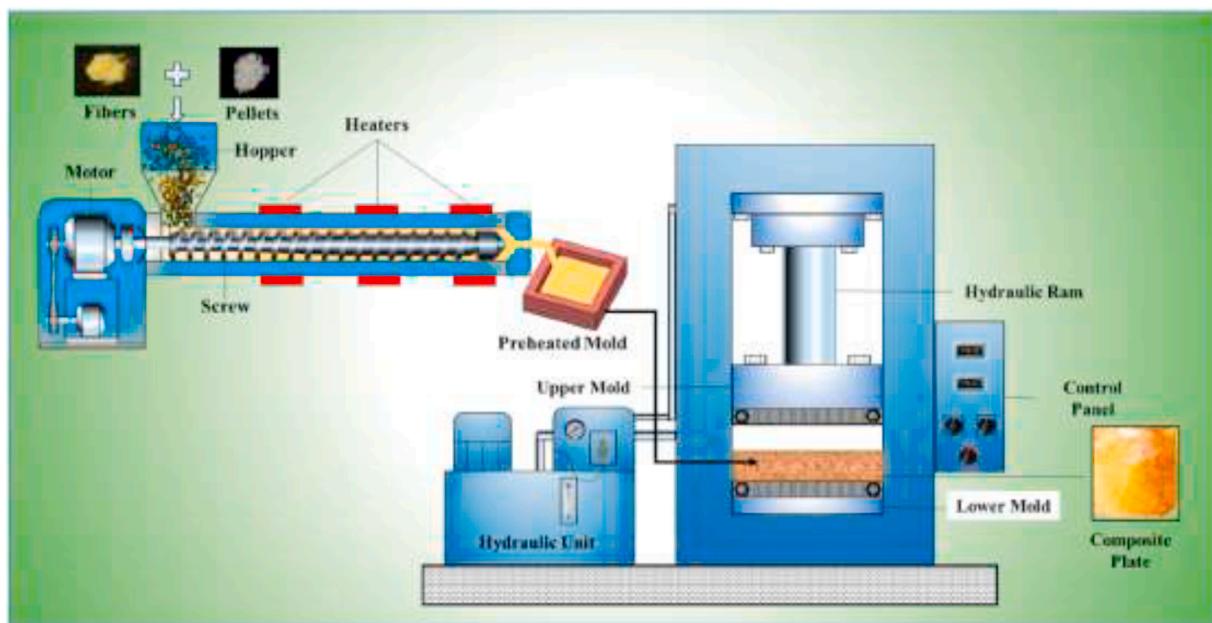
#### 4.6. Pultrusion

Pultrusion, whose schematic diagram is shown in Fig. 11, is another manufacturing technique through which we can produce cellulose fiber reinforced PLA composites (Linganiso et al., 2014; Memon and Nakai, 2013). It is predominantly used to manufacture long, constant cross-sectional members like solid rods, tubes, beams, flat sheets, etc. The fibers used are normally in the form of commingled yarns or continuous strand roving which are pre-impregnated with PLA.

When pultrusion is carried out using a thermoplastic resin, the fibers require a preheating step. Therefore, the fibers are first pulled through a pre-heating chamber which can be heated by hot air blowing,



**Fig. 7.** Process layout of extrusion injection moulding (reproduced with permission from Elsevier, license number: 4965130202761) (Komal et al., 2020).



**Fig. 8.** Process of extrusion compression moulding (reproduced with permission from Elsevier, license number: 4965130202761) (Komal et al., 2020).

microwave heating or both, which enhances fiber drying (Linganiso et al., 2014). The yarns are then slowly pulled through the electrically heated die which is kept at a temperature of 220–240 °C. In the heated die, the resin will melt and completely impregnate the fibers. This will be followed by the compaction and cooling of the composite at room temperature. The entire composite is pulled through the dies by a pulling system placed at the end of the production line.

#### 4.7. Additive manufacturing

A technique that has been gaining attention recently for the manufacture of PLA based natural fiber composites is additive manufacturing or 3-D printing. Among various 3-D printing methods, fused deposition modeling (FDM) is widely adopted as it is effective with various polymer materials such as PLA, ABS, polycaprolactone etc (Fig. 12). In this process, a filament comprising of the matrix and reinforcement is used. This

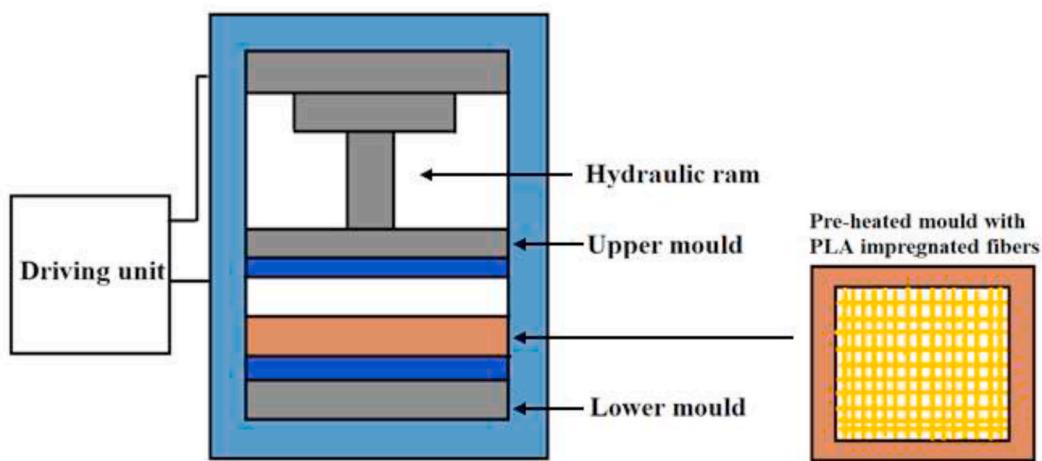


Fig. 9. Compression moulding setup.

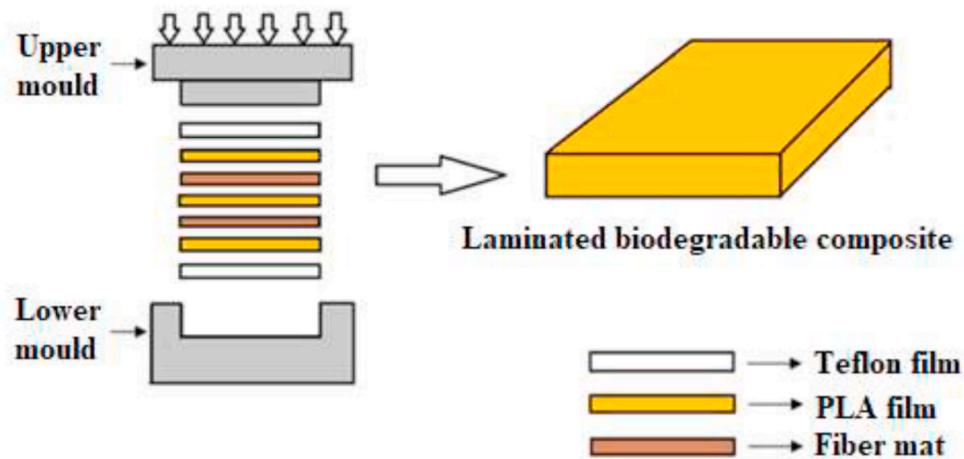


Fig. 10. Schematic of film stacking method.

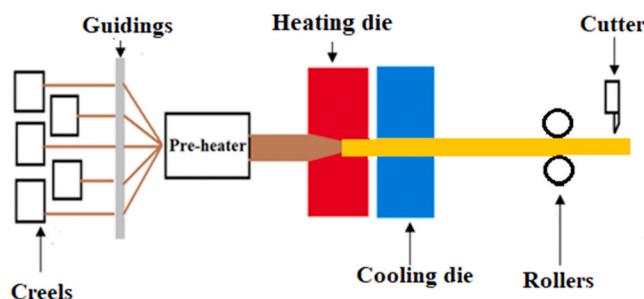


Fig. 11. Pultrusion process.

filament is guided through a heated nozzle with the help of a feeding roller and the resultant melt is deposited on a working table layer by layer. When the material solidifies, the partially liquefied layer fuses with the previous layer (Li et al., 2020).

The number of studies related to 3-D printing of natural fiber composites with PLA as a matrix material is less when compared to other conventional methods. Among the existing studies, there is a study in which properties of PLA/wood fiber composites fabricated through FDM process and injection moulding are compared. Here, different wt.% of wood was reinforced in PLA and the manufactured composites were subjected to tests such as mechanical, thermal, water absorption and

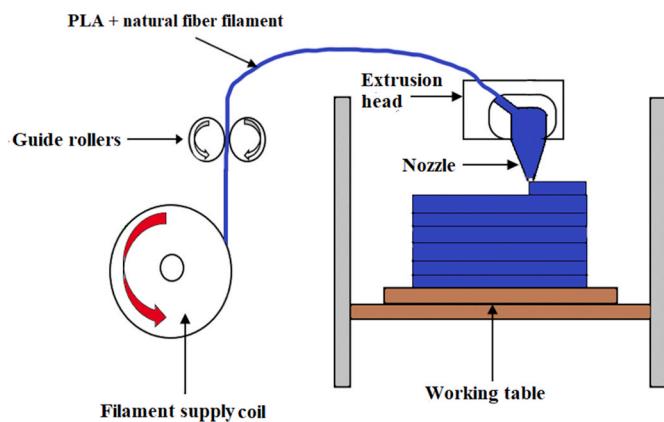


Fig. 12. Additive manufacturing of PLA/natural fiber composites.

morphological analyses. Samples produced through injection moulding had compact structure, while 3-D printed structures exhibited porous structures. Additionally, mechanical properties of 3-D printed structures were inferior to injection moulded samples (Ecker et al., 2019). Despite these results, it is important to note that additive manufacturing of natural fiber composites is still in its infancy and a substantial amount of research studies are required to understand the process and optimize it

**Table 5**

Benefits and drawbacks of different manufacturing methods used to produce PLA-green composites (Araújo et al., 2008; Chand and Dwivedi, 2006; Corradini et al., 2009; Mulinari et al., 2009; Peng et al., 2012; Santos et al., 2007; Zou et al., 2010; Li et al., 2020).

Manufacturing method	Benefits	Drawbacks
Injection moulding	<ul style="list-style-type: none"> <li>High production rate</li> <li>Ability to produce precise parts</li> <li>Intricate shapes can be manufactured</li> <li>Low operational cost</li> <li>Mass production is possible</li> </ul>	<ul style="list-style-type: none"> <li>Initial setup cost is high</li> </ul>
Hot pressing	<ul style="list-style-type: none"> <li>Produced parts have good surface finish</li> <li>Low wastage produced</li> <li>Fast setup time</li> <li>Low cost for large and complex parts</li> <li>Even distribution of pressure among the composites</li> </ul>	<ul style="list-style-type: none"> <li>Can be used only to manufacture flat or slightly curved composite parts</li> <li>Production speed is low</li> </ul>
Extrusion moulding	<ul style="list-style-type: none"> <li>Initial setup cost is less</li> <li>Fast setup time</li> <li>Low production costs</li> </ul>	<ul style="list-style-type: none"> <li>Mediocre precision can only be obtained</li> <li>Production speed is moderate</li> <li>Suitable only for parts with uniform cross-section</li> </ul>
Pultrusion	<ul style="list-style-type: none"> <li>It is highly automated</li> <li>Fast processing speed</li> <li>Economical method</li> <li>Increased composite strength can be achieved</li> </ul>	<ul style="list-style-type: none"> <li>Cost of heated dies can be high</li> <li>It is limited to parts with uniform cross-section.</li> </ul>
Additive manufacturing	<ul style="list-style-type: none"> <li>Easy and fast manufacturing</li> <li>Reduced thermal degradation</li> <li>Less expensive tools</li> <li>Better control of fiber volume and direction</li> </ul>	<ul style="list-style-type: none"> <li>The properties are inferior compared to conventional methods</li> <li>More research is necessary to optimize the process</li> <li>Nozzle clogging is prevalent</li> </ul>

(Balla et al., 2019).

Table 5 presents the benefits and drawbacks of the different manufacturing methods discussed in this article. In addition, Table 6 presents the manufacturing methods used to fabricate different PLA composites and also their properties.

## 5. Characterization

### 5.1. Mechanical properties

The mechanical properties of natural fiber composites have significant impact on the determination of the structural and light weight applications for polymer composites. Natural fibers from different plants and trees are effectively used as reinforcement material for PLA and their properties are influenced by fiber parameters (Bajpai et al., 2012; Du et al., 2014; Yu and Li, 2014). The sisal fiber incorporation at 20 wt. % enhanced the tensile, flexural and impact properties of PLA based composites up to 80.6 MPa, 249.8 MPa and 106.06 kJ/m<sup>2</sup>, respectively (Bajpai et al., 2012). Sisal fiber incorporation from 20 to 40 wt.% further enhanced the tensile strength up to 352 MPa and modulus to 13.9 GPa in PLA based composites (Orue et al., 2016). The fiber length of 1.24 mm in bleached kraft softwood showed improved tensile strength and modulus properties (Du et al., 2014). The fiber length of lyocell at the optimum range adds to the tensile and impact strength of PLA based composites, thereby enhancing the adhesion of fiber/matrix phase and improved their properties (Graupner et al., 2016).

Addition of 30 wt.% ramie fiber showed good flexural, tensile and impact properties of 105.2 MPa, 55.3 MPa and 10.2 kJ/m<sup>2</sup>, respectively, in PLA based composites (Yu et al., 2009). With the same percentage of

**Table 6**

Manufacturing methods used for the fabrication of different PLA composites and properties investigated.

S. No.	Manufacturing method	Fiber used	Properties studied	References
1.	Injection moulding	Oil palm	<ul style="list-style-type: none"> <li>Mechanical</li> <li>SEM analysis</li> <li>Thermal</li> <li>Moisture absorption</li> <li>FT-IR spectroscopy</li> <li>Optical microscopy</li> <li>Viscosity measurement</li> </ul>	Mamun et al. (2013)
2.	Injection moulding	Crushed kenaf and Long kenaf	<ul style="list-style-type: none"> <li>Mechanical</li> <li>SEM analysis</li> <li>Rheological</li> <li>Thermal characterization</li> </ul>	Serizawa et al. (2006)
3.	Injection moulding	Recycled cellulose	<ul style="list-style-type: none"> <li>Mechanical</li> <li>Thermal</li> <li>Dynamic mechanical analysis</li> <li>SEM analysis</li> </ul>	Huda et al. (2005)
4.	Injection moulding Extrusion injection moulding Extrusion compression moulding	Banana	<ul style="list-style-type: none"> <li>Hardness</li> <li>Thermal characterization</li> <li>Stereo-microscope Analysis</li> <li>FTIR spectroscopy</li> <li>XRD analysis</li> <li>Mechanical</li> <li>Dynamical mechanical analysis</li> <li>Morphological analysis</li> </ul>	Komal et al. (2020)
5.	Extrusion injection moulding	Sisal	<ul style="list-style-type: none"> <li>Stereo microscope</li> <li>Mechanical</li> <li>SEM analysis</li> </ul>	Chaitanya and Singh (2017)
6.	Extrusion injection moulding	Abaca Man-made cellulose	<ul style="list-style-type: none"> <li>SEM analysis</li> <li>Mechanical</li> </ul>	Jaszkiewicz et al. (2013)
7.	Film stacking	Jute	<ul style="list-style-type: none"> <li>ESEM analysis</li> <li>Mechanical</li> <li>Size exclusion chromatography</li> <li>Tribological</li> <li>XRD analysis</li> <li>Thermal</li> <li>Mechanical</li> </ul>	Plackett et al. (2003)
8.	Film stacking	Nettle <i>Grewia optiva</i> Sisal	<ul style="list-style-type: none"> <li>Thermal</li> <li>Mechanical</li> <li>Dynamic mechanical thermal analysis (DMTA)</li> <li>Water uptake</li> <li>Thermal analysis</li> <li>Mechanical</li> <li>SEM analysis</li> </ul>	Bajpai et al. (2013)
9.	Pultrusion	Jute	<ul style="list-style-type: none"> <li>Mechanical</li> </ul>	Memon and Nakai (2013)
10.	Pultrusion	Flax	<ul style="list-style-type: none"> <li>SEM analysis</li> <li>Mechanical</li> <li>Dynamic mechanical thermal analysis (DMTA)</li> <li>Water uptake</li> <li>Thermal analysis</li> <li>Mechanical</li> <li>SEM analysis</li> </ul>	Linganiso et al. (2014)
11.	Film stacking	Hardwood Softwood, Bleached Kraft softwood	<ul style="list-style-type: none"> <li>Mechanical</li> <li>Dynamic mechanical</li> <li>Thermal</li> <li>Mechanical</li> <li>Thermal</li> <li>SEM analysis</li> <li>Soil burial test</li> <li>Thermal</li> <li>Dynamic mechanical</li> <li>SEM analysis</li> </ul>	Du et al. (2014)
12.	Extrusion injection moulding	Kenaf Rice husk	<ul style="list-style-type: none"> <li>Mechanical</li> <li>Thermal</li> <li>SEM analysis</li> <li>Soil burial test</li> </ul>	Yussuf et al. (2010)
13.	Compression moulding	Bleached red algae	<ul style="list-style-type: none"> <li>Thermal</li> <li>Dynamic mechanical</li> <li>SEM analysis</li> </ul>	Sim et al. (2010)
14.	Extrusion injection moulding	Silk	<ul style="list-style-type: none"> <li>Hardness</li> <li>Mechanical</li> <li>Dynamic mechanical</li> </ul>	Cheung et al. (2008a, 2008b)

(continued on next page)

**Table 6 (continued)**

S. No.	Manufacturing method	Fiber used	Properties studied	References
15.	Extrusion injection moulding	Bagasse	<ul style="list-style-type: none"> <li>• Thermal</li> <li>• SEM analysis</li> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• SEM analysis</li> <li>• XRD analysis</li> <li>• Density and surface roughness</li> <li>• Accelerated weathering</li> </ul>	Lila et al. (2019)
16.	Extrusion injection moulding	Abutilon straw	<ul style="list-style-type: none"> <li>• FTIR analysis</li> <li>• Thermal</li> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• Rheological</li> <li>• Hardness</li> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• FTIR analysis</li> <li>• Thermal</li> <li>• SEM analysis</li> <li>• Water absorption test</li> <li>• Density and porosity</li> </ul>	Wang et al. (2019)
17.	Rotational moulding	Agave	<ul style="list-style-type: none"> <li>• Hardness</li> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• FTIR analysis</li> <li>• Thermal</li> <li>• SEM analysis</li> <li>• Water absorption test</li> <li>• Density and porosity</li> </ul>	Kumar and Das (2017)
18.	Compression moulding	Cow dung	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• Dynamic mechanical</li> <li>• Water absorption</li> <li>• Soil burial test</li> <li>• SEM analysis</li> </ul>	Yusefi et al. (2018)
19.	Compression moulding	Bleach Eucalyptus Kraft pulp	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• FESEM analysis</li> <li>• FTIR analysis</li> <li>• SEM analysis</li> </ul>	Nanthananon et al. (2018)
20.	Melt mixing	Posidonia oceanica	<ul style="list-style-type: none"> <li>• Calorimetric analysis</li> <li>• Mechanical</li> <li>• Dynamic mechanical</li> <li>• Density</li> <li>• Rheological</li> </ul>	Scaffaro et al. (2018)
21.	Extrusion compression moulding	Olive husk Flour	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• SEM analysis</li> <li>• Thermal</li> <li>• Bio degradation</li> </ul>	Hammiche et al. (2019)
22.	Extrusion	Date pit	<ul style="list-style-type: none"> <li>• Specific mechanical Energy</li> <li>• Thermal degradation</li> </ul>	Mohamed et al. (2018)
23.	Hot pressing	Bamboo	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• Optical microscopy</li> <li>• SEM analysis</li> </ul>	Masruchin et al. (2017)
24.	Extrusion injection moulding	Short flax seed fibers	<ul style="list-style-type: none"> <li>• Optical microscopy</li> <li>• SEM analysis</li> <li>• Mechanical</li> <li>• Thermal</li> </ul>	Agüero et al. (2020)
25.	Injection moulding	Coir	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• SEM analysis</li> </ul>	Nam et al. (2010)
26.	Compression moulding	Typha latifolia	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> </ul>	Daud et al. (2018)
27.	Dry mixing + Hot pressing	Corn starch	<ul style="list-style-type: none"> <li>• Thermal degradation</li> <li>• Enzyme degradation</li> <li>• Burial test</li> <li>• Thermal</li> </ul>	Ohkita and Lee (2006)
28.				

**Table 6 (continued)**

S. No.	Manufacturing method	Fiber used	Properties studied	References
29.	Extrusion compression moulding	<i>Manicaria saccifera</i>	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• SEM analysis</li> </ul>	Porras et al. (2016)
30.	Injection moulding	Elephant grass Jute Sisal	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• Water absorption</li> <li>• Soil burial degradation test</li> <li>• Enzymatic degradation test</li> <li>• SEM analysis</li> </ul>	Gunti et al. (2018)
31.	Hot pressing	Ramie	<ul style="list-style-type: none"> <li>• FTIR</li> <li>• Mechanical</li> <li>• Dynamic mechanical analysis</li> <li>• Thermal</li> <li>• SEM analysis</li> </ul>	Yu et al. (2010)
32.	Solution casting	Sucrose palmitate	<ul style="list-style-type: none"> <li>• Thermal</li> </ul>	Valapa et al. (2014)
33.	Extrusion injection moulding	Chicken feather	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Dynamic mechanical analysis</li> <li>• SEM analysis</li> </ul>	Cheng et al. (2009)
34.	Compression moulding	Hemp	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• Optical microscopy</li> <li>• SEM analysis</li> <li>• X-ray analysis</li> </ul>	Masirek et al. (2007)
35.	Injection moulding	Pineapple	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• SEM analysis</li> <li>• Water absorption</li> </ul>	Kaewpirom and Worrarat (2014)
36.	Wood flour	Wood flour	<ul style="list-style-type: none"> <li>• Hardness</li> <li>• Tribological</li> <li>• Microscopic observation</li> <li>• Mechanical</li> <li>• Dynamic mechanical analysis</li> <li>• Thermal</li> <li>• Abrasive Wear</li> <li>• SEM Analysis</li> </ul>	Mysiukiewicz and Sterzyński (2017)
37.	Hot pressing	Jute	<ul style="list-style-type: none"> <li>• Dynamic mechanical analysis</li> <li>• Thermal</li> <li>• SEM analysis</li> <li>• Mechanical</li> <li>• Tribological</li> </ul>	Goriparthi et al. (2012)
38.	Injection moulding	Coconut Flax Cellulose Fleece Wood	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• Water absorption</li> <li>• SEM analysis</li> </ul>	Béhálek et al. (2012)
39.	3-D Printing	Microcrystalline cellulose	<ul style="list-style-type: none"> <li>• SEM analysis</li> <li>• Dynamic mechanical analysis</li> <li>• Thermal</li> <li>• Water absorption</li> </ul>	Ecker et al. (2019)
40.	3-D Printing	Wood	<ul style="list-style-type: none"> <li>• Mechanical</li> <li>• Thermal</li> <li>• SEM analysis</li> </ul>	Murphy and Collins (2018)
				Kain et al. (2020)

ramie fiber addition (30 wt.%), an improved tensile strength of 64.3 MPa with PLA based composites was observed (Yu et al., 2014). The incorporation of 30 wt.% ramie fiber showed an enhanced flexural strength of 132.5 MPa and an impact strength of 11.3 kJ/m<sup>2</sup> in PLA based composites (Yu and Li, 2014). The area of cultivation significantly determines the physical and chemical properties of natural fibers, that changes the mechanical properties of polymer-based composites (Yu et al., 2009, 2014; Yu and Li, 2014). Incorporation of flax reinforcement also adds to the mechanical properties of PLA composites. Extrusion with compression moulding, micro-compounding with compression

moulding and extrusion with injection moulding fabrication process were carried out in flax/PLA based composites (Le Moigne et al., 2014; Oksman et al., 2003; Zhang et al., 2018). The flax fiber of 20 wt.% showed maximum tensile strength and impact strength up to 76.2 MPa and 21.6 kJ/m<sup>2</sup>, respectively (Le Moigne et al., 2014). Tensile modulus (8.1 GPa) has its maximum range with 30 wt.% flax (Oksman et al., 2003).

Addition of 20 wt.% kenaf fibers produced a high flexural strength of 90 MPa and flexural modulus of 3.4 GPa (Yussuf et al., 2010). The kenaf fibers at 30 wt.% showed a tensile strength of 59.5 MPa which is higher than the kenaf bast fibers with same percentage of addition (Ibrahim et al., 2010; Tawakkal et al., 2018). Substitution of 40 wt.% cellulose improved the tensile and impact strength up to 510 MPa and 27.1 kJ/m<sup>2</sup>, respectively, comparing to 30 wt.% and 0–2 wt.% cellulose fiber addition (Graupner et al., 2016; Kowalczyk et al., 2011; Spiridon et al., 2016). The cellulosic fiber along with the crystallinity of PLA based composites, showed good dimensional stability (Graupner et al., 2016). Addition of 15 wt.% jute fiber at a processing temperature of 210 °C showed maximum mechanical properties in polymer-based composites (Ma and Joo, 2011). The addition of 40 wt.% bamboo fiber exhibits maximum tensile and flexural properties in PLA based composites. Tensile and flexural properties enhanced up to 58.1 MPa and 83.4 MPa, respectively (Gamon et al., 2013).

The addition of kenaf, hemp and jute fibers in volume percentage also improved the mechanical strength of PLA based composites (Hu and Lim, 2007; Huda et al., 2008; Singh et al., 2020). Substitution of up to 50 vol.% kenaf fibers enhanced the flexural strength of PLA based composites up to 101.4 MPa (Huda et al., 2008). Hemp fiber substitution showed maximum mechanical properties at 40 vol.%. A tensile strength of 54.6 MPa, a tensile modulus of 8.5 GPa, a flexural strength of 112.7 MPa and a density of 1.25 g/cm<sup>3</sup> were observed during the substitution (Hu and Lim, 2007). Addition of 30 vol.% jute fiber showed optimized tensile and flexural properties in PLA based composites. The even fiber distribution in the matrix created a tight bonding in the PLA based composites that adds to the property enhancement. A maximum tensile strength of 64.13 MPa, a tensile modulus of 3.39 GPa, a flexural strength of 97.74 MPa and a modulus of 7.36 GPa were produced during this addition (Singh et al., 2020). Oil palm empty fruit bunch fibers of 30 wt.% showed high tensile strength of 67.3 MPa and a modulus of 2.73 GPa (Alam et al., 2012). The poor quality of fibers reduced the tensile properties of oil palm empty fruit bunch fibers at same combination (Alam et al., 2014).

A study by (Goriparthi et al., 2012) dealt with the influence of surface modification of jute fibers on the mechanical properties of PLA/jute composites. The fibers were treated with different chemical agents such

as alkali, silane 1, silane 2 and potassium permanganate. According to the findings, jute fibre composites treated with silane 2 had the best properties. The mechanical properties of PLA/jute composites were found to increase with fiber treatment. This is attributed to the enhancement of fiber-matrix bonding which was also confirmed from the SEM images (Fig. 13.) of the tensile fractured surface of untreated and treated PLA/jute composites.

It is apparent from the images that the fractured surface of untreated composite (Fig. 13a) has large number of fiber pullouts, which indicates the poor adhesion between matrix and reinforcement. On the other hand, fractured surface of silane 2 treated composites (Fig. 13b) exhibits fiber breakage rather than fiber pull out, which proves the improvement in the interfacial bonding between PLA and jute fibers after silane treatment. Due to this phenomenon, better stress transfer will take place which will lead to improved mechanical properties (Goriparthi et al., 2012).

Furthermore, Table 7 provides a comprehensive list of mechanical properties of PLA bio-composites that employ various natural fibers as reinforcement.

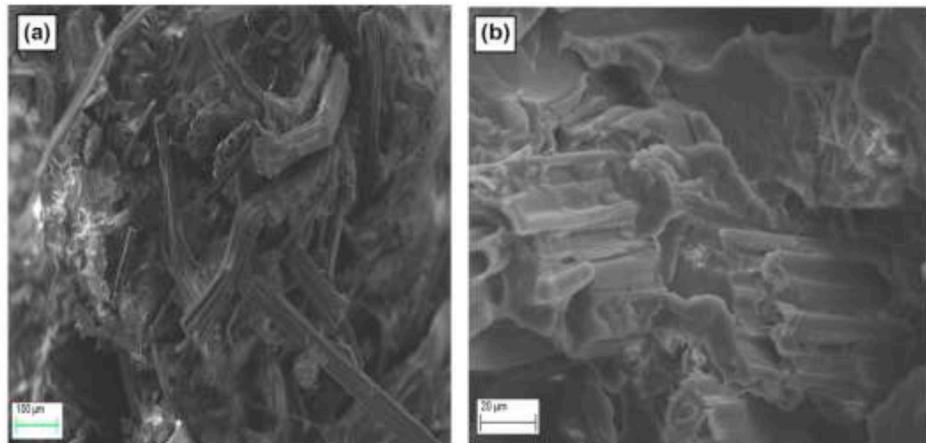
## 5.2. Dynamic mechanical analysis

It is pivotal to conduct dynamic mechanical analysis as it can act as an efficient tool to determine the viscoelastic characteristics and morphology of polymers and their composites (Saba et al., 2016). In addition, other parameters like dynamic fragility, density of cross-linking, stress-relaxation modulus and complex viscosity can be evaluated (Ornaghi et al., 2012; Pistor et al., 2012; Qazvini and Mohammadi, 2005). There are three major dynamic mechanical properties namely: storage modulus, loss modulus and damping factor.

The storage modulus ( $E'$ ) indicates the stiffness of the material and its ability to store energy. It can be used to obtain information about composite stiffness, degree of crosslinking and the bonding between matrix and fibers (Jawaid et al., 2012). It generally depends on length of the fiber, matrix material, bonding between matrix and fibers and load applied to the fibers (Nair et al., 2001).

The loss modulus ( $E''$ ) refers to the ability of the material to dissipate the energy that has been applied to it under cyclic loading. In contrast to storage modulus which is also termed as elastic response, loss modulus can also be defined as viscous response of a material (Ramakrishnan et al., 2021). The energy is generally dissipated in the form of heat. The loss modulus depends on various factors like morphology, structural homogeneities, molecular motions and relaxation processes (Jawaid et al., 2013; Naveen et al., 2019; Saba et al., 2016).

On conducting dynamic mechanical analysis, we obtain a value



**Fig. 13.** SEM images of tensile fractured surfaces of (a) untreated PLA/jute composites and (b) silane 2 treated PLA/jute composites (reproduced with permission from Elsevier, license number: 5046471264297) (Goriparthi et al., 2012).

**Table 7**  
Mechanical properties of natural fiber/PLA composites.

S. No.	Fiber combination	Fabrication technique	Addition of fiber (wt.%)	Best combination (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Impact strength (kJ/m <sup>2</sup> )	Elongation at break (%)	Density (g/cm <sup>3</sup> )	Hardness (Shore D)	References
1.	Crushed kenaf	Injection moulding	5–30	20	—	—	93	7.6	3.1	—	—	—	Serizawa et al. (2006)
2.	Long kenaf	Injection moulding	5–30	20	—	—	115	7.1	5.5	—	—	—	Serizawa et al. (2006)
3.	Banana	Injection moulding	10–30	20	52	1.43	104	4.8	—	—	—	84	Komal et al. (2020)
4.	Banana	Extrusion injection moulding	10–30	20	57	1.61	104	5.6	—	—	—	83	Komal et al. (2020)
5.	Banana	Extrusion compression moulding	10–30	20	18.5	1.52	30	4.1	—	—	—	83	Komal et al. (2020)
6.	Abaca	Extrusion injection moulding	30	—	79	7.6	—	—	3.7	—	—	—	Jaszkiewicz et al. (2013)
7.	Man-made cellulose	Extrusion injection moulding	30	—	120	6.5	—	—	8.2	—	—	—	Jaszkiewicz et al. (2013)
8.	Sisal	Compression moulding	20	20	80.6	5.3	249.8	9.75	106.06	6	—	—	Bajpai et al. (2012)
9.	Bleached Kraft softwood pulp	Film stacking	30–50	50	115.34	9.5	—	—	—	—	—	—	Du et al. (2014)
10.	Ramie	Compression moulding	0–50	30	55.3	—	105.2	—	10.2	3.8	—	—	Yu et al. (2009)
11.	Rice husk	Extrusion compression moulding	20	—	—	—	90	4.5	—	—	—	—	Yussuf et al. (2010)
12.	Cellulose	Compression moulding	30	—	62.3	4.1	—	—	15.2	—	—	—	Spiridon et al. (2016)
13.	Bamboo	Extrusion injection moulding	10–40	40	58.1	11.2	83.4	6.4	—	—	—	—	Gamon et al. (2013)
14.	Hemp	Compression moulding laminates	30–50 (vol. %)	40 (vol.%)	54.6	8.5	112.7	—	—	—	1.25	—	Hu and Lim (2007)
15.	Jute	Compression moulding	25–50 (vol. %)	30 (vol.%)	64.13	3.39	97.74	7.36	—	—	—	—	Singh et al. (2020)
16.	Oil palm empty fruit bunch	Extrusion	30	—	64.3	2.23	—	—	17.8	—	1.22	—	Alam et al. (2014)
17.	Bagasse	Extrusion injection moulding	30	—	80.2	2.62	111	5.24	3.58	—	—	—	Hong et al. (2019)
18.	Miscanthus	Extrusion compression moulding	20–40	20	59.1	3.18	—	—	—	—	—	—	Ragoubi et al. (2012)
19.	Wood flour	Extrusion injection moulding	20–40	30	63.3	5.3	116.6	8.9	—	—	—	—	Huda et al. (2006)
20.	Coir	Extrusion injection moulding	1–7	5	56	1.69	—	—	3.05	11.05	—	—	Sun et al. (2017)
21.	Silk	Extrusion injection moulding	1–7	5	62.08	2.54	—	—	—	10.29	—	19 (HV)	(continued on next page)

**Table 7 (continued)**

S. No.	Fiber combination	Fabrication technique	Addition of fiber (wt.%)	Best combination (wt.%)	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Impact strength (kJ/m <sup>2</sup> )	Elongation at break (%)	Density (g/cm <sup>3</sup> )	Hardness (Shore D)	References
22.	Abutilon straw	Extrusion injection moulding Extrusion injection moulding	1–5	5	50	0.48	–	–	–	12.5	–	–	Cheung et al. (2008a, 2008b) Wang et al. (2019)
23.	Coir	Hot pressing	30	–	13.96	4.35	24.6	3.96	–	–	1.13	–	Siakeng et al. (2019)
24.	Pineapple leaf	Hot pressing	30	–	21.86	5.11	38.53	5.56	–	–	1.02	–	Siakeng et al. (2019)
25.	Nettle	Compression moulding	10–90	50	50.82	3.34	36.89	–	28.76	2.45	0.825	–	Kumar and Das (2017)
26.	Agave	Rotational moulding	10–40	10	40	1.89	55	3	–	–	1.1	70	González-López et al. (2019)
27.	Cow dung	Compression moulding	10–50	30	45	–	–	5.3	–	–	–	–	Yusefi et al. (2018)
28.	Bleach eucalyptus kraft pulp	Compression moulding	10	–	89.03	1.948	–	–	–	5.95	–	–	Nanthananon et al. (2018)
29.	<i>Posidonia oceanica</i>	Melt mixing	10–20	20	39.2	2.219	72.9	4.73	–	2.32	1.2	–	Scaffaro et al. (2018)
30.	Olive husk flour	Extrusion	20	–	64	3.635	–	–	–	3.22	–	–	Hammiche et al. (2019)
31.	Short flax seeds	Extrusion & injection moulding	20	–	39.1	1.75	–	–	5.8	3.4	–	79.5	Aguero et al. (2020)
32.	<i>Manicaria saccifera</i>	Extrusion & compression moulding	40	–	68.45	4.89	133.12	3.94	26.62	3.48	–	–	Porras et al. (2016)
33.	Elephant grass	Injection moulding	5–25	20	66	2.54	113	6.95	5.45	2.85	–	–	Gunti et al. (2018)
34.	Chicken feather	Extrusion & injection moulding	2–10	5	55	4.2	–	–	–	4.4	–	–	Cheng et al. (2009)
35.	Pineapple	Injection moulding	10–50	40	17.5	3.6	–	–	–	0.92	–	–	Kaewpirom and Worarat (2014)
36.	Flax	Extrusion & injection moulding	10–30	30	56	4.18	86	3.825	16.4	3	–	–	Běhálek et al. (2012)
37.	Coconut	Extrusion & injection moulding	10–30	30	56	3.975	91	3.618	8.6	2	–	–	Běhálek et al. (2012)
38.	Fleece	Extrusion & injection moulding	10–30	30	46	3.571	60	3.464	19.7	3	–	–	Běhálek et al. (2012)

**Table 8**

Dynamic mechanical properties of PLA green composites.

S. No.	Natural fiber	Fabrication method	Fiber addition (wt.%)	Best combination (wt.%)	Max. storage modulus (MPa)	Max. loss modulus (MPa)	Tg (for loss modulus, °C)	Max. tan δ	Tg (for tan δ, °C)	References
1.	Banana	Injection moulding	10–30	20	3565	601	63.5	0.853	68	Komal et al. (2020)
2.	Banana	Extrusion injection moulding	10–30	20	3863	673	63.5	0.973	68	Komal et al. (2020)
3.	Banana	Extrusion compression moulding	10–30	20	3378	304	65	0.198	69	Komal et al. (2020)
4.	Hardwood	Film stacking	30–50	40	7900	585	61	0.17	64	Du et al. (2014)
5.	Softwood	Film stacking	30–50	40	7850	595	62	0.14	67	Du et al. (2014)
6.	Bleached kraft softwood	Film stacking	30–50	40	8200	710	60	0.18	64.5	Du et al. (2014)
7.	Flax	Compression moulding	30	—	4019	—	—	0.3	73	Zhang et al. (2018)
8.	Cellulose	Compression moulding	2, 20	20	3400	580	60	—	66	Kowalczyk et al. (2011)
9.	Bamboo	Extrusion injection moulding	10–40	40	950	—	—	0.78	69.5	Gamon et al. (2013)
10.	Miscanthus	Extrusion injection moulding	10–40	40	970	—	—	0.55	69.8	Gamon et al. (2013)
11.	Kenaf	Film stacking	40	—	4500	660	63.5	0.46	65.5	Huda et al. (2008)
12.	Bleached red algae	Compression moulding	30–60	50	5400	—	—	0.1	97.6	Sim et al. (2010)
13.	Abaca	Extrusion injection moulding	30	—	5700	1100	66	0.79	73	Adam et al. (2013)
14.	Jute	Extrusion injection moulding	30	—	6200	1050	66	0.81	74	Adam et al. (2013)
15.	Man-made cellulose	Extrusion injection moulding	30	—	5000	760	68	0.47	73	Adam et al. (2013)
16.	Silk	Extrusion injection moulding	1–7	5	1200	240	67	0.82	69.5	Cheung et al. (2008a, 2008b)
17.	Coir	Hot pressing	30	—	2200	360	56	0.8	65	Siakeng et al. (2019)
18.	Pineapple leaf	Hot pressing	30	—	2300	400	57	0.72	66	Siakeng et al. (2019)
19.	Bagasse	Extrusion injection moulding	20	—	1020	145	70	0.78	69.6	Lila et al. (2019)
20.	Abutilon straw	Extrusion injection moulding	1–5	5	1800	—	—	0.9	76	Wang et al. (2019)
21.	Sisal	Extrusion injection moulding	5–15	15	3430	570	60.3	0.96	66	Samouh et al. (2019)
22.	Nettle	Compression moulding	10–90	50	4024	174.49	89.69	0.067	94.12	Kumar and Das (2017)
23.	Agave	Rotational moulding	10–40	10	1500	—	—	1.25	65	González-López et al. (2019)
24.	Cow dung	Compression moulding	10–50	50	2700	—	—	0.63	87	Yusefi et al. (2018)
25.	Bleach eucalyptus kraft pulp	Compression moulding	10	—	2500	—	—	1.61	67	Nanthananon et al. (2018)
26.	<i>Posidonia oceanica</i>	Melt mixing	10–20	20	5000	—	—	1.5	66	Scaffaro et al., 2018
27.	Ramie	Hot pressing	—	—	2650	530	52	1.52	—	Yu et al. (2010)
28.	Chicken feather	Extrusion injection moulding	2–10	10	5650	—	—	0.89	66.3	Cheng et al. (2009)

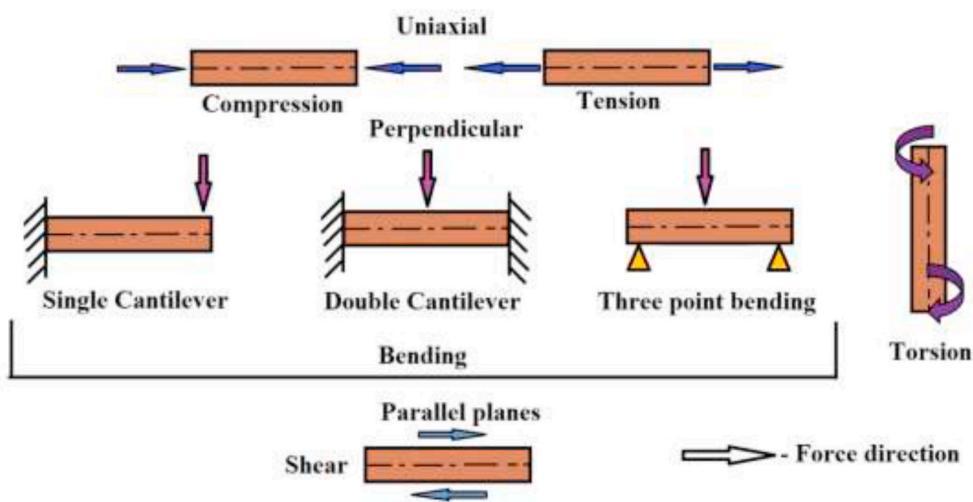


Fig. 14. Various configuration of testing used in dynamic mechanical analysis.

called the complex modulus, which is obtained based on the behavior of the material under sinusoidal loading. The ratio of stress to strain of the specimen gives the complex modulus, which is expressed in Eq. (1) (Gupta, 2017):

$$\text{Complex modulus } (E) = E' + iE'' \quad (1)$$

Here, the real part in Eq (1). is the storage modulus ( $E'$ ) and the imaginary part is the loss modulus ( $E''$ ).

$$\text{Storage modulus } E' = E \cos \delta \quad (2)$$

$$\text{Loss modulus } E'' = E \sin \delta \quad (3)$$

where,  $\delta$  is the phase lag between stress and strain.

The damping factor or  $\tan \delta$  value is defined as the ratio between loss and storage moduli. It has been reported that damping factor has a dependence on the fiber-matrix bonding. If a strong bond exists between fiber and matrix, then the damping factor will be low. This is because of the reduction in the mobility of polymer chains (Mohanty et al., 2006). The  $\tan \delta$  value is connected to different parameters like viscoelasticity, phase and grain boundaries, dislocations and molecular movements (Zhang et al., 2012). Table 8 provides the dynamic mechanical properties of various PLA green composites.

Dynamic mechanical analysis of composites can be carried out under different conditions and configurations. Fig. 14, represents the various configurations under which dynamic mechanical analysis is conducted.

### 5.3. Thermal analysis

When composites are subjected to heat, they are likely to experience prominent alterations in their physical and chemical properties. The material will undergo processes such as evaporation, sublimation, water absorption, etc., which is a function of both temperature and time. As a consequence, there will be variations in mechanical, thermal, electrical and magnetic properties (Ray and Cooney, 2018). In PLA based green composites, the reduction in the thermal stability may be predominant due to the degradation of the natural fibers (Ngaowthong et al., 2019). The degradation of natural fibers generally takes place at the processing temperatures of PLA (i.e., 200–210 °C) whereas the degradation of PLA begins at around 300 °C. Hence, it is crucial to study the thermal degradation of cellulose based PLA composites.

The thermal stability of composites is studied through thermogravimetric analysis (TGA) (Hammiche et al., 2019; Masruchin et al., 2017; Mohamed et al., 2018). During TGA, the mass of the sample is monitored as the temperature of the sample is increased. TGA is conducted in a thermogravimetric analyzer. The sample is placed on a sample pan. A thermocouple is present adjacent to the sample to monitor the temperature of the sample. This setup is enclosed in a furnace like setup which contains heating coils and cooling elements. Protective tubing is present to safeguard the sample from the heating coils. A purge gas which is generally an inert gas like nitrogen or argon is allowed to flow over the sample at a certain flow rate. As the temperature rises, the mass reduces as the sample decomposes and a graph (thermogram) between the sample mass change and temperature is obtained (Saadatkhan et al., 2020). A typical TGA thermogram is shown in Fig. 15. The different sections of the curves and their significance are elucidated below.

The thermal degradation of PLA bio-composites can be divided into three distinct zones. In the first zone, a weight loss of 5–10% can be seen in the composites. Here, the weight loss happens due to the evaporation of moisture that is trapped within the natural fiber. The second zone of degradation corresponds to the decomposition of hemi-cellulose and cellulose present in the fibers as well as to the degradation of the PLA matrix. Out of the three most important constituents of cellulose fibers; cellulose, hemi-cellulose and lignin, hemi-cellulose has the lowest thermal stability and it was the first compound to degrade among the three. Furthermore, it is followed by cellulose degradation while degradation of lignin takes place at a higher temperature (Beall and Eickner, 1970). Simultaneously, PLA degradation also happens in this zone. The mechanism behind the degradation of PLA comprises four different processes i.e., hydrolysis by traces of water, intra-molecular trans-esterification that results in the formation of cyclic oligomers,

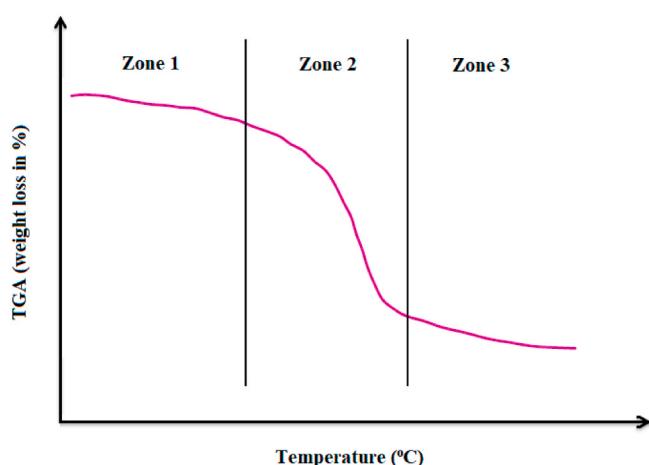
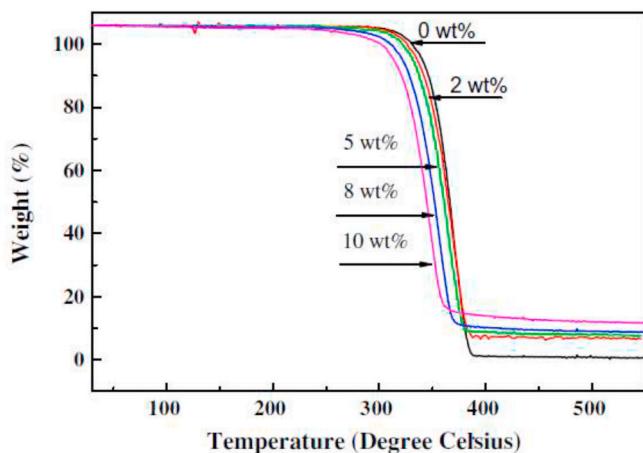


Fig. 15. Typical TGA thermogram.



**Fig. 16.** TGA curves of PLA and PLA/chicken feather composites (reproduced with permission from Elsevier, license number: 5046470617400) (Cheng et al., 2009).

cis-elimination that leads to acrylic acid and the formation of acetaldehyde and CO<sub>2</sub> due to fragmentation (Haafiz et al., 2013; Valapa et al., 2014). Finally, the third zone of degradation is attributed to the decomposition of lignin and other non-cellulosic molecules present in the natural fibers. The final residue that is obtained at the end is known as char.

Thermal analysis of PLA and its composites reinforced with different contents of chicken feather was carried out. The corresponding TGA curves are presented in Fig. 16. A drastic weight loss was observed for both PLA and PLA/chicken feather composites, starting from 300 °C. This transition is due to material degradation that happens above this temperature. Furthermore, there is a second transition that starts at around 360 °C when the materials begin to decompose. Additionally, PLA/chicken feather composites exhibited better thermal stability than pure PLA as the reinforcement served as barriers for improved heat insulation and prevented the permeation of volatile degradation products into the composite (Cheng et al., 2009).

The results obtained from the thermogravimetric analysis of various natural fiber reinforced PLA composites are presented in Table 9.

It can be seen clearly from the table that the initial degradation temperature of PLA based green composites varies in the range of 223–336 °C. In this range, the weight loss of the composites differs from 5 to 12%. Similarly, the final degradation temperature of the composites is between 324 and 415 °C and the weight loss is seen to vary from 75 to 95%. PLA composites reinforced with hemp, pineapple, recycled cellulose, chicken feather, and elephant grass demonstrate the highest thermal stability when compared with other PLA green composites.

#### 5.4. Tribological properties

Natural fiber-based composites have wide applications in machine tools, automotive interiors, bushing, sliding panels, and textile industries (Mohan and Kanny, 2019; Rajeshkumar et al., 2021; Rajeshkumar, 2020b). As the surfaces are not perfectly smooth, there is friction between them and this results in the deterioration of the component due to wear. This effect of friction and wear can be predominantly witnessed in applications such as machine tools, clutches, cam, rollers, bearings, etc. (Ahmed et al., 2019; Rajeshkumar, 2020b; Karthi et al., 2021). Tribology is the science that studies the phenomena of friction, wear and lubrication (Ravikumar et al., 2020). It is indispensable to understand the tribological properties of PLA/cellulose fiber composites and how cellulose fibers aid the enhancement of tribological properties of PLA composites.

The tribological properties of cellulose fiber reinforced PLA

composites were studied by incorporating three different fibers namely, nettle, *Grewia optiva* and sisal (Bajpai et al., 2013). The composites were fabricated using film stacking method and the wear test was conducted with the help of a pin-on-disc rotating type friction and wear tribometer. In a pin-on-disc method (Fig. 17.), a composite specimen of constant cross-sectional area is made to slide on a rotating disk which is predominantly made from ground stainless steel (Yousif and El-Tayeb, 2008). In the above experiment, the co-efficient of friction and specific wear rate were measured and in addition, the worn surfaces were analyzed using SEM. It was found that the minimum friction co-efficient of PLA/nettle, PLA/*Grewia optiva* and PLA/sisal increased by 34%, 20% and 31%, respectively when compared to neat PLA whereas the maximum friction co-efficient of PLA/nettle, PLA/*Grewia optiva* and PLA/sisal substantially dropped down by 44%, 10% and 32%, respectively. On the other hand, the minimum specific wear rate of PLA/nettle, PLA/*Grewia optiva* and PLA/sisal reduced by 28%, 5% and 54%, respectively, when compared to PLA, whereas maximum specific wear rate of PLA/nettle, PLA/*Grewia optiva* and PLA/sisal drastically slumped by 79%, 82% and 72%, respectively. This means that the wear performance of PLA is significantly enhanced by the addition of cellulose fibers and these composites can be used as a viable alternative in tribological applications (Bajpai et al., 2013).

The experiment Mysiukiewicz and Sterzyński (2017) involved the study of static and dynamic co-efficients of friction of PLA/wood flour composites. The composite specimens were produced by incorporating 10, 20 and 30 wt.% of wood flour (WF). The horizontal plane method was used to measure the co-efficient of friction. The study showed that both static and dynamic co-efficients of friction decreased with the addition of wood flour. One explanation for this can be that, hardness of polymer composites increases with the addition of wood flour (Kord, 2011). Due to this, the deformation during sliding is low. The static friction co-efficient was almost similar to neat PLA when 10 wt.% of wood flour was added but this quantity reduced by 23% with the addition of 20 wt.% wood flour. This was further reduced by 30% when 30 wt.% of wood flour was added to PLA. On the contrary, the dynamic friction co-efficient was reduced by 24% in PLA/WF (10 wt.%) as compared to PLA. Furthermore, there was a 54% reduction in the dynamic friction co-efficient of PLA/WF (20 wt.%) and PLA/WF (30 wt.%) when compared to neat PLA (Mysiukiewicz and Sterzyński, 2017).

The tribological properties of a PLA green composite can be enhanced by improving the adhesion between the matrix and the fiber (Wang et al., 2014). This can be done by chemically treating the surface of the cellulose fiber. One such study looks into the effects of fiber surface treatments on the wear performance of PLA/jute composites (Goriparthi et al., 2012). The jute fibers were treated with 5% NaOH, permanganate acetone, benzoyl peroxide, silane 1 and silane 2 solution and the composites containing 50 wt.% jute fibers were manufactured by hot pressing method. The composites were then subjected to wear tests using pin-on-disc apparatus. The results indicated that the surface treated PLA/jute composites had comparatively higher wear resistance than untreated PLA/jute composites. Among the different chemical agents used, PLA/silane 2 treated jute composites had the highest wear resistance whereas PLA/alkali treated jute composites had the lowest among all. Upon analyzing the SEM micro-graphs (Fig. 18.) of the worn surfaces of untreated composite and silane 2 treated composite, it was found that micro-cutting mechanism was the major cause for material removal. In addition, shallow ploughed grooves were observed on PLA/silane 2 treated jute composites, which is attributed to the exceptional wear resistance (Goriparthi et al., 2012).

Another interesting study involved the measurement of static and dynamic co-efficients of friction of PLA composites reinforced with coconut, flax cellulose fibers and fleece fibers (Běhálek et al., 2012). The composites were prepared using Extrusion injection moulding process for different quantity (10 wt.%, 20 wt.%, 30 wt.%) of fibers. The friction co-efficient was calculated using a device which contained a horizontal desk made from anodized aluminum. A constant force of F<sub>N</sub> = 106.6 N

**Table 9**

Thermal analysis results of different natural fibers reinforced PLA composites.

S. No.	Natural fiber	Fabrication method	Fiber addition (wt.%)	Best combination (wt.%)	Initial degradation		Final degradation		Residue	References
					Temperature (°C)	Weight loss (%)	Temperature (°C)	Weight loss (%)		
1.	Recycled cellulose	Injection moulding	30–40	30	325	5	394	75	7	Huda et al. (2005)
2.	Nettle	Film stacking	20	–	320	10	360	80	1.9	Bajpai et al. (2013)
3.	<i>Grewia optiva</i>	Film stacking	20	–	312	10	357	80	2.7	Bajpai et al. (2013)
4.	Kenaf	Extrusion injection moulding	20	–	321	10	357	75	4	Yussuf et al. (2010)
5.	Rice husk	Extrusion injection moulding	20	–	305	10	340	75	5	Yussuf et al. (2010)
6.	Bleached red algae	Compression moulding	30–60	30	299.2	5	354.3	75	4.9	Sim et al. (2010)
7.	Silk	Extrusion injection moulding	1–7	5	300	5	360	80	18	Cheung et al. (2008a, 2008b)
8.	Abutilon straw	Extrusion injection moulding	1–5	5	255	7	335	60	28	Wang et al. (2019)
9.	Sisal	Extrusion injection moulding	10–30	20	304	5	370	90	3	Ngaowthong et al. (2019)
10.	Olive husk flour	Extrusion compression moulding	20	–	260	7	324	86	8	Hammiche et al. (2019)
11.	Flax seeds	Extrusion injection moulding	20	–	279.1	5	368.4	90	3.4	Agüero et al. (2020)
12.	Flax	Hot pressing	34	–	310	5	380	90	4	Foruzanmehr et al. (2016)
13.	Coir	Injection moulding	10–50	10	336	10	385	82	10	Nam et al. (2012)
14.	<i>Typha latifolia</i>	Compression moulding	10–40	10	260	8	380	92	1.32	Daud et al. (2018)
15.	Corn starch	Dry mixing and Hot pressing	10–50	30	304	7	377	95	3.4	Ohkita and Lee (2006)
16.	<i>Manicaria saccifera</i>	Hot pressing	40	–	296	8	354	83	11.1	Porras et al. (2016)
17.	Elephant grass	Injection moulding	5–25	20	315	10	390	89	2.5	Gunti et al. (2018)
18.	Ramie	Hot pressing	30(vol%)	–	268	5	347	90	4.5	Yu et al. (2010)
19.	Sucrose palmitate	Solution casting	1–10	1	288	10	347	90	2	Valapa et al. (2014)
20.	Banana	Compression moulding	10–40	–	223	7	367.28	90	1.5	Jandas et al. (2011)
21.	Bamboo	Extrusion compression moulding	40	–	250	5	350	84	5	Lin et al. (2018)
22.	Wood flour	Melt compounding	10–40	10	300	7	370	81	4	Lee et al. (2008)
23.	Chicken feather	Extrusion injection moulding	2–10	2	330	5	390	92	5	Cheng et al. (2009)
24.	Hemp	Compression moulding	1–30	20	303	10	398	95	1.2	Masirek et al. (2007)
25.	Pineapple	Extrusion injection moulding	10–50	30	325	10	415	80	18	Kaewpirom and Worrarat (2014)
26.	Jute	Hot pressing	50	–	310	9	378	93	2.4	Goriparthi et al. (2012)

was applied at the centre of the sample. The friction force was recorded with respect to the displacement when the solid was sliding at a constant velocity of  $v = 100$  mm/min. The static co-efficient of friction was determined from the maximum frictional force that acts on the sample after the start of displacement. In contrast, dynamic co-efficient of friction was found from the average frictional force between the displacement 10 mm and 70 mm (Béhálek et al., 2012). Table 10 illustrates the percentage reduction of static and dynamic friction co-efficients of PLA composites.

### 5.5. Vibration analysis

When composites are utilized in engineering applications such as in automobile industry, they will be subjected to vibrations. If a material undergoes continuous vibration, it can lead to fatigue cracks, premature wear and generation of noise. Additionally, if the loads acting on the

material make it to vibrate at its natural frequency, then this can lead to failure (Etaati et al., 2014; Geethamma et al., 2014). Hence, the study of vibration on composites is pivotal. The vibration damping characteristics of natural fiber composites is dependent on various factors such as fiber volume fraction, aspect ratio of reinforcements, stacking sequence, properties of matrix and reinforcements, quality of fiber-matrix interface and loading direction (Akoussan et al., 2016; Chung, 2003; Etaati et al., 2014; Ni et al., 2015). In polymers, the vibration energy is dissipated by means of shear deformation (Gibson et al., 1982). However, there are various ways of energy dissipation from the composites namely: thermoelastic and visco-plastic damping, viscoelasticity of matrix, energy loss at cracks and the friction produced at the fiber-matrix interface (Chandra et al., 1999; Chauhan et al., 2009).

The damping properties of composites are observed to be greater than traditional metals (Jeyaraj et al., 2009). Furthermore, natural fibers possess better vibrational damping properties than synthetic fibers,

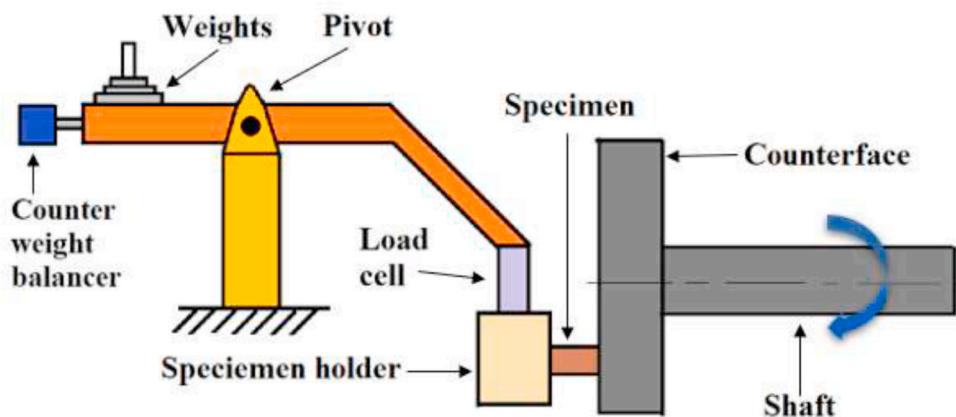


Fig. 17. Pin-on disk test setup.

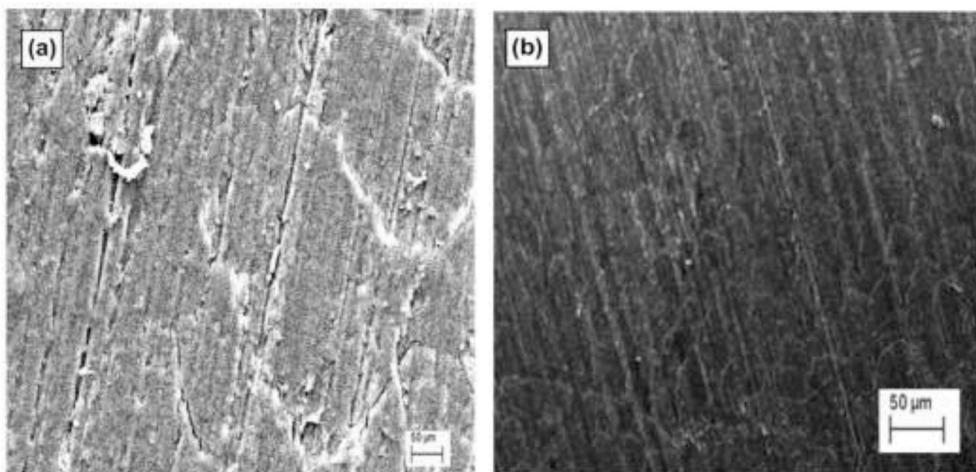


Fig. 18. SEM micro-graphs of worn surfaces of (a) untreated PLA/jute composites and (b) silane 2 treated PLA/jute composites (reproduced with permission from Elsevier, license number: 5046471264297) (Goriparthi et al., 2012).

**Table 10**  
Percentage reduction of static and dynamic friction co-efficients of PLA composites when compared to neat PLA.

Friction co-efficient	Quantity (wt. %)	Natural fibers			
		Coconut	Flax	Cellulose	Fleece
Static friction co-efficient	10	25	14.28	18.75	18.75
	20	25	25	25	25
	30	31.25	25	25	25
Dynamic friction co-efficient	10	30	40	40	30
	20	40	50	50	40
	30	50	50	50	50

which results in enhanced vibrational damping performance of natural fiber reinforced composites over synthetic ones (Hanipah et al., 2016). A method employed to evaluate the vibration damping characteristics is known as an impulse hammer test. The schematic of the test setup is presented in Fig. 19. One end of the specimen is fixed and the free end is subjected to vibrations with the help of an impulse hammer. An accelerometer is attached to the free end of the test piece and it is used to measure the displacement that occurs. Finally, the data are sent to a Fast Fourier Transformation (FFT) analyzer for further analysis (Kamal and Taha, 2010).

Vibration tests performed for the PLA based natural fiber composites are very minimal. Nevertheless, the vibration damping properties of PLA green composites of one particular work is presented in Table 11. In this

study, vibration damping analysis of PLA bio-composites reinforced with three natural fibers, namely: cotton, flax and bamboo, was reported. The natural frequency and damping ratio of the first three natural modes were experimentally investigated.

## 6. Conclusions and perspectives

Bio-composites play a major role in a number of applications and its properties can be enhanced by the addition of various materials in the base matrix. Bio-composites are fabricated from divergent renewable and non-renewable materials. Among the renewable polymers, PLA is a unique natural thermo-plastic polymer which is very much compatible to the environment and has better properties. This review compiles the overview of PLA synthesis, degradation and its applications in various fields, characteristics of various natural fibers and their sources, different manufacturing methods available for the fabrication of natural fiber based PLA composites and impact on the distinct properties with the addition of natural fiber in PLA. Injection moulding and hot-pressing methods are used to produce medium sized PLA based composites whereas, pultrusion process is used for long, constant cross-sectional PLA based composites. The addition of natural fiber improved the mechanical and thermal properties of PLA by creating good interfacial bonding and physical entanglement in the PLA matrix. Wear performance of PLA is significantly enhanced with the addition of cellulose fibers and these composites can be used as a viable alternative in tribological applications. However, details about the vibrational

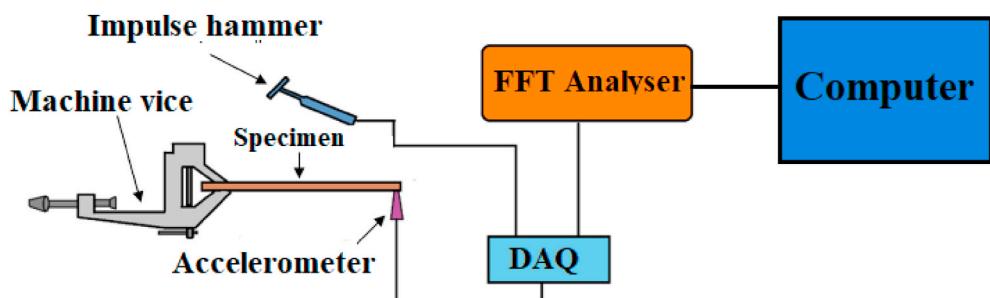


Fig. 19. Setup of impulse hammer test.

**Table 11**  
Vibration damping properties of PLA green composites.

S. No.	Natural fiber	Fabrication method	Fiber addition (wt.%)	Mode 1		Mode 2		Mode 3		References
				Natural frequency (Hz)	Damping ratio	Natural frequency (Hz)	Damping ratio	Natural frequency (Hz)	Damping ratio	
1.	Cotton	Hot pressing	50	96	0.055	372	0.04	764	0.021	Zhang et al. (2019)
2.	Bamboo	Hot pressing	50	228	0.048	646	0.06	1299	0.04	Zhang et al. (2019)
3.	Flax	Hot pressing	50	238	0.0405	501	0.0395	1022	0.057	Zhang et al. (2019)

analysis of PLA based natural fiber composites are very minimal in the literature and need further focus. In conclusion, improving the properties of PLA based natural fiber reinforced bio-composites is a quiet dynamic research area and various possible approaches to the inclusion of low cost natural fiber in the PLA needs to be investigated. This review could help the readers to understand the properties and impact of cellulose fiber reinforcement in PLA.

#### 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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