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Natural fibre filament for Fused Deposition Modelling (FDM): a review

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ABSTRACT
Fused Deposition Modelling (FDM) gets the most attention in development and manufacturing industries. The demand for FDM in industries increases gradually over time and attracts many researchers to enhance the quality of the FDM's fillers. The most popular filler reinforcements in use are synthetic or carbon fibre. However, these fibres are harmful to the environment. To overcome the issue and replace the current fibres and achieve the bio-composites filler, researchers suggested using natural fibre to replace the synthetic and carbon fibres as the reinforcement, which is also combined with bio-polymer matrix such as thermoplastics as the polymer matrix in FDM's industries. Many experiments and tests are conducted to prove the capability of the natural fibre as the main material in composite industries. FDM is a world-wide technology that aims to be environmentally friendly, thus, this paper focuses on biodegradable fillers for FDM.

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Fused Deposition Modelling (FDM); biodegradable; natural fibres; polymer; fillers; composites

1. Introduction

Positive characteristics such as light-weight, high strength, and low cost make natural fibre reinforced thermoplastics favourable by several industries. Two major components of the composite material are matrix and reinforcement (Ali et al. 2018). Natural fibre is also known as having biodegradable properties complying with the rules of green materials. Nevertheless, the problem that still needs emphasis when using this kind of composites is that the adhesion bonding between reinforced and polymer matrix leads to costly consequences, such as wetting problem, swelling, and dimension instability. This interface problem can affect the mechanical and physical properties itself. From the physical properties' perspective, natural fibres have hydrophilic characteristics that reduce water and absorb moisture, which are important to prevent dimension swelling and composite shape change. This prevention happens when the chemical treatment of natural fibres is complete (Obada et al. 2020).

Due to the future environmental concerns, the development of polymer composites using materials that can be decomposed or recycled is very important (Tholibon et al. 2019). Replacing synthetic and carbon fibre with natural fibre has many advantages that overcome the negative effects of synthetic, such as air toxicity, respiratory problems, recyclability, renewability, mechanical properties, and waste issues (Sanjay et al., 2016).

Development of the composite materials is a new generation that keeps up with the growing demands of technological and industrial changes. Mechanical property enhancement is a result of the composite's combination (Tholibon et al. 2019). The main role of the reinforcement is to act as the crack stopping and load bearing material. It also enhances the stiffness in mechanical properties and achieves good physical properties of the matrix (kumar, 2016).

L.Y. Mwaikambo's finding stated that 'cellulose is a skeletal polysaccharide, ubiquitous in the plant kingdom and one of the commonest naturally occurring fibrous materials. Strictly speaking all plant fibres are single-cell materials' (Mwaikambo 2017). One of the most popular natural fibre is kenaf fibre, which gets high attention among the researchers for the production of composites (Tholibon et al. 2019).

The governments required the use of green materials that can be recycled and reused. The decreasing value of petroleum resources has made societies realise the importance of preserving renewable sources for future generations, and industrialists understand the concept of sustainability in production. Currently, researchers are struggling with their research to develop base composites by using natural fibre reinforced biodegradable polymer matrix as the first biodegradable and sustainable product. For example, Japan has developed many products by using kenaf fibre reinforced polylactic acid (PLA) (Netravali 2005).

Environmental issues such as air pollution and waste disposal that may affect the whole ecosystem have driven researchers to investigate biodegradable composites to replace the usage of petroleum as the main renewable product and also investigate a bio-friendly polymer process (Coppola et al. 2018). A fabrication that has complex parts and geometries is from a 3D design software. A computer aided design (CAD) without requiring a mould is called additive manufacturing

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technologies of fused deposition modelling (FDM). The advantages of using additive manufacturing technologies are low processing time, better flexibility, the ability to build complex shapes, and a better finish (Montalvo and Hidalgo 2020). FDM is also more popular than other additive technologies for its ease of use and low cost (Coppola et al. 2018). PLA was found in combination with the natural fibres (kenaf, flax, or hemp) as a matrix to produce biodegradable composites (Daver et al. 2018).

2. Natural fibres

Due to environmental issues, global warming, pollution, health hazard, reduction of fossil materials, and declining economic benefits, renewable and biodegradable materials have been introduced to the production market as eco-friendly materials to health and the earth (Preet Singh et al. 2017). One of the alternatives discussed that would improve the quality of the environment and the new product (C. Wang et al., 2019) is the replacement of the synthetic and carbon fibre with natural fibre, which has been investigated by many scientists and researchers. Natural fibres have been in use for centuries. Hunters used to utilise natural fibres for their daily equipment such as rope, basket, mat (Sreenivasan et al. 2013), and even rigid structural buildings (Sanjay et al. 2018). This proves that natural fibre is a material that is readily available as compared to the synthetic materials, glass, and carbon fibre.

(C. Wang et al., 2019) stated that automotive, aeronautics, sports equipment, marine, electronics circuit, and even construction (Mahjoub et al. 2014) industries consume natural fibre materials as their main material fillers with the combination of polymer matrix. Natural fibre also has been used as the main component in cosmetics, cigarettes, drinking straw, and as an automotive structure in car bumper, car door, etc. (Kicińska-Jakubowska, Bogacz, and Zimniewska 2012). The use of natural fibres is increasing because they are biodegradable, light weight, and have a low aspect ratio and high specific strength compared with glass fibre (Akil et al. 2011). They are also cheaper than synthetic fibres and easy to manufacture (C. Wang et al., 2019).

Different countries consume different types of natural fibres for their manufacturing products, sometimes importing or exporting them to other regions. (Peças et al. 2018) wrote that the automotive production in Europe consume flax and hemp as their main fibres in their industry. The European countries required fibres such as jute and kenaf from Bangladesh and India, banana from Philippines and sisal from South Africa, the United States, and Brazil. In Germany, flax fibre is the most common fibre used in automotive production. Figure 1 shows the percentage of natural fibre usage in the European automotive industry, where wood was used the most, followed by cotton and other fibres. Table 1 shows other author that classified natural fibres consumption in automotive parts. The automotive industry and many other industries have used natural fibres as their main materials. These industries aimed to use a product that is more lightweight, easy to process, and also low in cost (Peças et al. 2018).

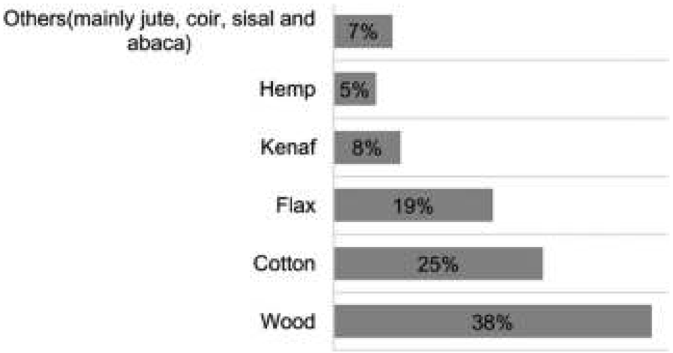

Figure 1. Europe's natural fibre usage in automotive production in 2012 (Peças et al. 2018).

Table 1. Automotive company that use natural fibre in vehicle parts (Furtado et al. 2012).

Automotive Company	Vehicle	Part
Audi	A2, A3, A4, A6, A8, Avant, Roadstar and Coupe	Seat back, side and back door panels, boot lining, hat rack, spare tire lining
BMW	3,5 and 7 series and others	Door panels, headliner panel, boot lining and seat back
Mitsubishi	Space star and Colt	Door panels, instrumental panels
Volkswagen	Golf A4, Passat Variant and Bora	Door panels, seat back, boot lid finish panel and boot liner
Ford	Mondeo CD 162 and Focus	Door panels, B-pillar, boot liner

Natural fibre is known for their strength. Natural fibre is one of the best replacements for synthetic fibre and artificial fibre in manufacturing because it is lightweight, biodegradable, and safe (Saba, Paridah, and Jawaid 2015).

There are two classes of natural fibre, which are organic and inorganic fibre. Organic fibre usually comes from living things, such as plants and animals, while inorganic fibre includes mineral fibres (Figure 2).

Akil et al. (2011) also stated that natural fibres are classified into three classes based on what they are derived from, which are plants or vegetables, animals and minerals. Fibres extracted from plants or vegetables are derived from cellulose which is the strongest part of the plant, while proteins which are hair, silk, and wool are normally extracted from animals. Most developers extract fibres from plants (Furtado, Silva, & Alves, 2012)Table 2.

The plant or vegetable fibre is popular among the production industries. Scientists and researchers have found that it has seven categories, which are fruit, seed, leaf, bast, wood, grass, and reed and stalk (Figure 3). Mwaikambo (2017) only emphasised four categories of plant fibres, which are seed, fruit, leaf, and bast fibres. Table 3 stated the classification of natural fibres.

2.1 *Physical, mechanical and thermal properties of natural fibres*

Kenaf, flax, and hemp are popular fibres that have been used as a manufacturing medium because they are easily found in Malaysia (Maslinda et al. 2017). Table 4 shows that kenaf fibre has the highest value of strength compared to hemp and flax (Mohamed et al. 2018; Sreenivasan et al.

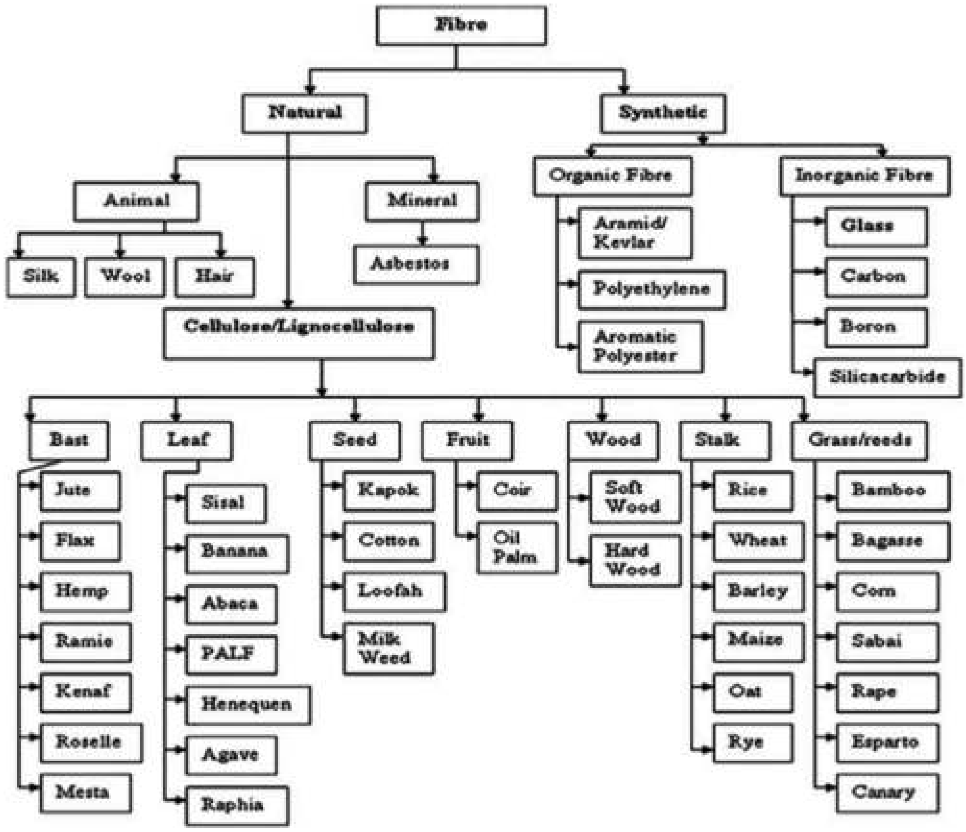

Figure 2. Classification of natural fibres (Siakeng et al. 2019).

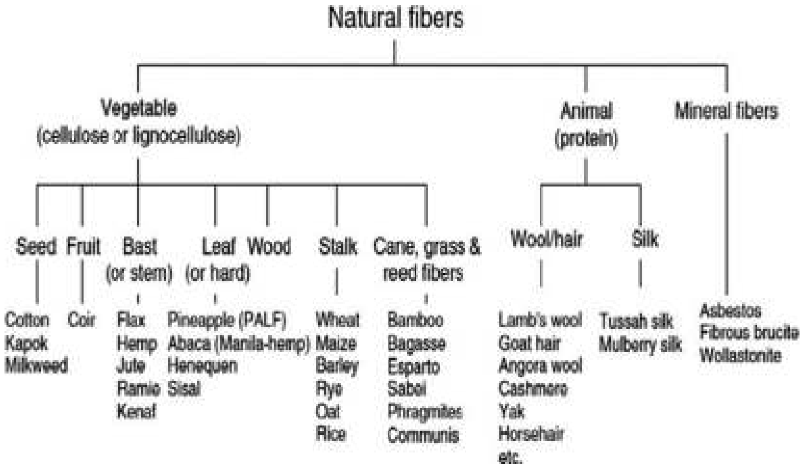

Figure 3. Classification of natural fibres (Akil et al. 2011).

2013; Nurul Fazita et al. 2017) while Table 5 is the thermal properties of the insulation materials.

A family of the Malvacea, *Hibiscus Cannabinus L.* or known as kenaf fibre (Figure 4) is one of the materials that can be used as fillers or reinforcements of bio-composite products (Akil et al. 2011; Tholibon et al. 2019). K. Gentian stated that kenaf only takes 4 to 5 months to grow, depending on the surroundings. Mohamed et al. (2018), on the other hand, stated that a kenaf plant grows in 150 days.

Core fibre and bast fibre (Tholibon et al. 2019) are types of kenaf elements (Mohamed et al. 2018). The kenaf fibre bast is known for its high aspect ratio and strength as a reinforcing agent and filler for a matrix polymer (Akil et al. 2011).

Most of the natural fibres have different chemical properties. The chemical composition of each fibre consists of major components, such as cellulose, hemicellulose, and lignin (Akil et al. 2011). Three of the natural fibres and their chemical properties are illustrated in Table 6.

One of the important components in plant is cellulose. Stability and strength of a plant are provided by cellulose (Kabir et al. 2012) via hydrogen bonds (Akil et al. 2011). Another chemical contained in plant is known as hemicellulose, which is responsible for moisture and water absorption,

Table 2. Fibres characteristic values for the tensile strength (MPa), young's modulus (GPa), elongation (%) and density (g/cm^3) (Akil et al. 2011; Mahjoub et al. 2014; Saba, Paridah, and Jawaid 2015; Siakeng et al. 2019; Sreenivasan et al. 2013).

Fibres	Tensile Strength (MPa)	Young's modulus (GPa)	Elongation (%)	Density (g/cm^3)
Cotton	287–800	5.5–12.6	3.0–10.0	1.5–1.6
Jute	393–800	10–30	1.16–1.8	1.3–1.6
Flax	345–1500	27.6	1.2–3.2	1.4–1.5
Hemp	550–900	70	1.6–4.0	1.47–1.48
Sisal	400–700	9.0–38.0	2.0–14	1.33–1.5
E-glass	2000–3500	70–73	2.5–3.4	2.50–2.55
Carbon (standard)	3400–4800	230–425	1.4–1.8	1.4–1.78
Kenaf	930	53	1.6	1.2–1.45
PALF	170–1627	60–82.5	1.6–2.4	1.56

Table 2 summarises characteristic values for the density and mechanical properties, of natural (plant), synthetic fibres and glass fibres.

Table 3. Natural fibre classifications (Sreenivasan et al. 2013).

Natural Fibre	Cellulose/ Lignocellulose	Bast	Flax, Hemp, Jute, Kenaf, Ramie
		Leaf	Abaca, Banana, Pineapple, Sisal
		Seed	Cotton, Kapok
		Fruit	Coir
		Wood	Hardwood, Softwood
		Stalk	Wheat, Maize, Oat, Rice
		Grass/ Reed	Bamboo, Corn
	Animal	Wool/ Hair	Cashmere, Goat hair, Horse hair, Lamb wool
		Silk	Mulberry
	Mineral	-	Asbestos, Ceramic fibres, Metal fibres

Table 4. Natural fibres' mechanical properties (Mohamed et al. 2018; Sreenivasan et al. 2013; Nurul Fazita et al. 2017).

Fibres	Density (g cm^{-3})	Diameter (μm)	Tensile strength (MPa)	Young's modulus (GPa)	Elongation break (%)
Kenaf	1.2–1.45	20–200	930	53	1.6
Hemp	1.47	25–500	690	30–70	1.6
Flax	1.4–1.5	40–600	345–1500	27.6	2.7–3.2

Table 5. Thermal properties of insulation materials.

Natural fibre	Thermal conductivity (W/mK) (Arenas and Asdrubali)	Thermal conductivity (W/mK) (Kumar Ghosh et al. 2016)
Hemp	0.04	0.038–0.040
Kenaf	0.044	-
Coconut fibre	0.043	-
Sheep wool	0.044	-
Wood wool	0.065	-
Cork	0.039	0.038–0.070
Cellulose	0.037	0.035–0.040
Flax	0.040	0.038–0.040
Wool	-	0.038–0.040
Jute	-	0.038–0.040

Table 6. Natural fibres' chemical properties (Nurul Fazita et al. 2017; Maslinda et al. 2017; Balla et al. 2019).

Fibres	Cellulose %	Hemicellulose %	Lignin %	Moisture %
Kenaf	72	20.3	9	6.2–12
Hemp	68	15	10	6.2–12
Flax	60.0–81.0	14.0–18.6	2.0–3.0	8–12

Figure 4. Kenaf fibre plants (Akil et al. 2011).

Table 7. Difference between thermoset and thermoplastic polymers (Kabir et al. 2012).

	Advantages	Disadvantages
Thermoset	Low resin viscosity Good fibre wetting Excellent thermal stability once polymerised Chemically resistant Recyclable	Brittle Non-recyclable via Standard techniques No post-formable Poor melt flow
Thermoplastic	Easy to repair by welding and solvent bonding Post formable Tough	Need to be heated above the melting point for processing purpose

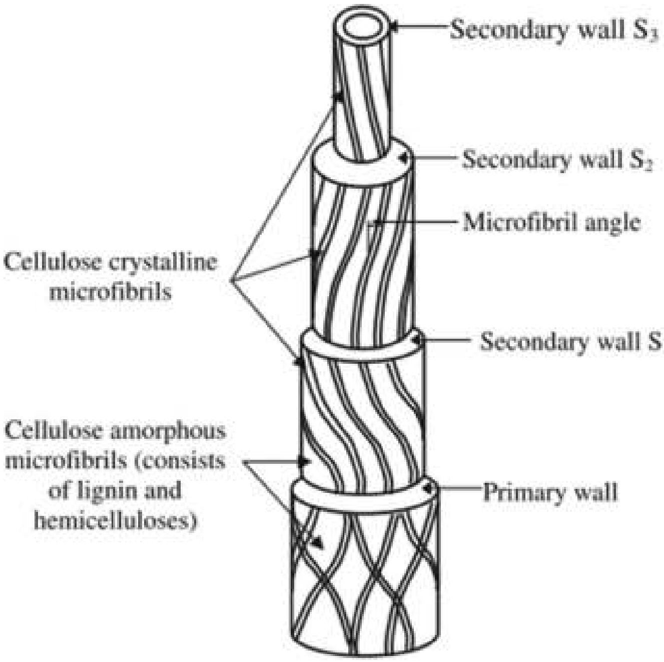

Figure 5. Structure of natural fibre (Kabir et al. 2012).

biodegradation, or fibre decomposition and thermal degradation. Lignin or pectin which has a cross linking structure (Kabir et al. 2012) is responsible for UV endurance and also thermal stability (Akil et al. 2011). A complete illustration of the natural fibre is shown in Figure 5.

Produce composites by combining polymer and natural fibre as filler, can produce the insulator materials, such as wire and cable wrapper. In Narayan Nayak (Narayan Nayak, Dr. Reddappa H. N, Ganesh R Kalagi, & Vijendra Bhat, 2017) paper has extract about electrical properties of natural fibre reinforced polymer. Dielectric strength is one of the important parameter in electrical which measure the withstand of voltage without breakdown. Along with the mechanical, physical and thermal properties, electrical properties also play an important role in producing composites. Mechanical is about the durability in term of tensile, bending, fatigue, and impact. Physical are about the durability of composites in moisture, flow, and density, while thermal properties are about how high the composite can withstand in certain temperature without degrade. In this paper also stated that 1.8–2.6 is the constant for dielectric constant that non-polar polymer lies and might be greater than that one for another polymer. Lesser the value of dielectric constant, the more efficient it might be (Narayan Nayak et al. 2017). But due to some of disadvantages of natural fibre especially the hydrophilic properties, it might increase the value of dielectric and lower the efficiency itself. In way to prevent this issue, the chemical treatment is done towards the natural fibre as to decrease the moisture absorption. By doing the alkaline treatment, not just can increase the efficiency in electrical insulator properties, but also in term of mechanical and physical properties (Narayan Nayak et al. 2017).

2.2 Potential polymer composites of FDM

Fibre reinforced polymer composites or known as fibre-reinforced polymer (FRP) is a new material introduced to replace the current materials in worldwide applications. It is also used with concrete and steel in building and construction. The composites have many advantages; they are lightweight, and have large strength value and specific modulus, making them a highly demanded material as car features, sports components, etc., (Saba, Paridah, and Jawaid 2015). Even though natural fibres are said to be the best replacement for synthetic or carbon, they have the disadvantage of attracting the hydrogen bond in composite production, causing swelling and change in dimensions. A polymer matrix has been introduced as a matrix that combines with natural fibres as a binder reinforcement to overcome this disadvantage. The use of polymer as a binder which has hydrophobic characteristics helps to repel the hydrogen molecules as illustrated in Figure 6 (McKeen 2016).

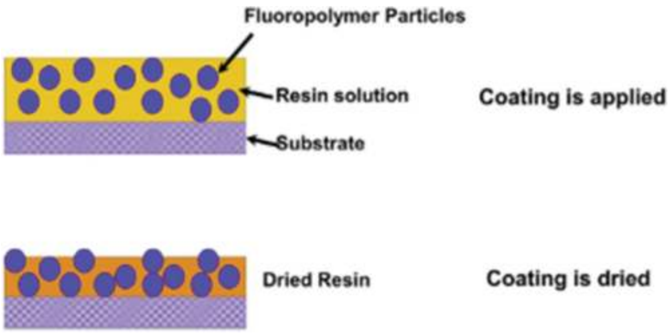

Figure 6. Polymer as a binder (McKeen 2016).

Table 8. Thermoplastic and thermoset mechanical properties (Furtado et al. 2012).

Resin		Density (g m^{-3})	Young's modulus (MPa)	Tensile strength (MPa)
Thermoplastic	Nylon	1.1	1.3–3.5	55–90
	PEEK	1.3–1.35	3.5–4.4	100
	PPS	1.3–1.4	3.4	80
	Polyester	1.3–1.4	2.1–2.8	55–60
	PC	1.2	2.1–3.5	55–70
	PTFE	2.1–2.3	-	10–35
Thermoset	Epoxy	1.2–1.4	2.5–5.0	50–110
	Phenolic	1.2–1.4	2.7–4.1	35–60
	Polyester	1.1.-1.4	1.6–4.1	35–95

The main function of a matrix as the binder of reinforcement is to provide a good grip to increase the strength of the composite and as a load transfer (Kabir et al. 2012). Table 8 shows the mechanical properties of several thermoplastics. Furtado, Silva, and Alves (2012) stated that there are three types of matrices, which are metallic, ceramic, and polymeric, which are mostly used in the production of unsaturated epoxy, polypropylenes, and polyesters. Polymeric or polymer can be divided into two group; thermosets and thermoplastics (Furtado et al. 2012). These thermosets and thermoplastic have different characters and chemical structures. The difference in characteristics is tabulated in Table 7 (Kabir et al. 2012).

Thermoplastic polymer produces the most biodegradable product because thermoplastics are recyclable (Furtado et al. 2012) and degradable. This polymer can generate a variety of edge shapes as their rheological properties are impressive. It also has viscosity at high temperature, low cost, and easy to manufacture. There are valid reasons for thermoplastics to be a favourable replacement for thermosets in the production industry (Ozsoy et al. 2017). The strength of thermoplastics is usually higher compared to thermosets as thermoplastics can absorb higher energy impact (Furtado et al. 2012). Thermoplastics melt when the polymer is heated, and the bonds between hydrogen molecules and Van der Waals are broken temporarily allowing the molecules to move (Peças et al. 2018). It has the same characteristics as metals; when the polymer is heated until its melting point, it melts and can be reused in another product. This process can be repeated (Dhinakaran et al. 2020) because thermoplastics are recyclable (Sreenivasan et al. 2013).

A thermoset polymer is in a three-dimensional (3D) linkage and known as a permanent structure that does not allow the reshaping or reprocessing of their structure, unlike thermoplastics. The permanent linkage shape occurrs when the thermoset polymer is already 'cured' and hard to recycle, but some findings have discovered that some thermosets can be recycled (Peças et al. 2018; Sreenivasan et al. 2013). Certain industries prefer to use thermosets rather than thermoplastic because thermosets resist water better than thermoplastics, and have good thermal behaviour. However, thermosets cannot be reprocessed or recycled, unlike thermoplastics (Furtado et al. 2012). Table 8 extract the mechanical properties of thermoset and thermoplastic. Polymer degradation is found to be

Table 9. High temperature of several thermoplastics (Muzzy and Kays 1984).

		Transition Temperatures	
Polymer	Symbol	T_g, °C	T_m, °C
Poly (butylene terephthalate)	PBT	40	228
Poly (ethylene terephthalate)	PET	80	265
Polysulphones	PS	190	^{b}
Poly (phenylene sulphide)	PPS	93	288
Poly (ether sulphone)	PES	230	^{b}
Poly (ether ketone)	PEEK	143	340
Polyimides	PI	>280	^{b}
Poly (ether imide)	PEI	210	^{b}

Where Tg: Temperature Glass Transition; Tm: Temperature Melting Transition; b: amorphous

a disadvantage and limits the value of demands in industries (Mohamed et al. 2018).

Due to their mechanical properties and thermo-oxidative stability at high service temperatures, thermosetting polymers are an obvious choice for advanced additive manufacturing. Many studies have proven that 3D printing can be successfully done by using thermosetting epoxy ink. The thermosetting polymer used chopped carbon fibre with a low-volume percentage, and by using clay as a shear thinning rheological modifier it is then extruded at room temperature. Before it is thermally cured, this method is used to hold the extruded shape. Compared with epoxy ink base, this formulation reflects the best mechanical properties in terms of modulus and strength values (B. G. Compton & J. A. Lewis, 2014). Applications in aerospace and defence are examples that extend the combination of traditional high-grade composite technology with additive manufacturing techniques. Flexibility in material, part geometry, cost, and lead time are examples of four additive systems illustrated in the repair applications for a complex and unique structure. Additive systems are ideal for unique or low-volume aircraft parts because part replacement of an aircraft can cost thousands of dollars (Hiemenz, J., 2013).

J.D Muzzy has stated in his review that the most delicate thing to be considered for thermoplastic is their melting temperature because the melting points of the thermoplastics are different considering their type of polymers, environment, and applications. The polymer is processed based on the temperature required by the customers. The maximum temperature for the different types of thermoplastics are shown in Table 9 (Muzzy and Kays 1984).

By using biodegradable polymers like thermoplastic and a mix of natural fibres, environmentally friendly bio-composites will be produced. As the biodegradable materials are blended, they can be used to make more products such as car doors, sports equipments, daily products, and so on. An example of biodegradable polymer is polylactic acid (PLA) that received a lot of awareness because of its properties as a renewable source (Mohamed et al. 2018). PLA resins can increase the contact area of the mechanical properties and produce sustainable bio-composites (Saba, Paridah, and Jawaid 2015).

2.3 *Enhanced bonding between fibres and matrix*

Silane, alkali, acylation, benzoylation, malleated coupling agents, permanganate, acrylonitrile and acetylation grafting, stearic acid, peroxide, isocyanate, triazine, fatty acid derivative, sodium chloride, and fungi are examples of various types of chemical treatment that are available (Mohamed et al. 2018; Akil et al. 2011). The main purpose of the treatment is to increase fibre-matrix interfacial bonding and stress (Mohamed et al. 2018). To enhance the matrix-fibre adhesion, chemical treatments are used by increasing the roughness through a fibre surface cleaning process to remove any impurities and by disrupting the moisture absorption process through of coat of -OH groups in the fibre (Krishna and Kanny 2016). In other words, by modifying the surface and cleaning the fibre surface, reduction of the moisture absorption process and upsurge of surface unevenness can be done by a chemical treatment or known as pre-treatment (Saba, Paridah, and Jawaid 2015). The chemical modification method or the treatment of natural fibre surface including kenaf, is carried out using reagents that contain functional groups that are capable of bonding with the hydroxyl group from the natural fibre itself (Akil et al. 2011). Fiore, Bella, and Valenza (2015) stated that the chemical method in modifying the natural fibre involved introducing a material that is compatible with both fibres and matrix.

Natural fibres that use the method of chemical surface modifications are well documented in the literature, including alkaline treatment (Mahjoub et al. 2014). One research was on alkalisation which consists of treating kenaf fibre using an alkaline solution to remove lignin, pectin, and waxy substances and natural oil covering the external surface of the fibre cell wall to improve the mechanical ability and physical ability of the natural fibre (Oushabi et al. 2017; Sreenivasan et al. 2013) and also reduce the fibre diameter (Fiore, Bella, & Valenza, 2015). The chemical treatment that uses an alkaline solution is also known as mercerisation (Sreenivasan et al. 2013). This treatment increases the interfacial bonding strength between lignocellulosic fibres and thermoset resins (Fiore, Di Bella, and Valenza 2015). Alkaline treatment or mercerisation is a well-known chemical treatment of surface modification of natural fibre reinforced polymer composites (Mahjoub et al. 2014). One of the most familiar and effective alkaline solutions applied in kenaf fibre is sodium hydroxide (NaOH) solution (Akil et al. 2011; Kabir et al. 2012). This solution is normally used for kenaf reinforced thermosets and thermoplastics polymer composites. The addition of sodium hydroxide in an alkaline treatment to the natural fibre promotes the ionisation of the hydroxyl group on the alkoxide (Akil et al. 2011). Figure 7 shows the equation of natural fibre and alkaline solution (Mahjoub et al. 2014).

The natural fibres were immersed in an alkaline solution for a limited time (Mahjoub et al. 2014) and as a result, the researchers discovered that the alkaline treatment had improved the mechanical properties of kenaf fibres (Saba, Paridah, and Jawaid 2015). The fibre surface finish became more uniform due to the elimination of micro voids, thus the stress distribution capacity between the ultimate cells were

$$\text{Fiber}{-}\text{OH} + \text{NaOH} \rightarrow \text{Fiber}{-}\text{ONa}^+ + \text{H}_2\text{O}$$

Figure 7. Equation of natural fibre and alkaline solution (Mahjoub et al. 2014).

Table 10. Parameters of alkaline treatment.

Sources	NaOH concentration	Time immersion	Time to dry
S. A. N. Mohamed (Mohamed et al. 2018)	6%	-	-
V. Fiore (Fiore, Di Bella, and Valenza 2015)	6%	48 h and 144 h	100°C at 6 h
A. Oushabi [13]	0%, 2%, 5% (optimum), 10%	-	-
R. Mahjoub (Mahjoub et al. 2014)	5% (optimum), 7%, 10%, 15%	1 h, 3 h and 24 h	-
A. M. M. Edeerozey [14]	3%, 6% (optimum), 9%	3 h	Room
temperature 24 h			
S. Scrrnivasan (Sreenivasan et al. 2013)	9%	-	-

improved (Kabir et al. 2012). It was noted that increasing the alkaline concentration can damage the natural fibres, resulting in a decreased mechanical (Mohamed et al. 2018; Kabir et al. 2012) and physical properties. As a conclusion, alkaline treatment is the most effective treatment compared to others because an alkaline solution can stand for a long period of time and has a lasting effect on the mechanical behaviour of the natural fibres, especially on their strength and stiffness (Akil et al. 2011).

The optimum concentration for alkaline treatment is 6%, consistent with the literature review.

From Table 10, it is proven that a 6% concentration gives the best result in mechanical and physical properties. The hydrophilic properties of the kenaf fibre can be reduced and enhance the interfacial bonding between fibre and polymer itself.

V. Fiore (Fiore, Di Bella, and Valenza 2015) stated in her findings that pre-treatment of kenaf fibre in 6% NaOH solution in a water bath led to the best results. Other than that, S. A. N. Mohamed (Mohamed et al. 2018) also reported that an alkalisation treatment with sodium hydroxide improved the mechanical properties and good stress dispersion of the kenaf fibre compared to an untreated kenaf fibre. The report also stated that the optimum value of concentration showed good results for the chemical treatment methods and led to greater interfacial bonding. Figure 8 shows the SEM micrograph, which illustrates that the immersion time in a NaOH solution affects the surface of natural fibre significantly. Figure 8(a) shows the fibre not treated with NaOH solution, Figure 8(b) shows the fibre immersed for 48 hours in NaOH solution, while Figure 8(c) shows the surface of the natural fibre that rotted due to long period of immersion (Mohamed et al. 2018).

Torrado, David, and Wicker (2014) stated that the use of silanes in addition to surface modification after the alkaline treatment improved some of the minor factors such as dispersion and adhesion of reinforcement and polymer matrix. In a past experiment, Petchwattana et al. (2019) stated that a silane coupling agent enhanced the bonding interaction between wood flour hydrophilic and PLA polymer hydrophobic. Figure 9 shows the bonding between fibre and PLA polymer untreated and treated with a silane coupling agent. Figures 9(a,c) show poor interfacial adhesion between fibre and polymer with untreated fibre, while Figure 9(b,d) show good interfacial bonding with treated fibre.

3. Filament for FDM

Additive manufacturing or known as AM is one of the technologies that offers potential savings, prevents material waste, and increases product variety with complex geometries. In Fused filament fabrication (FFF) is one example of additive manufacturing technology (Brenken, Barocio, Favaloro, & Pipes, 2018). Figure 10 shows the classification of rapid manufacturing method. Before using 3D printing method, several methods that been introduced to produce geometrical shape such as sand casting and injection moulding. Other than using 3D printing also, several conventional methods have been introduced, such as rapid prototype (RP) by using direct laser fabrication. Table 11 demonstrate in detail example of product that produce using different fabrication process.

In (Sithole, Nyembwe, and Olubambi 2019) reviewed about sand casting process by using molten metal and sand to produce small parts. The advantage by using sand casting is the material itself can be recycled and reused which can save cost. The process is first let the metal melts and pour into mould, which compact with sand. After the metal solidifies, the product will be taken out from the cast. This sand-casting method is popular in producing small and medium parts.

Injection moulding involves mixing between two phases, which is metal and polymer as binder. In (Dehghan-Manshadi et al. 2020) has extract the injection moulding process and stated the parameters that need to be list out before run the process as example the injection speed, temperature and also pressure. The shape that produces using injection moulding is determined by selection of dies and tools that include the

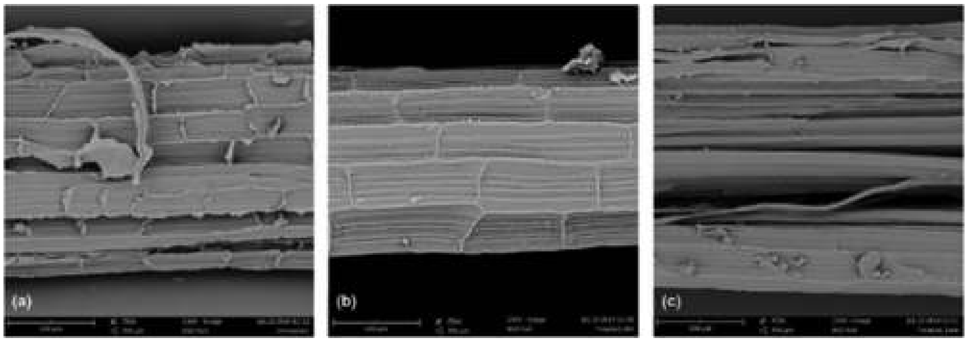

Figure 8. NaOH solution SEM (Mohamed et al. 2018).

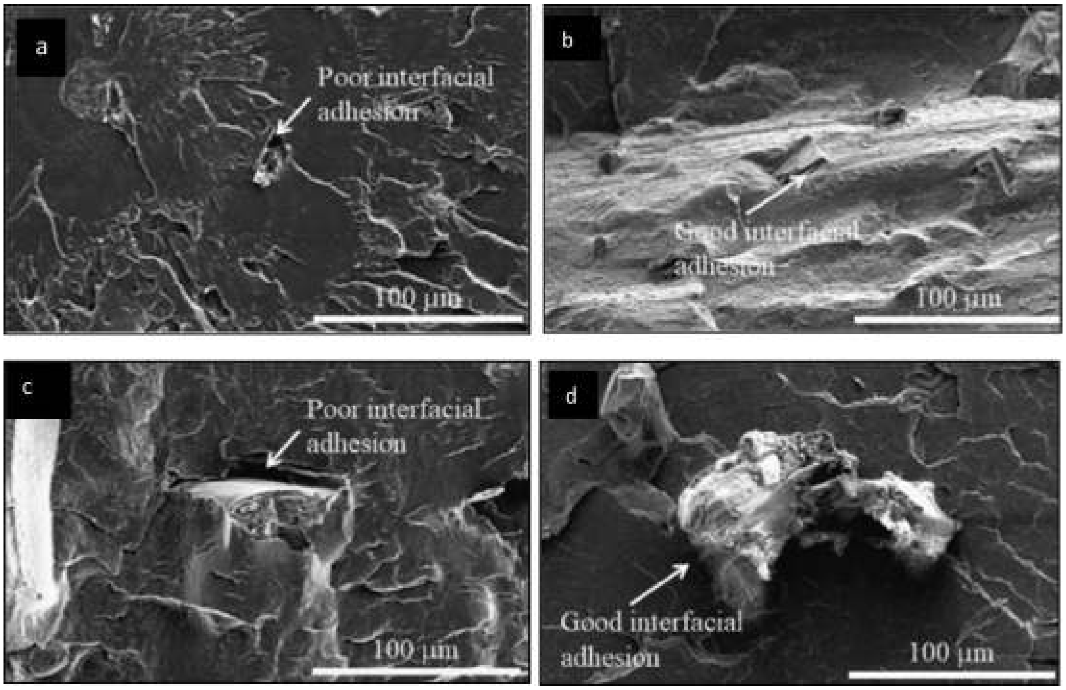

Figure 9. SEM (Petchwattana et al. 2019).

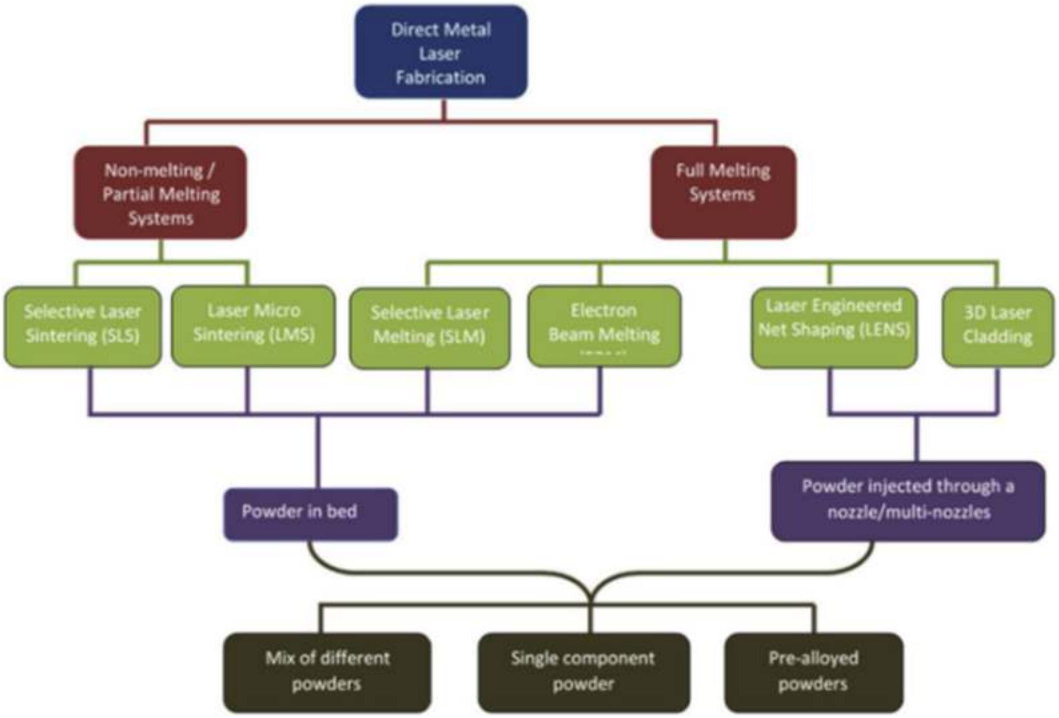

Figure 10. Rapid manufacturing methods using direct laser fabrication classification (Ahmed 2019).

design respect to the part of geometries and characteristic of the shape.

Other than using 3D printing that define the process layer by layer through heating the filament, rapid prototype (RP) has introduced direct laser built through layer by layer and the parts that design also use 3D CAD same like 3D printing process without using dies (Ahmed 2019). Naveed's paper also stated that using laser printing have several materials that can be used which are polymer, paper, plastic, wax, and metal powders. Laser rapid forming (LRF) is one of the process that develop under rapid prototype (RP). Below is a figure that demonstrate the process that using LRF as a process.

Selective laser sintering/melting (SLS/SLM) one of manufacturing process that well known. Olakanmi (Olakanmi, Cochrane and Dalgarno, 2015) write about SLS/SLM method that use aluminium allow powder for this metal and also extract the method by using this process. By using CAD drawing to create parts and through laser energy application

Table 11. Different type of fabrication process.

Manufacturing process	Materials	Process	Ref.
Sand-casting	Metal molten	The sand-casting process requires moulding, metal melting, solidification, shake out and finishing.	(Sithole, Nyembwe, and Olubambi 2019)
Injection moulding	Metal polymer composites	The process involves mixing thoroughly metal and polymeric binder, injection moulding, solution and thermal de-binding and sintering	(Dehghan-Manshadi et al. 2020)
Rapid protoptype Example: SLM/SLS	Polymer, paper, plastic, wax, and metal powders	Using direct laser fabrication to melt the materials in 2D layer which bonded and form complete 3D product	(Ahmed 2019)

towards powder beds. The cross-sectional area, which requires laser spot and to melt the materials, sinter and particles were bonded together in a thin lamina, then the process is repeating until layer by layer up until the product is obtain.

Fusing or deposition layer by layer is process that had been used in 3D printing(Mazzanti, Malagutti, and Mollica 2019; Pu et al. 2020; Sezer and Eren 2019). Additive manufacturing (AM) and rapid prototype (RP) are other name for 3D printing(Pu et al. 2020). Before FFF was introduced, fibre reinforced polymer (FRP) manufacturing methods (Dhinakaran et al. 2020) like vacuum-assisted resin transfer moulding (VARTM), hand layup (which is a popular method because of low cost), compression moulding, and autoclave curing had been used. The FFF process has overcome many minor factors from using the previously manual method, such as using a hand roller tool to reduce bubbles and gaps.

Peng Wang and Bin Zhou (Wang, Zou, and Ding 2019) previously emphasised that thermoplastic is one of the main extrusion in fused deposition modelling (FDM) in printing technology as shown in Figure 11. This printing technology has been used in many industrial platforms for its ease in printing products that consists of small dimensions and hard geometries. In the last few years, FDM is a popular process in aerospace, biomedical, mechanical, and electrical industry productions for its good mechanical and thermal properties that are convenient to the users and consumers (Wang, Zou, and Ding 2019).

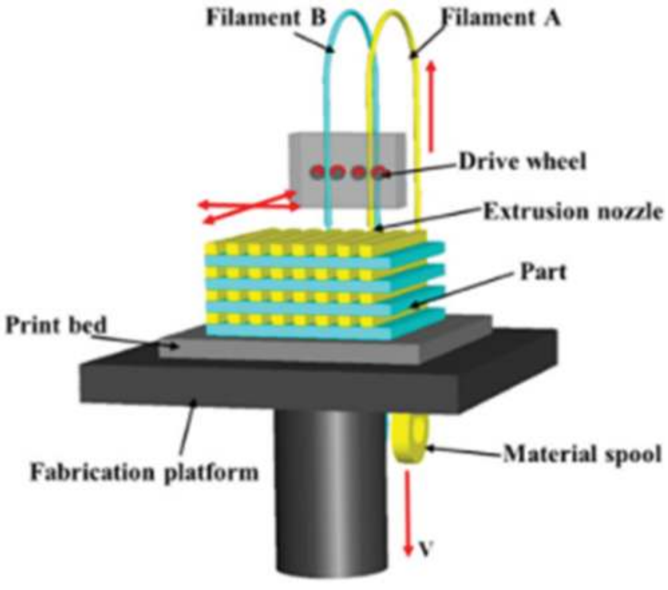

Figure 11. FDM process diagram (X. Wang et al. 2017).

In paper (Pu et al. 2020) stated that in year 1999, medical equipment has used FDM printing technique and expected further development in biomedical industries. Such of printing design that used 3D printing as main platform by producing surgical tools, implants and fixtures. Rather than using milling lathe and any traditional method in producing tools, 3D printing has outstanding finishing in term of complex parts and also can save time. In 3D printing development design, software such as computer-aided design (CAD) has been used(Pu et al. 2020; Sezer and Eren 2019) and been converted into stl. format and transfer into 3D machine. Thermoplastics, such as acrylonitrile – butadiene styrene (ABS) and polylactic acid (PLA), are the most common product that been used in FDM where the process can control parameters such as temperature and PLA and ABS are suitable in terms of rheological and thermal.

Next, (Sezer and Eren 2019) has wrote about multi wall carbon nano tubes reinforced ABS composites in FDM 3D printing parts and stated that it can enhanced the mechanical and also electrical properties of the composites. The process involves CNT as the main filler which the disadvantages of this addition can increase the strength of composites and improve electrical/heat conduction and other mechanical properties that correlated. Twin screw extruder was used in mixing both filler and polymer matrix and produce the filament for printing purpose. The samples are generated by using 3D printing following a guide from ASTM. The different parameter in this process is the direction of printing and also the weight percent of the filler. As conclusion, for electrical conductivity, the higher the percentage of filler in composites, the better electrical conductivity will obtain and also the different printing direction been studied and effect the final results (Sezer and Eren 2019).

The most popular development now is the printing technique using short or natural fibre particles. This printing technique is used in the manufacturing product platform to replace carbon fibre and glass fibre reinforcements. Natural fibre has poor physical properties as a reinforcement, thus, researchers have done many experiments to improve the interfacial bonding between reinforcement and polymer and surface treatment on natural fibre to remove the chemical content that may lead to poor mechanical properties. Therefore, by using natural fibre reinforced polymer in a printing platform, an environmentally friendly filler filament can be achieved and a bio-waste product can be developed (Rahim, Abdullah, and Md Akil 2019)

In their past paper, Peng Wang and Bin Zhou (Wang, Zou, and Ding 2019) used neat poly-ether-ether-ketone (PEEK) as a filament in an FDM process, as illustrated in Figure 12. In this finding, they studied the behaviour of this polymer and noted that it had a high melting temperature of over 334°C, high chemical resistance, and excellent thermal stability, but had poor rheological behaviour at high temperatures.

Figure 12. Neat poly-ether-ether-ketone (PEEK) (Wang, Zou, and Ding 2019).

Bates-Green (Wa and Wa 2017) used pure PLA and acrylonitrile butadiene styrene (ABS) as printing filaments and studied the difference between the two materials. They found that the glass transition of ABS is higher (110°C) than PLA (65°C). The high glass transition of ABS leads to the possibility of product shrinkage. PLA has a low glass temperature, therefore, it is much stiffer than ABS. ABS produces toxic fumes and can cause minor headache, unlike PLA, which is toxic free.

Nawadon Petchwattana (Petchwattana et al. 2019) developed natural fibre reinforced polymer using wood flour and PLA polymer, in producing FDM printing filament. They used particles of different sizes (74 μm and 125 μm), fibre with different weight percentages (1 wt%-5 wt%), and treated and untreated fibre with a silane coupling agent in the study. The results showed that 74 μm fibre treated with 5 wt% showed positive results, as its optimum fibre dimension can prevent porosity, and the optimum weight percentage acts as a barrier and good interfacial bonding between fibre and polymer to remove impurity by using a silane coupling agent.

In another work, Xin Wang (X. Wang et al. 2017) used titanate ($BaTiO_3$)/ABS, Al and Al_2O_3/Nylon-6, $BaTiO_3$/ABS, $CaTiO_3$/Polypropylene, Tungsten/PC $BaTiO_3$/ABS and the same polymer, which is ABS. The addition of copper and iron particles as a printing filament can improve the thermal expansion and reduce the distortion of final finishing in printing products. Aluminium can reduce the coefficient of friction, and by using $BaTiO_3$, improves dielectric permittivity.

3.1 Biodegradable natural fibre composites as filament Fused Deposition Modelling (FDM)

Fused Deposition Modelling or FDM as shown in Figure 13, is an application that is commonly used in industries such as print prototypes and wide applications such as automotive, airplane parts, and medical devices (Stoof, Pickering, and Zhang 2017). FDM makes for shaping 3D objects or shaping complicated parts easier. It is also popular in the consumer market for the past few years for its affordable. This method can also be applied for human tissue and organ modelling (Stoof, Pickering, and Zhang 2017). This technology is still in the enhancement and improvement stage, especially product printing, for better manufacturing. FDM is a reliable and low-cost method (Rahim, Abdullah, and Md Akil 2019) making it a high range of additive manufacturing technology (Stoof, Pickering, and Zhang 2017). The reason this technology is reliable is that it is easy and simple to operate to achieve effective products and environmentally friendly (Rahim, Abdullah, and Md Akil 2019). Some shapes and geometries including those with small dimensions or sizes are complicated to produce or obtain through standard methods of polymer manufacturing, thus, additive manufacturing technology is one of the methods that can help to fabricate this kind of product. The rapid diffusion in both industry world and household settings is the result of additive manufacturing techniques' valuable characteristics (Mazzanti, Malagutti, and Mollica 2019).

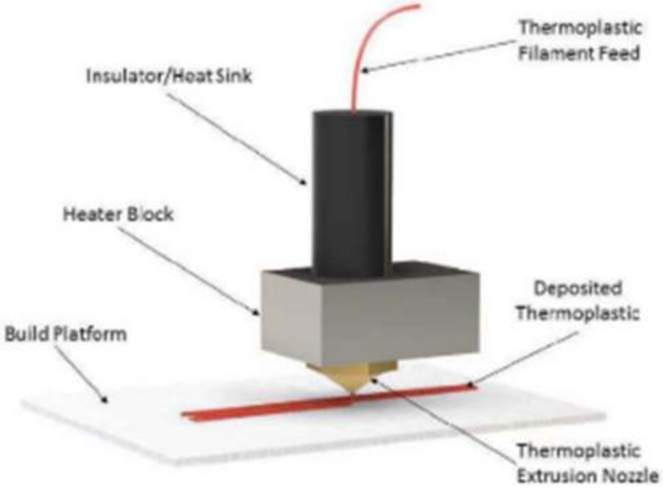

Figure 13. Schematic of FDM (Nagaraju 2019).

The operation of the FDM technology needs a few steps. Petchwattana et al. (2019) has identified five steps that are needed to design any 3D design software. The first step is to develop an accurate measurement and overall product. Next, the design from the software is sent to the 3D printer and the filament will be extruded through the die. The molten material is then stacked into layers. Lastly, the product is removed from the 3D printer machine. Samples of bio-composites by using the FDM method are low in strength compared with the bio-composites that endured extrusion, compression, and injection moulding processes. The parameters and processes are, therefore, improved to produce samples that have low porosity with suitable fibre content.

All in all, the production of natural fibre is a new issue that has been introduced by many researchers. Natural fibre reinforced polymer bio-composites using environmentally friendly FDM technology has attracted many parties in industries and also researchers. The implementation of using natural fibres in the filament of FDM to replace the current fillers has attracted many competitors and market platforms (Stoof, Pickering, and

Zhang 2017). The most popular polymer that acts as the main material in FDM is acrylonitrile butadiene styrene (ABS). However, the use of thermoplastics polymer as the main material for FDM is still not recommended. The important elements of a polymer are its mechanical properties, which are strength and stiffness. As stated before, the mechanical aspects of many based polymers have been investigated to enhance the technology of FDM. Acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA) are popular because they are stable. The most frequent thermoplastic that had been produced in this technology is PLA. The advantages of using PLA are it is recyclable and biodegradable and has a temperature of 145–160°C (Petchwattana et al. 2019). PLA is one of the biopolymers that is obtained from the fermentation of the recyclable product and has good mechanical properties such as tensile strength and low thermal stability that prevents crystallisation (Coppola et al. 2018). Figure 14 illustrates the filament of neat PLA polymer and the filament that combines hemp (natural fibre) and PLA polymer.

The finishing of a product will be affected by the thermoplastics that had been chosen in FDM as different selected polymers contain different properties and characteristics. Glass transition temperature (Tg), melting temperature (Tm), and coefficient of thermal expansion (CTE) are examples of the polymer properties that need to be considered and had already been discussed in Table 10 (Coppola et al. 2018). The temperature of the extruder while printing the polymer is affected because of the melting temperature of the polymer matrix. Hence, the adhesion or viscosity during printing is relatable due to Tg and CTE, as how much thermal stress is developed is important, in parallel with the strength, ductility, and solvent resistance (Manickavasagam et al. 2018).

Some authors have made an analysis regarding natural fibre as fillers for reinforced bio-polymer composites (Coppola et al. 2018). Researchers found that using thermoplastic bio-composite filaments is one of the steps that can be taken to address the industrial sector's concern on the importance of sustainable and renewable materials. As all the technical productions are using 3D printing technology in line with decreasing cost and increasing quality of life, the waste of the product might also increase. As a result, developers have come out with a new invention that uses biodegradable materials as the fillers. Table 12 states several applications that use FDM as the main application in some of the industries' platforms (Rahim, Abdullah, and Md Akil 2019).

Figure 14. PLA polymer and PLA/hemp (Coppola et al. 2018).

Table 12. Summary of several applications (Rahim, Abdullah, and Md Akil 2019).

Segment	Applications
Automotive and aircraft	Fibre reinforced composites structural components, degradable bio-composite structure, unmanned aerial vehicle, rapid tooling, prototype for mechatronic control unit.
Electronic	Electrically conductive structure, smart interphase.
Medical and dentistry	Nasal prosthesis, biomedical implantable devices, lumbar cage, scaffold for tissue engineering, biosensor.
Pharmaceutical	Patient-tailored/personalised tablets, thermo-labile drug.

As previously stated, PLA is getting attention as a biodegradable and renewable plastic. It is also environmentally friendly and the studied between natural fibre as reinforcement such as hemp and kenaf which combine with PLA using standard method also has been done (Mazzanti, Malagutti, and Mollica 2019). The fibre loading optimisation and also the chemical treatment of the reinforcement can affect the mechanical results of the product; hence, the natural fibre that combines with the PLA is hard and requires dried feedstock and storage (Mazzanti, Malagutti, and Mollica 2019).

A filament that is from the cured bio-polymer composites is fed into a small heated chamber. The filament then melts and becomes a high-viscous fluid and molten polymer. The melted polymer is extruded through the nozzle and ejected layer-wise on a heated table following patterns that have already been synced into the CAD software to achieve the correct dimension and geometric shapes (Mazzanti, Malagutti, and Mollica 2019).

3.2 Extrusion of filament

Figure 15 shows a schematic diagram of the filament extrusion with fibre content A journal article by Rahim, Abdullah, and Md Akil (2019) had emphasised the factor of printing filament

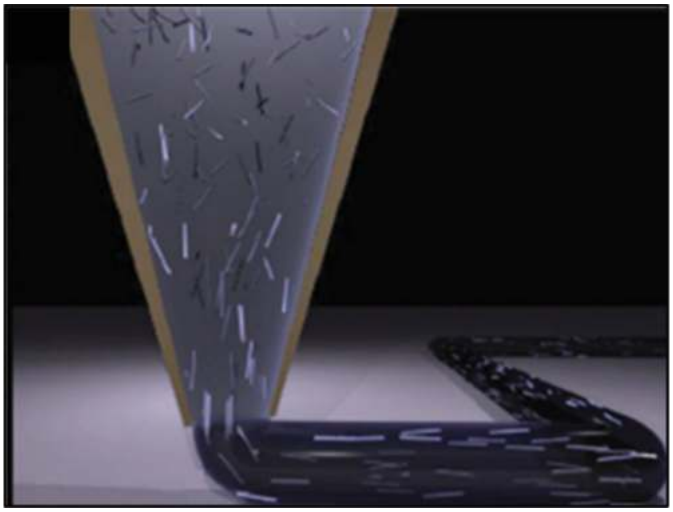

Figure 15. Schematic diagram of extrusion of filament with fibre content (X. Wang et al. 2017).

that needs to be considered. It stated that filament diffusion modelling is hot processing, which requires the filament to be extruded and printed layer by layer. The extrusion process requires the filament to be 1.75 mm or 3 mm in diameter.

The larger the diameter the stronger the filament (Rahim, Abdullah, and Md Akil 2019). However, a bigger filament has some disadvantages. The FDM process needs a higher pressure to push the filament into the nozzle and might have to stop if the filament is stuck. A bigger filament is also less flexible to rotate in the tube. A diameter of 1.75 mm is claimed to be the optimum size of FDM's filament, which requires less pressure and also prevents the nozzle from clogging.

The FDM process requires a temperature of 250°C–500°C depending on the type of printer used. After the printing process, the extruded semi-molten filament, which is called a bead, will return to a solid state. The cooling temperature is important to prevent shrinkage as the cooling process happens. However, not all semi-crystalline polymers are suitable for the FDM process (as illustrated in Figure 16) because some of them have a higher degree of crystallinity, which can lead to a higher degree of shrinkage and distortion of products. It can be concluded that challenges and unexpected issues exist in this process to produce the best new materials (Rahim, Abdullah, and Md Akil 2019)

Noraihan et al. (2019) also verified that a filament of 1.75 mm in diameter is preferable in the FDM printing process with a diameter tolerance of 0.01 mm. The tolerance takes into account serious issues that might arise during maintenance. For example, a small filament size can cause the gripping filament in the extrusion to fail, while a filament that is too wide for the nozzle would not be pushed out by the motor.

In a nutshell, product printing is a success if the following are met: 1) the suitable materials for filament are chosen, and 2) the parameters are set correctly during the FDM process (Rahim, Abdullah, and Md Akil 2019). Lastly, J. I. Montalvo N, M.A claimed that a good filament has good interfacial bonding between two phases from the surface modification.

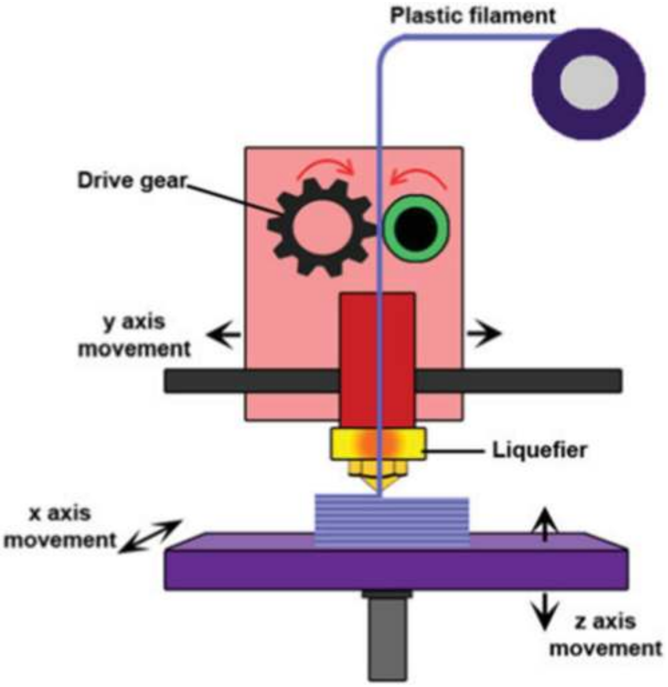

Figure 16. Schematic diagram of fused deposition modelling (Rahim, Abdullah, and Md Akil 2019).

Netravali (2004) stated that natural and synthetic polymers derived from plants and animals are called natural resins. As raw natural fibre might not satisfy some of the properties in the industries' platforms, a modification or blending process is done to enhance the mechanical, physical, or thermal properties of the raw fibre to make it easy to process. Some of the polymers might degrade due to enzymatic reactions in the environment and decompose naturally, while some polymers might degrade by the presence of alkali or acid. This degradation process makes us understand that the polymer that can degrade in any condition leads to green technology. Table 13 classifies the polymer into two (Netravali 2005).

In a journal article, Chun-Ying Lee and Chung-Yin Liu (Lee and Liu 2019) modified the 3D printer, as shown in Figure 17, and observed the effect of cooling airflow speed on the printing orientation of the samples (vertical and horizontal). The material used was PLA with a filament parameter of 1.75 mm. The filament was printed out in two directions, horizontally and vertically. The journal found that the speed of cooling airflow

Table 13. Polymer classifications.

Natural	Synthetic
Polysaccharides	Poly(amides)
Starch	Poly(anhydrides)
Cellulose	Poly(amide-enamines)
Chitin	
Pullulan	
Levan	
Konjac	
Proteins	Poly(vinyl alcohol)
Protein from grains	
Collage/gelatin	
Casein, albumin, fibrogen, silks, elastin	
Polyesters	Poly(ethylene-co-vinyl alcohol)
Polyhydroxyalkanoates, copolymers	Poly(vinyl acetate)
	Polyester
	Poly(glycolic acid)
	Poly(lactic acid)
	Poly(caprolactone)
	Poly(orho esters)
Other polymers	Poly(ethylene oxide)
Lignin	Poly(urethanes)
Shellac	Poly(phosphazines)
Natural rubber	Poly(acrylated)

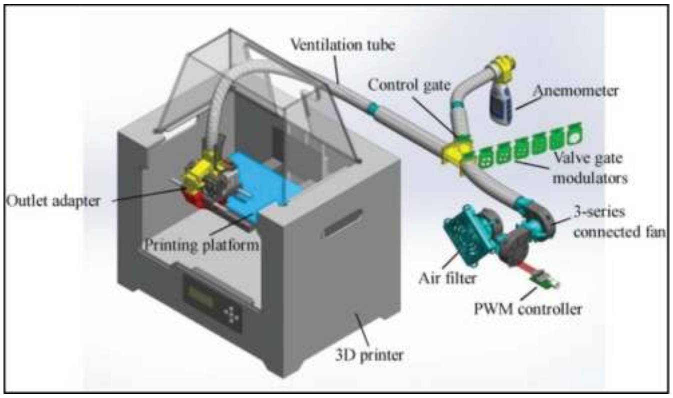

Figure 17. Modified printer (Lee and Liu 2019).

can affect the strength of the sample itself. The higher the speed of airflow, the lower the strength of the mechanical properties. This is due to the change in the crystallinity of thermoplastics. Also, the cooling process of the sample can affect the microstructure of the sample and the interaction between the bonding.

In the experiment, they used three-series connected fans to provide airflow at 5 m/s. If the airflow exceeds the value of airflow provided, it may be hard to print consistently. As such, a pulse width modulation (PWM) was provided to ensure the airflow did not go beyond the limit. Next, six different parameters of valve gate control, which were 1, 2, 3, 4, 5 m/s, were determined. The paper stated that an infrared picture near the printing head was captured during the extrusion process. It is illustrated that the temperature of PLA decreased as it left the printing nozzle. It can be concluded from the graphic picture and captured data that as the temperature of PLA increased, the airflow needed also decreased. The temperature of the vertically printed coupon became slightly lower than the horizontal one as the cooling airflow velocity was higher. This was because the glass temperature was lower, requiring a printing temperature of 60°C to prevent the damage of printing finishing (Lee and Liu 2019).

Tensile resulted in two different directions of printing with the cooling fan turned off, and the axial direction achieved higher results than the transverse direction. This showed that the direction of printing influenced the mechanical results. But, as the airflow was increased, the result of tensile strength in each direction decreased .

Figure 18 shows the different direction and speed of airflow during the printing process. It can be seen that airflow does not influence the finishing in horizontal direction. Compared to the horizontal direction, the axial direction takes a longer printing process. The bonding strength between particles and printed filament adjacent factor is influenced by airflow velocities. A cooling process that is quicker with larger airflow velocities results in many voids, as illustrated in Figure 18. There were more voids in the 5 m/s specimen compared with the 0 m/s specimen. This void content will influence the mechanical properties, especially in the horizontal direction (Lee and Liu 2019).

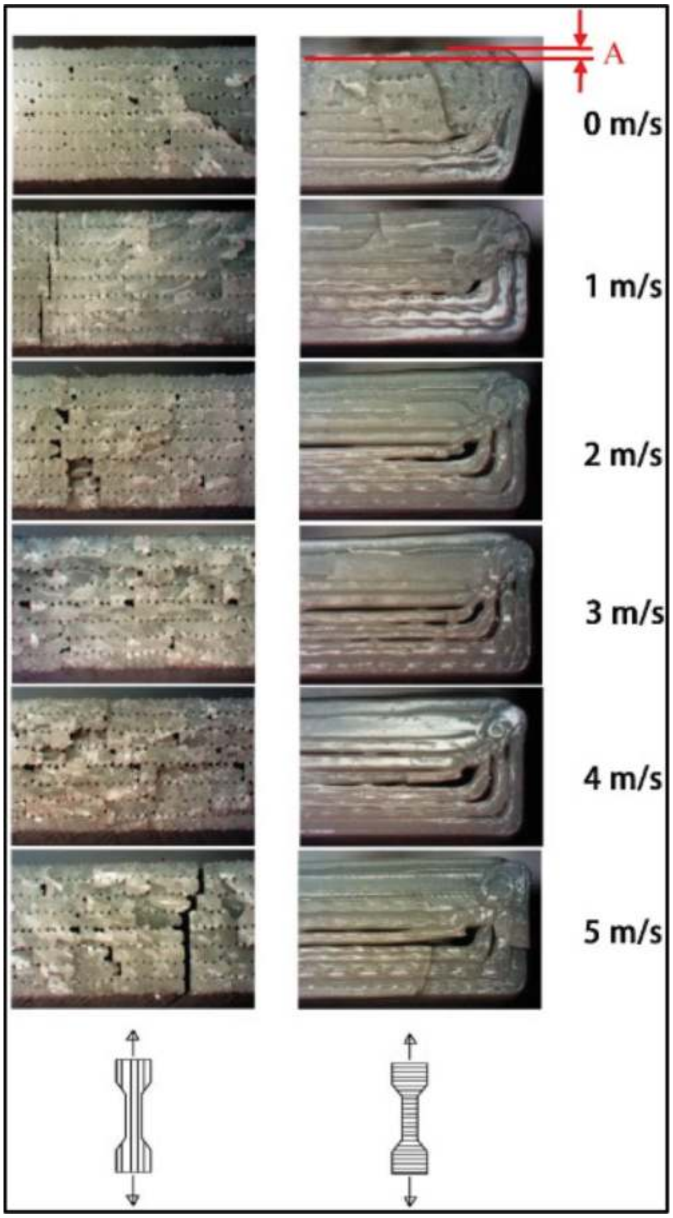

Figure 18. PLA printing with different direction and speed of airflow. (Lee and Liu, 2019)

3.3 Parameters of printing process

Thermoplastic is a raw material that is used in the FDM process to transform solid into liquid the size of the nozzle (Zhong et al. 2001; Brenken et al. 2018). Zhou's paper (Zhou et al. 2020) stated that there were four major parameter that need to be considered in the printing process; hopper temperature, temperature of the printing table, layer thickness, and speed of deposition. The explanation of each parameter, which were extracted from Noraihan's paper, is illustrated in Table 14 (Rahim, Abdullah, and Md Akil 2019). The aim of this paper was to compare the mechanical characteristic between printed neat polymer and printed natural fibre reinforced polymer form the major past researches.

From the table, we can configure that the size of the fibre was the main factor that can clog the nozzle. Nozzle clogging happens when the fibre gets stuck at the nozzle. Figure 19 defines the prototypes that are affected because of nozzle blockage. It can be seen that the finishing of the surface was not smooth and the fibre was porous. The worst effect is failure of the product due to the changes of the mechanical properties (Rahim, Abdullah, and Md Akil 2019).

Next, the coefficient thermal temperature is also important to prevent the shrinkage of the composite's product. Brenken et al. (2018) stated that most of the sharp corners of the product cool first, followed by the centre of the product. If the material cools faster than it should, the product could fluctuate off from the platform printer bed. The sample is as in Figure 20.

In Table 9, the cooling temperature emphasised on the temperature taken for the printed materials to solidify. The temperature of the printed layers is important to prevent sagging of materials. In other words, if the printed materials

Table 14. Major factor of FDM parameters (Rahim, Abdullah, and Md Akil 2019).

Parameters that need to be considered	Reason(s)	Explanation(s)
Fibre loading	Wetting problem of composites	Dimension stability
	Strength of the composites	Too stiff if there is more fibre and too brittle if lack of fibre
Fibre orientation	Influence on the primary viscosities	
Cooling temperature	To prevent sagging of the materials due to gravity effect	The print up layer is damage and the finishing are not compatible If the materials are not solidified enough and the deposition is continuing, the sagging of materials might be occurred.
CTE (Coefficient of Thermal)	To prevent shrinkage and internal stress	Due to the evolving material stiffness upon solidification bonds. Thermomechanical and crystallisation effect
Temperature of nozzle	Prevent degradation of the filament	
	To achieve optimal mechanical properties	
Size of fibre	Prevent nozzle from clog	
Speed of extrusion nozzle	Printing extruded speed	Speed which the head of print moves around and the plastic is extrude form the nozzle.
	Travelling speed	Speed of printing head is moving when it is not printing which usually higher than printing extruded speed If the speed setting is not been set-up properly, the printer may extrude too much plastic if the speed is too slow or otherwise less plastics will be printed if the speed is too high

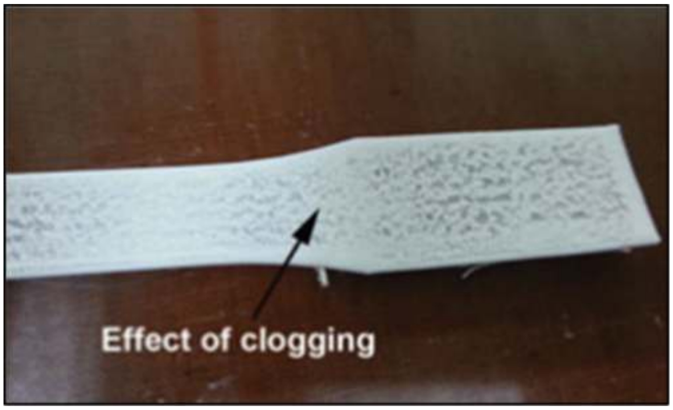

Figure 19. Effect of clogged surface (Rahim, Abdullah, & Md Akil, 2019).

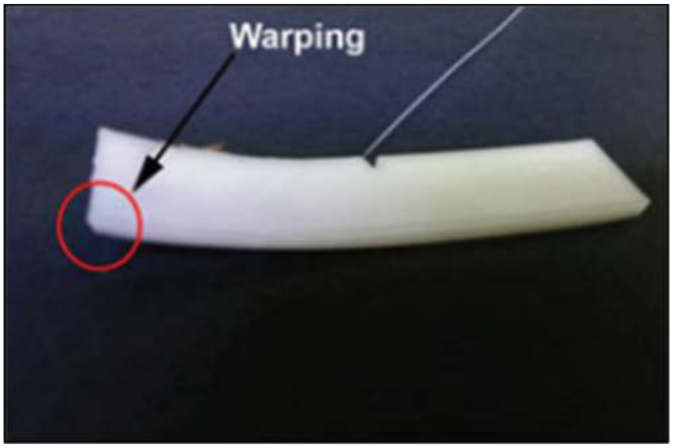

Figure 20. Effects of shrinkage (Rahim et al., 2019).

do not solidify perfectly in the specific amount of time, the printed layers would be damaged and the finishing of the product (i.e. mechanical and thermal properties) would be affected. The temperature of the nozzle is also important to prevent the degradation of the filament before the polymers are printed up on the bed. A degraded filament that extrudes from the nozzle causes the diameter of the print-up layers to be different from the actual dimension; thus, material stability is affected and mechanical properties are insufficient (Brenken et al. 2018).

Noraihan et al. (2019) stated that the optimum filament that is mostly used in the production of 3D printing, which is 1.75 mm 0.01 mm is brittle in nature. An optimum filler has been discovered to overcome the brittleness of the filament structure. The filler weight of nano-particles is 0.2 wt%-8 wt% while for micro-size, the weight is between 10%-40%. The reinforcement material makes it seem that the mechanical strength would be enhanced, however, too much fibre content leads to wetting problems and fibre content that is too low indicates the composites are brittle and the fibres are porous. Therefore, the optimum value should be discovered. The journal also stated that 40 wt% of fibre loading affects the nozzle extrusion process because clogging might occur. So, the right value of fibre loading for microparticle is 30 wt% while the lowest mechanical value is 10 wt%. As for nano-particles, the optimum fibre loading range is between 5 wt%-7.5 wt%. Table 15 is a tabulation of past experiments regarding the different types of fibre and fibre loading amounts. From the fibre analysis and observation, it can be concluded that the shrinkage of the FDM parts can be reduced by adding nanoparticles, which can lead to a low percentage of porosity (Zhou et al. 2020).

In the early process of FDM, many major problems relating to the size of filament, filling of material, nozzle clogging, and printing layers occur. These problems can be overcome by redoing the printing process repeatedly to find the optimum

Table 15. Different fibre-reinforced PLA composites (Rahim, Abdullah, and Md Akil 2019).

Polymer type	Fibre of filler reinforcement	Filler or fibre content (wt%)	Strength (MPa)	Modulus (GPa)
PLA	Tricalcium phosphate (TCP)	2.5	TS: 20–25	TM: 2.7–3.1
PLA	Kraft pine lignin	0, 10, 15, 20	TS: 41.3–55.9	TM: 2.31–2.41
PLA	Cork	0,5,10,15,20,25, and 30	TS: 10–60	TM: 0.05–0.4
PLA	Hemp and harakeke	0,10,20,30	TS: 24–35 (Hemp) TS: 28–37 (Hirakeke)	TM: 3.3–133 (CF) (Hemp) TM: 2.5–4.3 (Hirakeke)

a

Table 16. Thermoplastic and its properties (Wa and Wa 2017).

Property	Poly Lactic Acid	Acrylonitrile butadiene styrene	Nylon	Polycarbonate	Thermoplastic polyurethane	Polyethylene terephthalate
Extrude temp. (c)	180–220	220–240	235–270	270–315	230–260	2230–255
Bed temp. (c)	20–55	80–110	60–80	90–120	40–60	55–70
CTE (µm/m. c)	85	68–110	80–95	50–70	89–170	60–92
T_g (c)	60–65	105–110	47–60	145–150	−35	70–78

parameters for the specific type of plastic (Nikzad, Masood, and Sbarski 2011).

Jagenteufel (Jagenteufel & Hofstaetter, 2017) verified that speed of printing during the FDM process is crucial. If the speed of printing is low, a longer time in the nozzle results in high elongation due to gravitational force.

As a conclusion, different types of thermoplastics that are used will change the parameters of the FDM process. The differences have been analysed by Bates-Green, K. and Howie, T. (Wa and Wa 2017) in Table 14. Also Table 16 illustrates the parameters of the FDM printing process with different values, which depend on the filament's dimension and also the type of reinforcement and polymer.

Nevertheless, as the advantage has been stated in 3D printing industries, it cannot abide disadvantages that might raise during the process. From the author point of view, the most fragile part in producing the reinforcement filament is inconsistency of the diameter. From the reviewed, inconsistency of filament will affect the process of printing, for example, the motor cannot grip the filament perfectly if the size is small because it can slip and might slip, if the size of filament is exceeding than 1.75 mm, it will rather stuck at the motor. Other than that, if the filler contain is not being controlled, it might block in the nozzle or tube, which can affect the printing process. As 3D printing process for polymer composites is too fragile, so the parameter before the process need to be studied before endure the process. Other than that, the selection of reinforcement agent and polymer matrix should be compatible to produce high-strength of filament and products. Table 17, Tables 18 and Table 19 shows the properties of printed polymer and continuous filament.

Table 17. Printing parameters with different types of polymer and filler.

Parameter	Settings (Lee and Liu 2019)	Settings (Daver et al. 2018)	Settings (Le Duigou et al. 2016)	Settings (Torrado Perez, Roberson, and Wicker 2014)
Filament diameter (mm)	1.75	1.75	3.00	1.77
Melting point (c)	155–170	-	-	-
Glass transition temperature (c)	55–70	-	-	-
Print head temperature (c)	210	230	-	230
Build platform temperature (c)	45	60	70	
Print head speed (printing/ travelling) (mm/s)	30/50	30	18	55
Cooling airflow velocity (m/s)	0,1,2,3,4,5	-	-	-
Material	PLA	PLA/Cork	PLA/ Wood	ABS/Jute fibre

Table 18. Properties of printed polymer matrix.

Matrix	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural modulus (GPa)	Printing temperature (°C)
ABS	22–37	0.998	1.9	210–250
PLA	37–46	2.02	2.39	190–210
PP	20–40	1.1–1.6	1.2–1.6	230–260

Table 19. Properties of printed continuous filament.

Continuous filament	Tensile modulus (GPa)	Flexural modulus (GPa)
Carbon	54	51
Kevlar	27	26
Fiberglass	21	22
Jute	39.4	-

My point of view for Noraihan's paper (Rahim, Abdullah, and Md Akil 2019), most printed FDM products are still being used as conceptual prototypes rather than functional components due to the limited mechanical properties of 3D-printed parts. These shortcomings and concerns have driven material scientists to improve the performance of materials to ensure that their structural functionalities comply with specific functional requirements. One of the potential solutions is by developing polymer composites for FDM feedstock. With the inclusion of selected reinforcements in the polymer matrix, a system with a unique combination of properties, which could not have been attained by the constituents alone, can be achieved.

4. Thermoplastic starch (TPS) as feedstock in Fused Deposition Modelling (FDM)

3D printing is also known as additive manufacturing (AM) which involves the fabrication process of adding and joining components layer-by-layer to form a part. It also involves processes such as extrusion and binding (De Leon et al. 2016). One of the major advantages of AM is, it can produce complex shapes accurately with short lead times and small lot sizes (Stansbury and Idacavage 2016). The importance of 3D printing is, it can change the way a product is created, from the traditional manufacturing process. Besides, 3D printing contributes to the prototype industry by easily producing simplified prototypes and also minimising the cost of production and improving overall efficiency.

Several AM techniques have been discovered over the years, such as fused deposition modelling (FDM), stereolithography (SLA), and digital light processing (DLP). However, there are disadvantages of 3D printing applications, which include limitation of size and raw materials, with the most common

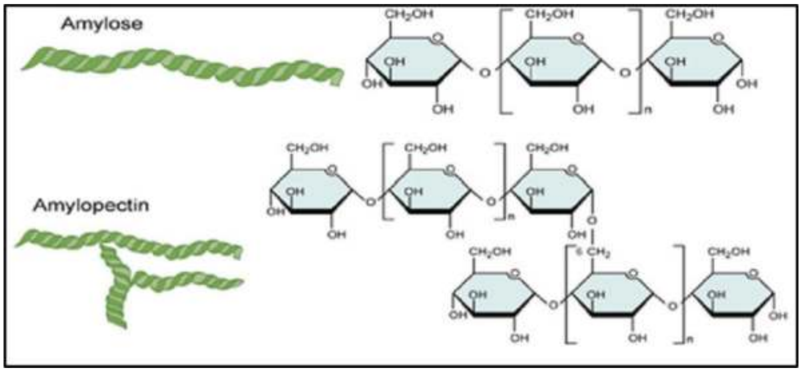

Figure 21. Molecular structure of starch (Amagliani et al., 2016).

thermoplastics used are acrylonitrile-butadiene-styrene (ABS) and polylactic acid (PLA) (Huang 2018). Hence, the evolution of 3D printing technology will develop new raw materials that are compatible with the machine.

The invention of 3D printing with thermoplastic materials replaces the traditional manufacturing, by enabling on-demand production at the final assembly site (Millholand 2016). Apart from that, thermoplastic is the most suitable material for 3D printing technologies such as FDM, where the technology mechanism is simple and highly adaptable.

Despite the increase in demand for 3D printing, the industry encountered a limitation of the thermoplastic material used as feedstock for 3D printer. According to Chia and Wu (2015), the thermoplastic material for 3D printing feedstock should exhibit good melt viscosity, which has a high enough viscosity to be printed but low enough viscosity for extrusion, and this is a challenge in FDM technology. Furthermore, the thermoplastic material should have good processability and is 3D printer-friendly which includes shear thinning behaviour and high zero shear viscosity for allowing easy 3D plotting of constructs (Kyle et al. 2017).

The most common thermoplastic materials as feedstock in 3D printing applications are acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA). Thermoplastic starch (TPS) is one of the alternative materials for the 3D printing feedstock, which is made from unmodified starch in the presence of a plasticiser (water, glycerol, etc.) under thermal and mechanical processing. A plasticiser improves the processability of the starch by lowering the viscosity of the starch melt. Starch itself is a natural substance that consists of many hydroxyl groups that can easily interact with the plasticiser due to its hydrophilic nature. The semi-crystalline structure in starch granules is restructured in the presence of the plasticiser in the starch-glycerol mixture. As a result, a more amorphous structure is formed in the starch melt, in the form of a viscous melt. The changes in the crystallinity of the starch may cause the TPS to retrograde and thicken after heat gelatinisation. The TPS could also have low crystallinity, and poor thermal and mechanical properties, which make it unsuitable for certain industries (Ghanbari et al., 2018). Various studies have been conducted to overcome these disadvantages, especially via chemical modifications, such as acid modification, esterification, and etherification (Huang 2018).

4.1 Starch

Starch is a versatile biopolymer that can be extracted from agriculture plants, such as rice, wheat, and corn. Starch has been used in the food industry and also as substitute component of crude oil and petroleum by-products in packaging, cleaning agents, etc. Starch is low cost and renewable and biodegradable as compared to petroleum-based substances.

Starch is an important biopolymer that could be transformed into a thermoplastic material through a continuous polymer-entangled phase with the presence of plasticiser (Luchese et al. 2015). Starch is biodegradable in a wide variety of environments. It can break down into glucose by micro-organisms or enzymes and then metabolised into carbon dioxide and water. There are many types of starch sources, such as sago, wheat, corn, sorghum, pea, tapioca, and yam. The article from the Sun (2013) reported that wheat, corn, potato, and sorghum are the major sources, with 70%–80% starch content. This number is predicted to increase along with the increase of research interest in the field of food and starch-based plastic products.

Starches from different sources have different overall structures (through chain length distribution in crystalline structure and amylopectin), distribution size of granule, shape, and amylose and lipid contents (Ahmad et al. 1999). Theoretically, starch is a homo-polysaccharide produced by a combination of units of glucose and stored in the carbohydrates in plants. There are two types of homo-polysaccharide, namely amylose and amylopectin.

According to Osorio-Díaz et al. (2002), amylose, also referred to as a linear polysaccharide, is formed by 5–600 glucose units that are linked by α-(1,4) glycoside bonds. The linear structure of amylose contributes to the strength and flexible structure of starch granules. The structure of helix with six glucose units takes place in the core of granules and is soluble in water, as shown in Figure 21. Meanwhile, amylopectin is a branched molecule α-(1,6) produced by thousands of glucose units which prevent the formation of a helix structure. Therefore, a semi-crystalline structure is the arrangement of amylose and amylopectin forming a matrix of starch granules with alternating crystalline and amorphous, known as the growth ring in superior plant starch (Jenkins, Cameron, and Donald 1993).

4.2 Modification of starch

Most natural polymers are brittle, water sensitive, and have poor processability causing their properties to deteriorate. Plasticisers such as water and glycerol have been to enhance their properties and performance. TPS is mainly an extruded starch with high viscous plasticisers, such as glycerol, that acts as a transportation for starch, and provides the movement of starch molecule chains. According to Hernández-Jaimes et al. (2014), TPS shows a shear thinning behaviour where the viscosity decreases as the shear rate increases. Water and glycerol are suitable as a natural polymer because they make starches flow easily and reduce T_g and T_m. During the preparation, the mechanical and heat energies are supplied to destroy the starch's internal hydroxyl bonds. Hence, water is not preferred to be the only plasticiser used due to its volatility; instead, glycerol is more preferred (Dufresne and Vignon 1998). The addition of plasticisers can also influence the ageing of starch-based polymers induced by retrogradation. Retrogradation results in a reduction in the final product embrittlement.

Many applications utilise TPS, such as packaging, textile, and paper industries. However, there are limitations in the physical and chemical properties of TPS that prevent its wider production compared to common thermoplastic materials. Some of the limitations are poor mechanical properties, low crystallinity, and increased brittleness over time because of retrogradation and low water stability that are not suitable for certain applications (Ribba et al. 2017). The limitation of TPS is the starch is hydrophilic and can easily interact with a plasticiser. Thus, various chemical and physical modifications have been suggested by many researchers, such as acid hydrolysis, esterification, and cross-linking, to improve the properties of TPS as the desired application. Chemical modifications such as acid hydrolysis governs the significant changes in starch behaviour, retrogradation, gelatinisation properties, thermal properties, rheological properties, and paste properties (López, Zaritzky, and García 2010). The acid hydrolysis method alters the structure of starch in the presence of an acid solution to reduce viscosity, while esterification method reduces retrogradation tendencies by a denaturation process. Crosslinking is a method that crosslinks the molecules between starch and crosslinking agents to increase the stability as well as mechanical strength of granules. These modifications are crucial to improve the processability of starch to be used as feedstock in a 3D printer, as the unmodified TPS molecules present a few limitations, which are not suitable with the application of 3D printing.

4.3 Application of thermoplastic starch

Since the 1990s, thermoplastic starch has become a new class of biodegradable material (de Carvalho and Trovatti 2016). A lot of effort has been made by many researchers to produce biodegradable materials that can be applied to meet both commercial and environmental demands (Akhtar et al., 2016). Due to its biodegradablility and low cost, TPS is most suitable for packaging of dry products, textiles, and films. For instance, food packaging needs to be cost-effective and follows the requirements of the industry and consumer demand; it must keep food safe and be able to control the impacts of the environment (Marsh and Bugusu 2007). TPS consumption contributes to the decreasing environmental pollution owing to the biodegradable characteristic of TPS. TPS also works as a non-supported film in food and agriculture applications because of its availability and low cost (Glenn et al. 2014). Apart from that, TPS can also be in the form of foams that are used for damping impact as to protect fragile products (Zhang, Rempel, and McLaren 2013). Hence, the development of thermoplastic starch processing technology required to play an important role to achieve the production of inexpensive plastic product and biodegradable.

4.4 Starch as material 3D printing

3D printed bioproducts are expected to grow in the global market by 2022, which involve bio-based products and materials (Yang et al. 2018). An article from the Star (Michael and Lai 2019) had been reported that the government of Malaysia came out with an idea to encourage the people to use eco-friendly products where single-use plastics are substituted as an effort to drive a more sustainable environment. Bio-based polymers are extracted from living organisms such as trees. As the bioplastic industry grows, a biodegradable material that can be degraded naturally by microorganisms is the effective way to overcome the environmental issues, such as reducing waste.

Considering the significance of biopolymer material, 3D printing offers a new perspective in manufacturing biopolymer products. For conventional manufacturing, new biopolymer products are currently being developed that can provide interesting opportunities to expand the biopolymer utilisation in 3D printing applications (Horvath 2017). A study by Sarah (2018) proved that the biopolymer is suited for 3D printing where the 3D printing produces functioning biological tissues from layers of cell-laden hydrogel structures. Based on the statistics from the Association of Plastics Manufacturers, the capacity of bio-based plastics in global production was around 1.4 million tonnes in 2012. Thus, the bio-based plastic is becoming more important than conventional plastics. The bio-based plastic is based on starch due to the low cost, steady availability of crops, and biodegradability. Starch-based plastics derived from plant is much easier to work with during processing (Bioplastics 2016).

Starch also has the potential to be the standard feedstock for 3D printing in the form of pellets or filament. This is due to the lower heating requirements to melt the material that will reduce energy consumption (Catriona & Jonathan, 2014). In addition, starch also has excellent gas barrier properties, which cause the TPS to be less toxic and odourless, and have better print quality due to a low glass transition of temperature. Furthermore, it also can be extruded into a continuous filament similar to the current feedstock for 3D printer. Therefore, there is a great potential for 3D to produce starch-based products on demand and optimise the use of starch. The combination of TPS with other polymers has also been the current interest among researchers. Kuo et al. (2016) have reported to successfully combine TPS with appropriate amounts of acrylonitrile-butadiene-styrene copolymers (ABS), compatibilisers, impact modifiers, and pigments in a single-screw extruder to produce TPS/ABS 3D filaments. According to their report, the TPS/ABS filaments exhibit properties superior to the commercial ABS filaments used as feedstock in the 3D printer.

5. Conclusions

This review is focused on biodegradable composites as a filler of fused deposition modelling (FDM) as one step to reduce pollution issues. FDM is one of the addictive technologies that have many advantages, such as rapid production, good finishing, ability to generate many shapes with complicated geometries and dimensions, and low cost. A natural fibre has biodegradable properties that can preserve the green house. The researchers stated that given their properties, natural fibres, with the correct method of modification and handling, can directly replace synthetic fibres. The main reason for the replacement is synthetic fibre is harmful to the earth and also not environmentally friendly. Users' demand drives manufacturers to produce a recyclable product by using renewable products other than petroleum. Supply of petroleum is decreasing, and the use of natural fibres combined with the biodegradable plastic in FDM technology can preserve this source.

PLA is one of the most highly demanded thermoplastic polymer. This is because of the enforcement to develop full biodegradable composites by combining natural fibres and bio-polymer composites. Many researchers investigate the properties between natural fibres and PLA polymer under different methods of processing. The investigation covered the physical and mechanical properties of the composite itself. Such mechanical properties are the adhesion bonding between natural fibre and polymer to achieve the perfect strength value. The physical properties include ways to overcome the hydrophilic characteristic of natural fibres. The reduction of hydrophilic properties and also chemical content of the natural fibres ensure that the material will not swell in dimension and not affect the user's activities. The pattern of the study should be analysed well to ensure the patterns of the PLA composites increase gradually to replace the synthetic as the main material consumed in production and to achieve fully biodegradable polymer composites.

It is confirmed that bio-polymer composites can successfully replace the usage of the synthetic fibre in industrial production parallel with the mechanical and physical properties of synthetic fibre. Considering the production of natural fibres, the difficulties of material storage are less worrying for natural fibres compared to petroleum. Next, the low cost of the bio-composites compared to other fibres is one of the biggest factors for this interchange between the fibres. As a conclusion to the whole review, the use of bio-polymer composites as the main filler of the FDM instead of synthetic fibre can feed the usage desire as the enhancement of the composites itself meets the users' and manufacturers' expectations.

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