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Review

Tensile Properties of Natural Fibre-Reinforced FDM Filaments: A Short Review

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Abstract: Fused deposition modelling (FDM) is considered the most popular technique of three-dimensional (3D) printing. This is a simple and sustainable method of materials manufacturing with rapidly spreading applications in diverse areas. In this method, a thermoplastic filament is extruded through a nozzle on a layer-by-layer basis to construct a 3D object in a benchtop environment. To further promote its acceptance, FDM printing currently has a significant focus on the use of natural fillers with thermoplastic polymer. Nevertheless, successful FDM printing is largely dependent on the strength and consistency of the feed material, the filament. Preparing such composite filaments is challenging due to possible manufacturing defects and inconsistency while mixing the filler and matrix. Studies showed that there are significant differences between the tensile properties of FDM filament when compared with their printed parts, caused by the variations in printing parameters, filament consumption, density, and architectural difference. Previous reports have confirmed that mechanical characteristics are the most common parameters used by scientists to evaluate the properties of the materials in the additive manufacturing field. Though several reviews are accessible on the tensile properties of FDM-printed materials, currently there is no review available on the tensile properties of the filament itself. This is the first review focused exclusively on the tensile properties of FDM filaments. The goal of this short review is to better understand the influential factors in the natural fibre-reinforced filament preparation process that affect the tensile properties and subsequently impact on 3D printing. Therefore, evaluation of the reported tensile properties, i.e., tensile strength and elongation at the break and modulus, was conducted in relation to different process parameters, such as filler concentration, filler size, extrusion methods, the combination of filler and polymer, and the interrelations among the parameters and properties were explored.

Keywords: additive manufacturing; extrusion; tensile strength; elongation at break; modulus of elasticity

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1. Introduction

Fused deposition modelling (FDM), also known as material extrusion (MEX) or fused filament fabrication (FFF) is a rapidly growing area in additive manufacturing technology. The use of FDM is expanding across diverse domains, including, but not limited to, agriculture [1], construction and building materials [2], composites [3,4], nanotechnology [5], automobile [6], aerospace [7], tissue engineering [8], pharmaceuticals [9], healthcare applications [10], scaffolds [11], and biomedical [12] and microfluidic devices [13], electromagnetic shielding and sensors [14], rapid tooling [15], four-dimensional printing [16], footwear [17], furniture [18], and other home products [19]. This method is rapid, flexible, and solvent-free, making it easy to create complexly designed prototypes and structures in a desktop environment [20]. The FDM approach outperforms even other 3D printing methods like selective laser sintering, with an 82% lower cost and 87% less waste produced [21].

FDM 3D printing requires filament as the input material. The commercial FDM filament market is mainly occupied by two main sorts of filaments, polylactic acid (PLA) [22] and acrylonitrile butadiene styrene (ABS) [23]. However, PLA needs elevated temperature (>50 °C) for degradation [24] and ABS is non-biodegradable [25], both of which hinder the full captivation of the sustainability advantage of FDM. Therefore, reinforcing different natural fillers in the filaments has been a keen area of interest in FDM research. This is because the incorporation of natural fillers commonly improves the biodegradation rate and is known to reduce the production cost, particularly if sourced from a waste stream [20]. However, the reinforcement of natural fillers impacts the tensile strength of the filament, which can have either a positive or negative impact. This often depends on the specific formulation used [26], with simultaneous influences from a few factors, such as the amount of filler [27], size of filler [28], surface modification [29], and the extrusion method [30].

The filaments used for FDM are of a distinctive kind of material. They should not be confused with textile filament that has a very fine diameter, or any other extruded and shaped (e.g., through compression or injection moulding) materials. Though the fundamental of the melt extrusion of textile filament and FDM filament is the same, the difference is mainly in the diameter of the extruded filament, which is much coarser in the case of FDM. Commercial FDM filaments come with standard diameters of 1.75 mm or 2.85 mm [31], though variation of this is also reported when preparing natural fibre-reinforced composite filaments, such as 1.3 to 2.1 mm [32] and 2.8 to 3 mm [33]. The produced filaments are firm but able to be wrapped around a spool, and just enough to be used as a feeding element in FDM printers.

Different parameters can affect the 3D printing performance. These include printing parameters, such as speed, nozzle diameter [34], extruder temperature [35], build plate temperature [36], infill pattern and percentages [37], as well as filament properties, such as its mechanical properties [38], viscosity [39], and consistency [40]. However, it has been confirmed (from a review of 1271 documents) that the analysis of the mechanical property is a significant component studied in additive manufacturing and it has been a subject of the highest interest in this field [41]. The keyword "mechanical properties" was found as the third highest (after "additive manufacturing" and "3D printing"), which shows its utmost importance in this area. In most cases, mechanical properties are analysed through tensile tests. Nevertheless, the tensile properties of the filament itself are often overlooked, and in most cases, the tensile tests are performed on the 3D printed materials, e.g., dog-bone shapes, to confirm their performance. However, filament and 3D printed specimens are two different categories of materials, even though they are produced from the same combination. During 3D printing, a varied range of parameters are used, such as type of infill patterns, percentages of infill, and the variation of design in different layers. All of these influence the density [40], weight, and filament consumption [42]. Therefore, a 3D printed part is perceived as a complex architectural object, compared to a plain composition of filament. Particularly in cases of fibre-reinforced composites, it is crucial to thoroughly understand the properties of filaments and any influential phenomena before printing, as their tensile properties could have been affected through the formation of voids and other faults in the interfaces of polymer and matrix [43].

Despite several comprehensive reviews available on the tensile properties of 3D printed parts from natural fibre-reinforced composites [20,44–47], all of them are dedicated to the tensile properties of the printed objects rather than the FDM filaments themselves. Although filaments are the key to successful 3D printing, currently there is no conclusive knowledge about what parameters affect their tensile properties and how those can impact 3D printing. Therefore, this short review aims to emphasise the tensile properties of reported FDM filaments, particularly those that are reinforced with different natural fibres and identify the key influential factors in their manufacturing process. To perform this review, the database of Web of Science was first filtered by using the keywords "tensile properties", "filaments", and "3D printing" (in the Title, Abstract and Keywords fields). However, many articles in this filtered segment did not report the tensile properties of filaments and demonstrated the tensile properties of printed dog-bone shapes. Therefore, all of those papers were reviewed to identify the papers that presented the tensile properties of filaments. After that, the impact of the process parameters observed on those papers was emphasised in relation to each other and their influence on 3D printing was evaluated.

2. Mechanism of FDM Printing

The FDM 3D printing method was invented by S. Scott Crump in the United States in 1989. The patent of this invention describes it as an apparatus including a movable dispensing head and a base member that is moved related to each other along the X, Y, and Z axes in a programmed pattern [48]. The dispensing head has a supply of material that solidifies at a predetermined temperature (e.g., thermoplastics). Thus, the build-up material discharged from the dispensing head onto the base at a controlled rate produces the 3D objects.

The majority of FDM 3D printers in the market operate using the Cartesian coordinate system [49]. The Cartesian coordinate system is a method of locating a point using the X, Y, and Z axis lengths. The supply of material comes in the form of filament to ensure the feed is continuous. Figure 1 depicts a typical mechanism of FDM 3D printing.

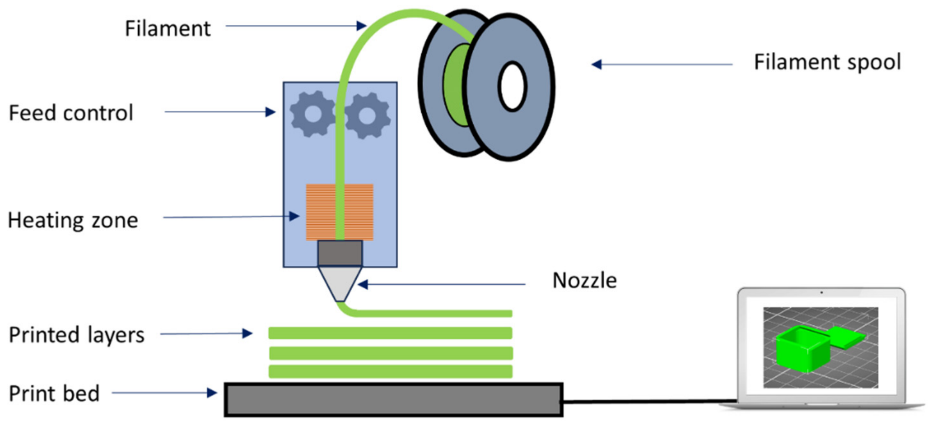

Figure 1. Schematic of 3D printing mechanism through fused deposition modelling.

The filament is guided through the heating zone to successfully melt the polymer and extrude through a nozzle (printing head) [50]. The extruded material is then lined up as per the pre-program onto a printing bed (base) on a layer-by-layer basis [51]. The printing temperature can be higher than the melting temperature of the thermoplastic to maintain a consistent flow of melts [42]. The printing bed temperature is also adjustable to synchronise the whole process of 3D printing.

3. FDM Filaments

Though ABS and PLA are two common polymers used in FDM printing [52], research on fibre-reinforced filaments is concentrated more towards the use of PLA rather than ABS. This is probably because of the so-called biodegradability of PLA, which matches natural fillers. Though the degradability of PLA needs a higher temperature [24], the addition of fillers can help start the degradation of the natural part in a natural environment, and at least reduce the fraction that is needed for degradation in customised conditions. Other than PLA, polycaprolactone (PCL) and polypropylene (PP) are also proposed for FDM where natural fillers were used. PCL is probably the fastest among the synthetic polymers that has a biodegradable profile [53]. Another advantage of PCL is its FDM processability at low temperatures (80–150 °C) [33] which is much lower than ABS and PLA (200–220 °C) [52], and is thus more energy-saving. Despite being a non-degradable polymer, a reason for interest in PP could be its abundance as waste [54] that co-matches with its melt processability through FDM at a comparable temperature (200–250 °C) [55] to PLA and ABS.

This is important to note that the consistency of filament tensile properties plays a crucial role in the printing performance [30]. This is more applicable to composite filaments rather than pure polymers as phase separation can occur in composites due to poor mixing [56] which can alter the filament properties, such as strength, in places. During

FDM printing, the 3D printer uses the diameter value of the filament supplied by the user to adjust the filament feed to match the final specimen [57]. Therefore, it is possible to print unevenly if the filament property is not consistent, such as when voids are present relative to the improper mixing of polymer and filler during filament preparation. Figure 2a,b show a practical example of a difference in composite samples (wool-reinforced PCL) due to large variations in the filament tensile properties in places. Figure 2c shows an example of the variation that occurs in the stress–strain curves of these samples. A filament with inconsistent properties produces a much lower maximum strength, breaking strength, and elongation in places compared to a consistent filament.

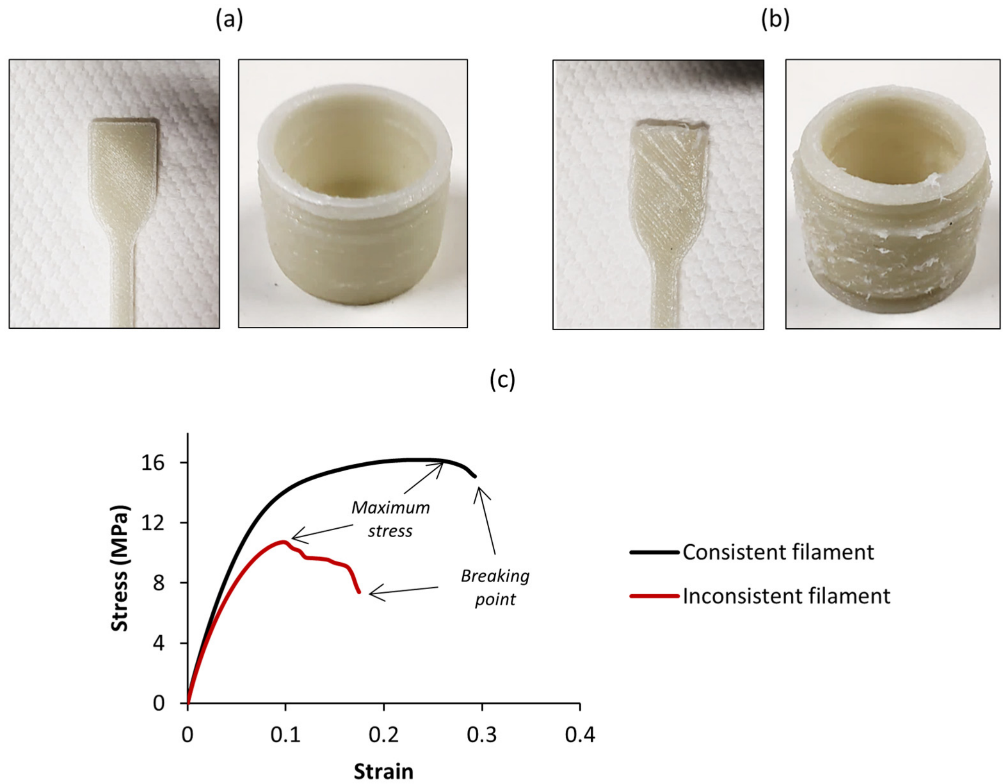

Figure 2. Examples of the impact of consistency of composite filament (wool/polycaprolactone) tensile strength on the 3D printed objects from (**a**) consistent filament and (**b**) inconsistent filament, and (**c**) variation in the stress–strain curves from consistent and inconsistent filaments (this work has been carried out by authors, unpublished results).

4. Tensile Properties of FDM Filaments

4.1. Method of Tensile Test

Interestingly, there is no standard yet established for the tensile test of 3D printable filaments, i.e., those from standard organisations, such as the ISO (International Organization for Standardization) or ASTM (American Society for Testing and Materials). The standards for tensile test for fibre-reinforced plastic materials are proposed mainly based on the dog-bone shapes, which are obtained after the 3D printing. A news release from the ASTM indicates that they are currently working on this, and a standard is on its way (ASTM WK82320) to cover the measurement of tensile properties, precisely for thermoplastic-based filaments used as a feed material for the MEX or FDM technique [58]. This also indicates the expanding awareness of the importance of the tensile properties of filaments, which is key for proper FDM printing.

Though the measuring technique of FDM filament's tensile strength somehow replicates to single fibre tensile test, the calculation is different from the test of single textile fibre (ASTM D3822), where the strength is expressed as force per linear density (i.e., tenacity) [59]. Since the properties of the filaments are more relevant to their plastic counterpart, i.e., the 3D printed object, to provide a more useful comparison, researchers have reported the values in a similar unit (e.g., MPa) to the 3D printed object (force per unit area). This indicates the consideration of the diameter of cylindrical filament for cross-sectional area calculation [33,52]. Even though there is no standard available yet, the studies have measured the tensile properties based on the fundamental principle of the tensile test [60], such as using the force per cross-sectional area for the tensile strength or increase percentage of gauge length to measure the elongation. This has provided the opportunity to compare the reported findings.

4.2. Impact of Extrusion Methods

Until now, three pathways have been used to produce the FDM filaments. As shown in Figure 3a, the first path is through the single-screw extrusion (SSE) [61]. Commonly, they have two to four temperature control zones to operate. Both the polymer pellets and fillers are added to the hopper and extruded directly. This method is quick and easy and often possible with a simple desktop setup but may lead to non-uniform mixing and defects in the composite [62].

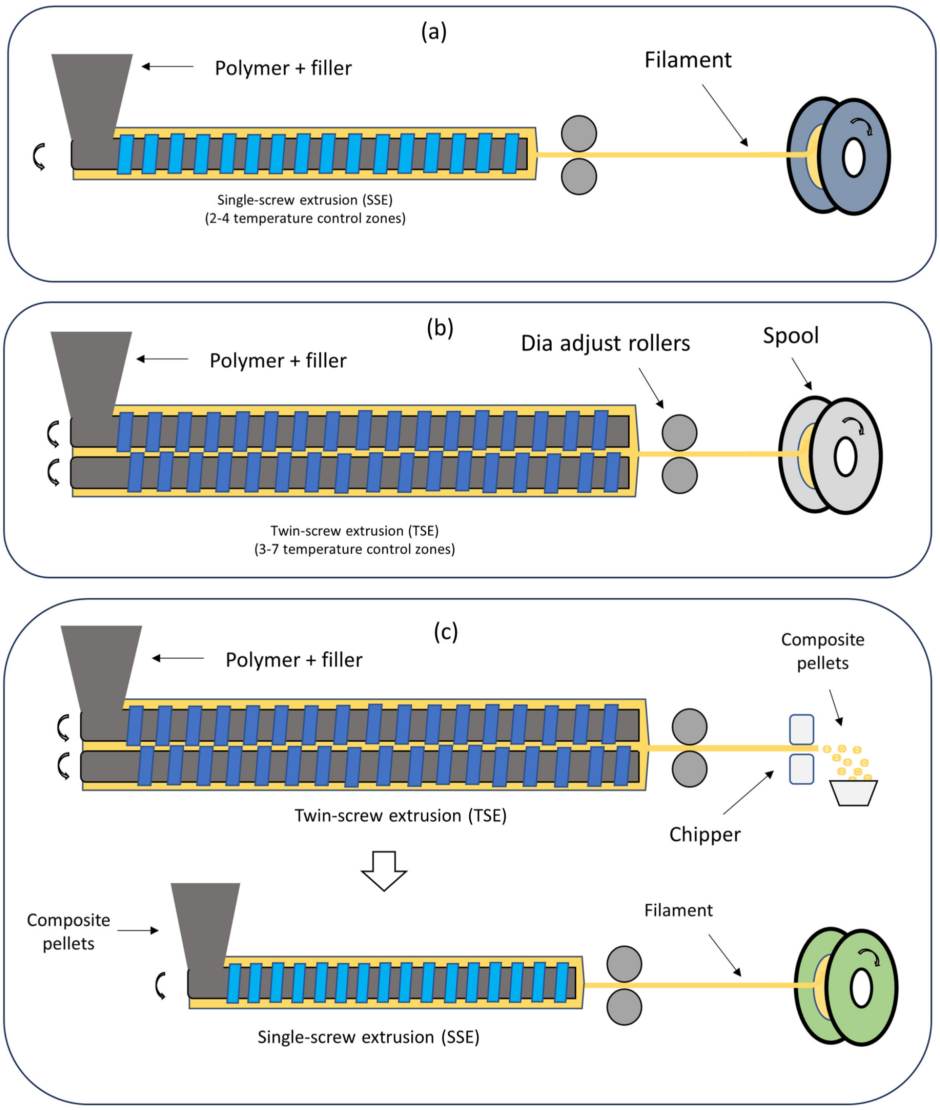

Figure 3. Different methods of extrusion used for filament preparation, reported in the literature, (**a**) single-screw extrusion, (**b**) Twin-screw extrusion, and (**c**) combination of both extrusions.

The second method (Figure 3b) is twin-screw extrusion (TSE) [63], which often needs a larger setup than SSE, due to accommodating two screws for extrusion as well as more temperature-controlling zones (often three to seven). While an additional screw helps with better mixing, more control over the temperature facilitates an improved mixing of the polymer and filler with limited phase separation [64]. The third method is a combination of TSE and SSE (Figure 3c) where the extrusion is performed in two steps. At first composite pellets are made from the TSE technique, which is later transformed into filaments by SSE [33]. This method is slower compared to the other two but provides the opportunity for further improved miscibility by taking advantage of both techniques [52]. Table 1 summarises the tensile properties of FDM filaments achieved from these three techniques when a similar loading (approximately 10%) was used. This is clear that the involvement of TSE, either solely or with the incorporation of SSE, was greatly influential for the improvement of tensile strength when compared to the results from SSE only. In one study only, the tensile strength of the control polymer was reduced after the TSE–SSE technique, where the fillers were used mainly as plasticisers to improve flexibility [33]. Since plasticisers are designed to occupy the intermolecular spaces of a rigid materials' structure and reduce different secondary interactions, including hydrogen bonds and van der Waals force, the required energy for molecular motion decreases, leading to increased flexibility and chain mobility [65]. Therefore, a decrease in the modulus as well as an increase in elongation was also evident from that study. Other than that, from the data available until now, TSE or the combination of TSE–SSE seem to be a great approach for FDM filament preparation.

Table 1. Impact of the extrusion methods on the composite filaments produced for FDM printing (SSE = Single-screw extrusion, TSE = Twin-screw extrusion).

Method	Polymer	Filler	Loading (%)	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)	Reference
SSE	PLA	Spruce pulp	10	36.6 (55.3)	-	-	[29]
	PLA	Egg shell	10	36 (~44)	-	2.7	[66]
	PCL	Cocoa shell	10	24 (27.5)	334 (304)	697 (702)	[67]
TSE	PLA	Lignin	20	50.8 (42.9)	2100 (2260)	4 (2.9)	[32]
	PLA	Flax	15	30 (31.3)	2400 (1300)	0.075 (1.8)	[68]
	PP	hemp	10	~26 (22)	1100 (~890)	11.5 (10.9)	[69]
	PP	hemp	10	28.2 (22.1)	1683 (892)	-	[54]
	PP	Harakeke	10	~25.5 (22)	1100 (~890)	7.9 (10.9)	[69]
	PP	Harakeke	10	27.6 (22.1)	1612 (892)	-	[54]
TSE-SSE	PLA	Beechwood	10	57 (55)	3630 (3270)	-	[40]
	PCL	Wool	10	17–17.1 (15)	239–254 (176.1)	23.1–23.7 (1780)	[52]
	PCL	Gum rosin	10	9.8 (13.7)	145.9 (184.8)	912.3 (558.7)	[33]
	PCL	beeswax	10	11.7 (13.7)	112.1 (184.8)	676.8 (558.7)	[33]

Data in the bracket shows the values of the control polymer.

4.3. Impact of Filler Amount

A common trend in the tensile properties of composites is an initial improvement in tensile strength by a limited amount of filler up to when the distribution of fibres is uniform [70]. However, with a higher amount of filler, it is often not possible to maintain a uniform distribution, which results in poor tensile strength [71]. This is due to the creation of voids in the fibre–matrix interfaces and greater interruption in the molecular chain of the polymer, all of which negatively affect the tensile strength. Therefore, the impact of the filler amount is probably the most affecting parameter in the tensile properties of FDM filaments. Table 2 shows the key findings regarding the tensile strength, Young's modulus, and elongation at break of the filaments impacted by the amount of filler.

The filaments produced using PLA showed a common trend of reduced tensile strength after filler inclusion (10 to 30%), with a few exceptions. When using beechwood as the fibre source, the strength was increased with 10% filler, though it was reduced after the inclusion of 20 or 30% filler [40]. In a separate study, lignin reinforcement in PLA resulted in an 18% improvement in tensile strength when 20% filler was used. However, no other ratio was tested to confirm the best possible lignin amount that could be incorporated [32]. In most cases, only 10% filler was enough to reduce the strength as well as the elongation at the break of the composites. However, regardless of the strength and elongation, Young's modulus was consistently increased by fillers, confirming more rigidity in composites compared to the control polymer [72]. In the case of PCL composites, a similar observation was found, i.e., decreased strength, elongation, and flexibility with only 10% filler [33,67], except the one with the wool/PCL composite, where both 10% and 20% wool fillers were able to improve the tensile strength of the composites [52]. This was claimed to be the effect of the hydrophobic nature of wool's surface, which was more matching with hydrophobic PCL. It is known that hydrophilic fillers (such as cellulosic) often lack compatibility with hydrophobic polymers, which is one of the major dilemmas in composite preparations [73–75].

However, the composite filaments reported using PP as the polymer showed improved strength from two different studies both with 10% and 20% fillers (hemp and harakeke). This was possibly affected by the large length of fillers used (such as 8–10 mm), keeping the fibre property more intact [54,69]. In between these two studies, Stoof and Pickering have used more temperature control zones (five zones) and a slower speed of extrusion (50 rpm) [54]. This has probably allowed more uniform mixing and led to a slightly higher tensile strength in all the combinations.

In a study with ABS by Ahmad et al. [76], oil palm fibre has reportedly influenced a reduction in the strength of ABS when a lower amount was used (3–5%), though slightly improved the strength with a 7% filler. Nevertheless, the nanosized fillers (carbon nanotube) were found highly effective in improving tensile properties, where only 1.5% of filler reportedly improved approximately 7% of strength in ABS [77]. Unfortunately, even though there are many studies conducted using nanofillers for FDM composites, the tensile properties of the 3D printed specimens are mostly reported, not the properties of the filaments [78–82]. Overall, this is obvious that in addition to the filler amount, filler size could have importantly impacted the properties of FDM filaments, which is discussed in the next section.

Table 2. Impact of filler amount on different composite filaments for FDM printing.

Polymer	Amount (%)	Filler	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)	Reference
PLA	1	Cellulose nanofibrils	~49 (~47)	-	~4.5 (~3)	[82]
	2.5	Cellulose nanofibrils	~57.5 (~47)	-	~4.5 (~3)	[82]
	4	Eggshell	49.3 (~44)	1890	6.6	[66]
	5	Cellulose nanofibrils	~50 (~47)	-	~3.9 (~3)	[83]
	6	Eggshell	~40 (~44)	-	~5 (~4)	[66]
	8	Eggshell	~39 (~44)	-	~5.5 (~4)	[66]
	10	Spruce pulp	36.6 (55.3)	-	-	[29]
	10	Beechwood	57 (55)	3630 (3270)	-	[40]
	10	Eggshell	~36 (~44)	-	2.7 (~4)	[66]
	12	Eggshell	~31 (~44)	2390	1.7 (~4)	[66]
	15	Flax	30 (31.3)	2300–2400 (1300)	0.07–0.1 (1.8)	[68]
	15	bamboo	22–23 (31.3)	1600–1800 (1300)	12–13 (1.8)	[68]
	20	Spruce pulp	32.4 (55.3)	-	-	[29]
	20	Beechwood	49 (55)	3940 (3270)	-	[40]
	20	Lignin	50.8 (42.9)	2100 (2260)	4 (2.9)	[32]
	30	Beechwood	48 (55)	3800 (3270)	-	[40]
	30	Recycled wood	27.3 (65.4)	-	-	[38]
	40	Beechwood	42 (55)	3860 (3270)	-	[40]

Table 2. *Cont.*

Polymer	Amount (%)	Filler	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)	Reference
PCL	40	Recycled wood	24 (65.4)	-	-	[38]
	50	Beechwood	30 (55)	3000 (3270)	-	[40]
	10	Wool	17–17.1 (15)	239–254 (176.1)	23.1–23.7 (1780)	[52]
	10	Cocoa shell	24 (27.5)	334 (304)	697 (702)	[67]
	10	Gum rosin	9.8 (13.7)	145.9 (184.8)	912.3 (558.7)	[33]
	10	beeswax	11.7 (13.7)	112.1 (184.8)	676.8 (558.7)	[33]
	20	Wool	16.1–16.8 (15)	249–275 (176.1)	11.7–19.3 (1779.5)	[52]
	20	Cocoa shell	17.5 (27.5)	329 (304)	538 (702)	[67]
	20	Chitosan	~4	-	-	[84]
	30	Cocoa shell	12.5 (27.5)	356 (304)	495 (702)	[67]
	40	Cocoa shell	9.4 (27.5)	338 (304)	129 (702)	[67]
	50	Cocoa shell	7.3 (27.5)	319 (304)	23 (702)	[67]
PP	10	Hemp	~26 (22)	~1100 (~890)	11.5 (10.9)	[69]
	10	Hemp	28.17 (22.12)	1683 (892)	-	[54]
	10	Harakeke	~25.5 (22)	~1100 (~890)	7.9 (10.9)	[69]
	10	Harakeke	27.56 (22.12)	1612 (892)	-	[54]
	20	Hemp	~31 (22)	~1800 (~890)	5 (10.9)	[69]
	20	Hemp	34.35 (22.12)	2261 (892)	-	[54]
	20	Harakeke	~33 (22)	~1700 (~890)	5.1 (10.9)	[69]
	20	Harakeke	35.94 (22.12)	2336 (892)	-	[54]
	30	Hemp	37.8 (22.12)	2681 (892)	-	[54]
	30	Hemp	34 (22)	2163 (~890)	-	[69]
	30	Harakeke	38.5 (22.12)	2767 (892)	-	[54]
	30	Harakeke	34 (22)	2202 (~890)	-	[69]
ABS	1.5	Carbon nanotube	41.2 (38.4)	1680 (2050)	4.05 (4.99)	[77]
	3	Oil palm	0.15 (0.4)	16.2 (14.1)	~1.9–3.9 (~4.9–5.4)	[76]
	5	Oil palm	~0.25 (0.4)	17.1	~1.9–2.8	[76]
	7	Oil palm	0.46 (0.4)	18.3	~2.4–3	[76]

Data in the bracket show the values of the control polymer.

4.4. Impact of Filler Size

Table 3 lists the tensile properties of the FDM filaments noting the size of particles, where the filler amount was relatively similar, approximately 10–15%. In the case of PLA composites, the strength of PLA was less affected when the filler's particle size was higher. For example, flax fibres 5 mm in size reduced the tensile strength by approximately 4% [68], though a 315 µm bamboo fibre reduced the strength by 26.5% compared to the control. The only exception was the beechwood fibre, in which the strength increased with a lower size of particles (<237 µm) [40]. Interestingly, a longer fibre also affected the modulus in the case of PLA, and a higher degree of increase in the modulus was seen by the longer fibre.

In the case of PCL and PP composites, the changes in tensile strength showed an opposite trend (the lower the particle size, the higher the strength); however, an increase in the modulus and a decrease in elongation were observed. It is true that a smaller size of particles helps produce a uniform distribution and thereby can positively influence the strength. Asmaller size of particles produces a lower disturbance angle in the matrix that is less prone to breakage compared to a higher disturbance angle produced from a larger size of particles [85]. This also helps the effective stress transfer when a tensile load is applied. But, it should also be noted that the longer size of fibres can also improve the structure due to a higher aspect ratio [71]. Therefore, it is highly important how the fibres are distributed in the matrix.

Figure 4a shows the impact of filler loading on the increase or decrease of tensile strength compared to the control of different polymers, and Figure 4b shows the impact of particle size on the change in tensile strength when the loading was constant (10%). It can be observed after 30% loading that the strength mostly reduced. However, when a lower filler loading was used, both increases and decreases were seen. In case of particle size as well, both increase and decrease were seen with similar size of particles, indicating the influence of multiple factors. Overall, the trend of tensile properties can be shortened as a synergistic influence from different variables, such as the loading amount, aspect ratio, particle size, and distribution of particles. Figure 4c thus summarises these common criteria observed in the tensile strength of FDM filaments that affected the tensile strength.

Table 3. Impact of the particle size of fillers on different composite filaments for FDM printing.

Polymer	Filler Size (µm)	Filler	Amount	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)	Reference
PLA	<25	Eggshell	10	36 (~44)	-	2.7	[66]
	<237	Beechwood	10	57 (55)	3630 (3270)	-	[40]
	315	Bamboo	15	23 (31.3)	1800 (1300)	0.13 (1.8)	[68]
	595	Spruce pulp	10	36.6 (55.3)	-	-	[29]
	630	Bamboo	15	22 (31.3)	1600 (1300)	0.12 (1.8)	[68]
	2000	Flax	15	30 (31.3)	2400 (1300)	0.075 (1.8)	[68]
	5000	Flax	15	30 (31.3)	2300 (1300)	0.1 (1.8)	[68]
PCL	22.2	Wool	10	17.1 (15)	239 (176.1)	23.7 (1780)	[52]
	46.3	Wool	10	17 (15)	254 (176.1)	23.1 (1780)	[52]
	50	Cocoa shell	10	24 (27.5)	334 (304)	697 (702)	[67]
PP	8000	Hemp	10	28.2 (22.1)	1683 (892)	-	[54]
	8000	Harakeke	10	27.6 (22.1)	1612 (892)	-	[54]
	10,000	Hemp	10	~26 (22)	1100 (~890)	11.5 (10.9)	[69]
	10,000	Harakeke	10	~25.5 (22)	1100 (~890)	7.9 (10.9)	[69]

Data in the bracket shows the values of the control polymer.

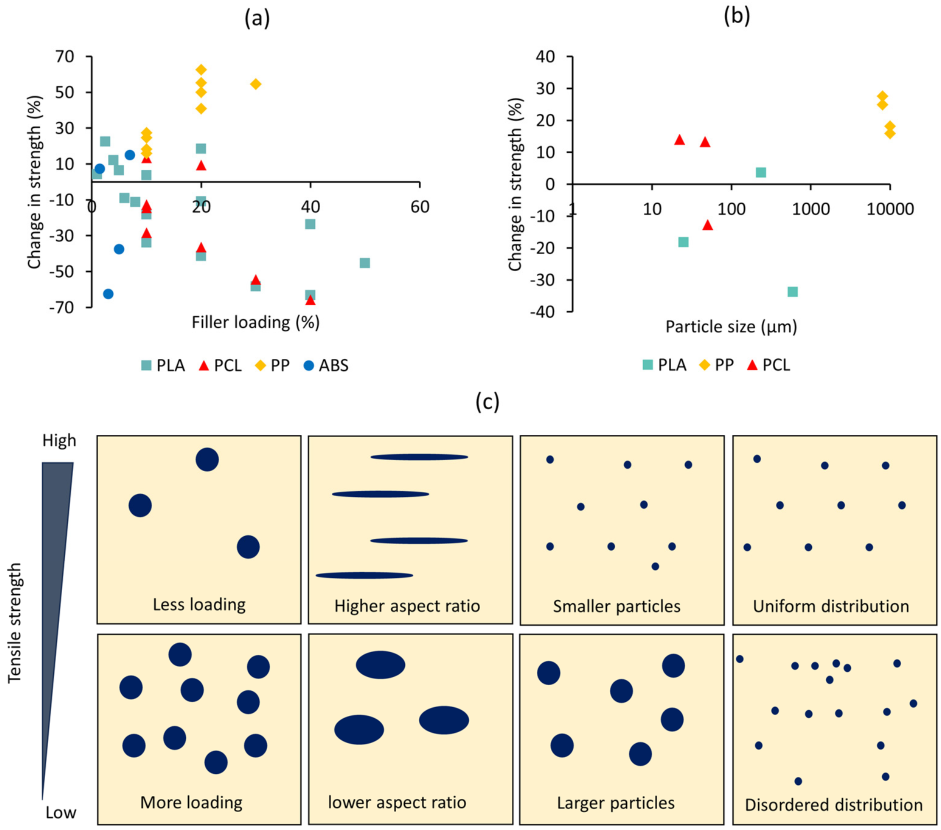

Figure 4. Impact of (**a**) filler loading and (**b**) particle size (10% loading) on the change in tensile strength of natural fibre-reinforced FDM filaments, and (**c**) summarised trend of the tensile strength affected by different parameters of the fillers sourced from natural fibres.

4.5. Impacts from Other Parameters

Some other parameters, such as chemical modification and the fineness of filler fibres and plasticisers, can have potential impacts on the FDM filament's tensile strength, but these areas are not widely explored. For example, in many cases, the chemical modification of fillers has been considered, though the tensile properties of the printed objects were directly reported, not the filament [86–88]. For that reason, despite there being diverse options in the route of chemical modification, data are very limited for filaments.

In only one study, Filgueira et al. reported the laccase-assisted modification of thermomechanical pulp fibres by octyl gallate and lauryl gallate to improve the adhesion behaviour with PLA [29]. Though both chemicals induced hydrophobicity, the surfaces of the fibres were found to be chemically different due to the variation in the aliphatic chain lengths of the two chemicals. The treatment with octyl gallate resulted in better hydrophobicity and better interfacial compatibility with the PLA matrix. This led to a better stress transfer from PLA to fibre during the tensile test and a higher strength was found in octyl gallate-treated filament. This result aligns with the claim for hydrophobic wool fibre's better compatibility with hydrophobic polymer (PCL) and minimal loss of strength in composite filament preparation [52].

Another parameter of filler that influences the filament strength could be the fineness of the natural fibres. A study on merino wool showed a finer diameter of fibre (16 µm) was slightly more influential than a coarser diameter of fibre (24 µm) to enhance the tensile strength [52]. Though the difference was not so prominent with a smaller amount of filler (10%) it became more evident when higher amounts of filler were used, i.e., 20–25%. However, there was no further study on any other natural fibres to confirm this influence.

The presence of a third component in the system, such as a plasticiser, could also be an influential factor in filament strength. In one study, a comparison of the performance of two plasticisers was reported [32]. Interestingly, uses of plasticisers in a small amount have shown a significant increase in the tensile strength and elongation behaviour and reduced the modulus. For example, the use of 2% polyethylene glycol (PEG) resulted in an 18% increase in tensile strength, a 7% decrease in the modulus, and a 35% increase in elongation at break. A similar effect was also seen when a commercial plasticiser (struktol TR451) was used. This kind of effect from plasticiser is uncommon since the use of plasticiser commonly impacts the strength negatively [89]. However, since no other FDM studies have reported this effect on the filament, further information is required before coming to any conclusion.

4.6. Impact of Tensile Properties on Printing Operation

Table 4 compares how different the tensile strength of filament compares to the 3D printed objects made from it. In the studies where tensile tests for both filaments and 3D printed specimens were performed, a significantly higher tensile strength is often seen in the case of the filament. This is because of the use of varied parameters during 3D printing, such as infill patterns, percentages of infill, and variations of designs in different layers. In the reports, often all the printing parameters were not reported which hinders a proper comparison. Ideally, a 100% infill should replicate the filament properties, other than smaller percentages of infill where gaps persist. However, even with a 100% infill, a lower strength was seen in the 3D-printed object compared to the respective filament [33]. Only in a series of studies, where filament properties were evaluated first [52], followed by the properties of printed objects with different printing parameters [42], a similar strength was seen in wool-reinforced PCL. This was attributed to the hydrophobic nature of the wool surface, which was claimed to be more compatible with PCL, thus promoting a uniform dispersion of filler in the matrix. This was noted that during the 3D printing, due to the use of a higher temperature (130 °C) than that of filament preparation (90 °C), the flowability of the polymer–matrix system was improved [42]. This could be a reason for better adhesion in the printed objects as well. However, hydrophobic fillers, such as protein fibres are not yet widely used in FDM printing; even while occasionally used, data for both filament and printed specimens are often not available. This area thus seeks further exploration.

Table 4. A side-by-side tensile strength results from some composite filaments and the 3D dog-bone shapes printed from them.

Polymer	Filler	Tensile Strength (MPa)		Printing Infill (%)	Reference
		Filament	Printed Object		
PLA	-	55.3	~7.5	-	[29]
PLA	Octyl gallate modified Spruce (10%)	57.7	~10.5	-	[29]
PP	-	22	~17	-	[69]
PP	Hemp (20%)	~31	14	-	[69]
PLA	-	65.4	26.8	55	[38]
PLA	Recycled wood (30%)	27.3	7.3	55	[38]
PCL	-	13.7	11.5	100	[33]
PCL	Gum rosin (10%)	9.8	7.9	100	[33]
PCL		15	13.9–14.7	50	[42,52]
PCL	Wool (10%)	17–17.1	16.8–17.5	50	[42,52]
PP	-	22.1	17	-	[54]
PP	Hemp (10%)	28.2	20.6	-	[54]
PP	Harakeke (10%)	27.6	19.9	-	[54]

The lowest permissible tensile strength of FDM filament that can be printed varies depending on the polymer used. Until, now the lowest value (0.15–0.46 MPa) is reported for ABS composites with 3–7% oil palm filler, with a particle size of 1 to 4 mm [76]. However, in terms of tensile strength, this value may not be appropriate since the tensile strength of the control ABS (0.4 MPa) was reported significantly different from that conveyed in different FDM 3D printing studies, such as 38.4 MPa [77], 36.8 MPa [88], and 24.7–33.7 MPa [90]. Nevertheless, it was testified that all of the ABS–palm oil filaments were successfully 3D printed into objects. It was also noted that with the higher amount of loading (e.g., 7%), the printed surfaces were coarser, and frequent clogging was reported in the printer nozzle. An important phenomenon in this aspect is the decrease in the density of filament with the increase of filler percentages (such as a 24% decrease by 7% filler) [76], due to the formation of voids in the system. This can also be confirmed by an increased porosity by increased filler loading, e.g., porosity raised from 8.5 to 17.6% when the filler amount was increased from 3 to 7%). The generation of such void spaces in filaments is likely to affect the 3D printing performance, though there is not enough study on this aspect.

A similar observation was reported in PLA composite filaments with a higher loading of wood particles (50%) where the strength of the filament was reduced to half compared to control [40]. In the printed parts, clusters of particles were seen, with the presence of voids on the specimens and dark spots on the edges, and the filament failed to be deposited consistently due to clogging and improper flow through the printer nozzle. The particle size of wood was reported less than 237 µm, which appeared suitable for printing through a 0.4 mm nozzle when the filler loading was less, i.e., 10–20%. Confirming the effectiveness of this range for PLA, another study reported successful printing with 15% loading of filler but using two different size ranges, i.e., 315–630 µm and 2 mm, which indicates even though the particle size is coarser, it can be compromised with the filler loading to attain the printing. In the case of PCL, at best 50% filler (particle size 50 µm) was used to produce filaments, resulting in a massive decrease (73%) in the tensile strength compared to the control [67]. Particularly, when more than 30% fillers were used, the strength radically decreased, and thereby filament containing only up to 30% filler was suitable for 3D printing with PCL. This was also found similar to PP, where 30% of fillers were successfully used in filaments and printed without any reported problems. However, it is worth noting, that a highly coarse size of filler particles (8–10 mm) was used for PP in two separate studies, therefore the nozzle size used was also large (1 mm) [54,69]. During the printing, the speed was maintained low, and in the software, the diameter of filament was inserted by 80% reducing the actual to further slow down the printing process [69]. The reason for the success in printing with PCL and PP using a higher amount of filler (up to 30%) than PLA (10–20%) was probably related to the more flexible nature of PCL and PP chains compared to PLA [91–93]. This provides the added advantage for the fillers to accommodate the spaces available in PCL and PP chains.

5. Conclusions and Future Scope

This study exclusively reviews the tensile properties of FDM filaments reinforced with natural fibres. The available data suggested that the tensile properties of FDM filaments are largely different to the tensile properties of the 3D printed parts due to different printing parameters set during printing and the structural differences between these two kind of materials. Further, a higher temperature is often needed for the fabrication of 3D printing objects, compared to filament which was observed to affect the rheological behaviour of the polymer–matrix system. Therefore, the tensile properties of filament should be treated as a separate phenomenon. The accumulated results indicated that twin-screw extrusion or its combination with single-screw extrusion can deliver filaments with better tensile properties, compared to only single-screw extrusion. The amount and size of the fillers also influenced the tensile strength, though this was more related to whether a uniform distribution of fillers was achieved to help effective stress transfer.

Data on the influence of different chemical modifications of the fillers on the filament strength is limited, which could be a future area to explore. Hydrophobic fillers (protein fibres) were found to be compatible with the hydrophobic thermoplastic interface, resulting in a better or marginal reduction in strength. However, more study is required to validate this interesting finding. The results suggest that the filler amount has the most influence on the filament's tensile properties and subsequent 3D printing performance. For PLA, 10–20%, and in cases of PCL and PP, up to 30% filler was found to be suitable for producing appropriate filaments that can be 3D printed without clogging in the nozzle and avoiding imperfection in the printed material. The particle size of filler was also found to be an important parameter that can affect the printing properties; however, a coarser size of filler (e.g., 10 mm) was also found manageable if the speed of printing reduced and the filler loading was in the correct range.

Overall, even though there are significant studies available on FDM 3D printing, often the tensile properties of the filament part are ignored which limits the data availability from both cellulosic and protein-based fillers. In addition, some other parameters, such as changes in density and porosity of the filaments by filler loading, melting temperature, and impact on the printing parameters including the printing speed, and the temperatures in the nozzle and bed need further understanding. Addressing this in future can provide more valuable insights into the parameters responsible for filament tensile properties and help better correlate them with the properties of 3D printed specimens.

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