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A REVIEW: RELATION ON NATURAL FIBER LOADING TO THE TENSILE PROPERTIES OF TPU COMPOSITES

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ABSTRACT

The natural fiber is considered recyclable and may have some potential for some application. Nowadays, this type of fiber has to gain popularity as a filler in polymer composites. Besides, it is readily available and environmentally friendly. It is also inexpensive because they are non-toxic and biodegradable waste. This paper presents the various natural fiber from fruit skin, fruit peel, agricultural residue, waste, and others as fillers in thermoplastic polyurethane (TPU). The author also reflects the key issue related to the effect of filler loading on the tensile properties of TPU composites. In general, the tensile strength of the natural fiber-filled polymer composites increases with fiber loading. The tensile properties are mainly influenced by the interfacial adhesion between the polymer and the fibers. Other than that, each type of fiber also plays a vital role in its properties and polymer adhesion. This review only focuses on the latest studies in the range of seven years from 2013 to 2019 [1].

Keywords: *Thermoplastic polyurethane, natural fiber, fiber loading, tensile properties, composites*

INTRODUCTION

The transition towards the production of green composite is taking place due to the increasing demand of consumers to reduce synthetic material, higher sustainability, biodegradability, environmentally friendly and recyclability, inexpensive, low density, etc. Natural fibers are biologically occurring materials that have two primary sources [1], which are agriculture production, for example, coconut shell [2] and production residue of crops when they are processed for primary uses, such as sugar palm [3]. Natural fiber is majorly used with thermoplastic because this plastic is recyclable in an effortless manner. It can be combined with other materials, and for this study, the material chosen is thermoplastic polyurethane (TPU) to form a new composite. The formation of composites structure is dependent on the fiber and the TPU. Natural fibers have recently become attractive to researchers due to their low cost, non-abrasive, eco-friendly, and bio-degradability. They are now being exploited as a replacement for conventional fiber such as glass, aramid, and carbon [4]. Thermoplastic polyurethane (TPU) is a polymer that is very useful and commercially used widely in industry and daily life. TPU has no cross-link and is described as a "bridging gap between rubber and plastic" [5]. Additionally, it has wear, acid, alkali resistance, excellent mechanical range, and good polymer field potential. TPU is a physically cross-linked copolymer [6] and the typical trait is caused by a two-phase structure, a hard segment, a soft segment, and diverse segments.

The main barrier of natural fiber with the thermoplastic is due to the natural fiber's hydrophilic nature. The combination of the hydrophilic nature of the natural fibers and the hydrophobicity of the polymer matrices make the combination incompatible [4]. The composites' mechanical properties depend strongly on the interfacial adhesion between filler and matrix components [7]. Remarkably, the tensile properties of natural fiber and their compatibility with the polymer may vary due to some factors such as the fiber loading, fiber size, the adhesion between filler-matrix, fiber distribution, the strength or surface of the fiber itself, etc. There are numerous mechanical tests and testing instruments with standardized and non-standardized testing methods to determine the mechanical properties of polymer composites. One of the essential criteria in determining the performance of polymer composites materials is the tensile properties.

Tensile properties of natural fibers filled TPU composites will be focused on in this review and verified are related to the fiber loading and the fiber type. The effects of fiber content on the properties of composites are particularly significant. It is often observed that the increase in fiber loading leads to an increase in tensile properties [4]. In general, high fiber content is required to achieve the high performance of the composites. However, sometimes up to a maximum or optimum value, the tensile properties will then drop. As things go generally, high fiber content produces good composites performance. Still, the matrix does not adhere well with the saturated number of fibers at certain limits, and the tensile strength of the composites will be decreased. The performance of a natural fiber itself depends on several factors such as chemical composition and physical properties related to the type of natural fiber. This review's beauty lies in the fact that it brings recycling awareness because natural fibers are the most abundant agricultural residue and waste from domestic or business. The origin of all the natural fiber can be classified into four main categories, i.e., fruit, stem,

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Table 1 The classification and origin of studied natural fiber [2],[8]-[15]

Fruit	Stem	Leaf	Others
Coconut husk Coconut shell Durian Banana Mangosteen Rambutan Sugar palm Cocoa pod	Sugarcane Kenaf Wood Roselle	Pineapple Tea waste Hemp	Corncob Coconut residue

leaf, and others, as shown in Table 1. Therefore, the addition of natural fiber into TPU reduces the cost of the composites and keeps in reducing the waste in the environment [7]. Hemp, wood flour, fiber from fruits skin as natural filler is essential to impart reinforcement. The development of these environmentally friendly composites materials has accelerated rapidly since they are safer to deal with.

FIBER AND COMPOSITES PREPARATION

Usually, the wastes from fruit are peel, rind, and skin. Some are the cob, bagasse, and residue after extraction. Primary data collection methods are from markets, stalls, villages, and factories, and small businesses. For the stem, most of them are collected from the plantation area. The wastes were washed and undergone natural sun drying, hot air oven drying, and freeze drying to dry those fibers. Grinding, chopping, or cutting is the following methods. Some of the fibers will be extracted using the water retting process. Figure 1 represents the flow chart on the essential fiber preparation, which is commonly done by many researchers.

Melt compounding using an internal mixer [16] or two roll mills [2],[13] was a popular method for thermoplastic composites among researchers. These two compounding machines are suitable for TPU due to their melting temperature and handling. For TPU, the temperature range for compounding is about 180°C to 200°C [11]-[14],[16],[17], depending on the machine and the type of TPU. Compounding was carried out with mixing speed within the range of 30 rpm to 60 rpm [9],[11],[13],[14] for 11 minutes to 20 minutes [9],[13],[17]. The TPU matrix was initially charged into the machine, and the fibers were then added. Once a homogeneous compounding of TPU and fibers is assumed, the sample was removed and rested for 24 hours at room temperature. After that, the samples were then hot-pressed and cut into dumbbell shape for tensile testing as followed to ASTM D638 [5],[6],[13].

FIBER CONTENT/LOADING EFFECT ON TENSILE STRENGTH OF TPU COMPOSITES

Polymer composites are a kind of high-performance and versatile material formed from a combination of different phases of materials. One of the materials is a polymer which is usually the matrix, and the other one is the filler. Combining these components results in unique mechanical properties that are infeasible to be achieved with a single material [17]. Matrix and reinforcement/filler are the two main phases that are essential in developing polymer composites. These two phases are usually composed of organic polymers as the matrix and fiber as the reinforcement. Generally, the fiber materials' strength and stiffness are much higher than those of the matrix material, making the fiber the primary load-bearing component in polymer composites.

On the other hand, the matrix serves as a load distributor by uniformly transferring the applied force to the fiber. Therefore, the matrix needs to hold the fiber firmly to establish an efficient load transfer, which increases the mechanical properties of the polymer composites [1]. The performance of polymer composites is generally determined by one factor: the ratio of the fiber to the polymer matrix in the composite. This referred to the fiber content, fiber loading, or fiber fraction in the composites.

Most of the research had reported different percentages of the optimum filler loading in TPU composites, such as 40% for hemp fiber [9], 30% for kenaf fiber [12], and even 5% for pineapple [13]. It depends on the type of filler itself and the way the filler distributes inside the TPU matrices. As the filler increases, accumulation and increased filler-filler interaction occurs, resulting

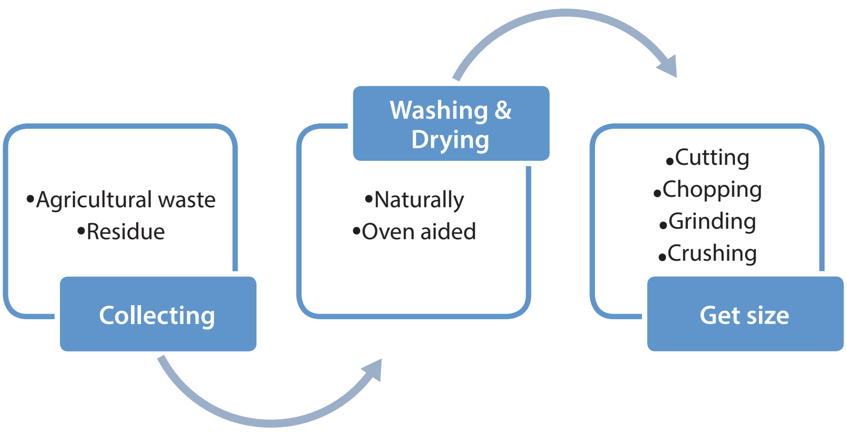

Figure 1 Flow chart of the basic fiber preparation process

in weak interaction and bonding between filler and matrix [18]. Besides that, the good adhesion, less micro-cracking, and less void also put in the reason on tensile strength [8]. Fiber or fillers work as carriers of load in the matrix [19]. The composites' power depends on several factors, such as strengths of fibers and matrix, volume fraction, and fiber-matrix interfacial bonding [17],[20].

Natural fibers are excellent reinforcing materials for preparing polymer matrix-based composites. Principally, the natural fiber reinforced polymer composites› tensile strengths increase with fiber content, up to a maximum or optimum value, and the value will then drop [21]. Based on Tina [9], they mixed fiber hemp as filler in TPU. As is known, it has also proven even supported with previous studies with the addition of fillers, and tensile will also increase. However, it is only limited so that a value is called the optimum value. Their finding proved the tensile increased until it reached 40% of hemp loading while 10% of hemp loading showed the lowest value of tensile strength (Figure 2). This can conclude that 40% of hemp in TPU composite could be the optimum loading since it reached the highest tensile strength.

Wood flour as filler from Özturk [21] research demonstrated that 70% of the filler was the optimum loading, leading to an increase in density, hardness, and tensile modulus. The tensile strength value has decreased up to 35% of filler but then increase with further addition of filler. This happened because some of the filler acted as reinforcement at certain loading. However, at some loading percentage, it may deteriorate. At high loading, usually, the filler tends to agglomerate or become lumped or clustered. The uneven fiber will be caused by poor fiber-matrix interaction, an essential obligation for excellent tensile strength. We assumed each filler has its optimum loading, which also influences the filler-matrix interaction and adhesion [5]. Hongjie [10] had done the same filler (wood flour) and exhibited low elongation at break when the filler increased. By referring to Figure 3, at higher filler loading, this composite showed low flexibility as we can see the composites showed lower elongation at break with the increasing filler loading. The tensile strength keeps dropping until it reached 20% of wood flour but slightly increased by 30% and 40%, respectively.

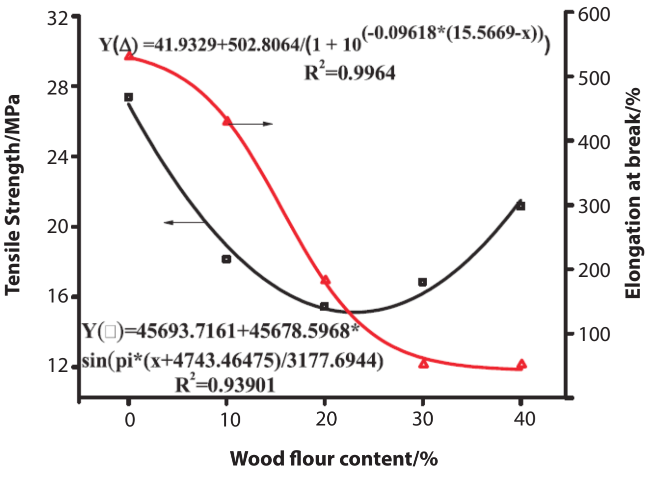

Figure 3 Tensile strength and elongation at break for various loading of wood flour in TPU [10]

The increasing cocoa pod fiber proved by El-Shekeil [11] has increased the tensile strength and tensile modulus consequently. They added 20%, 30%, and 40% of cocoa pod fiber into TPU. They proved this to the fact that the trend indicated the excellent interfacial bonding between fiber and matrix. The fibers are effectively participating in the stress transfer. The composites' strength is supported by fiber strength, matrix strength, and fiber/matrix interfacial bonding [11]. Figure 4 shows the strength-modulus plot vs. the cocoa pod loading in TPU composites.

Kenaf has been widely used in TPU as filler, as well as in other polymer composites. El-Shekiel [12] and Sapuan [16] showed their findings. For the kenaf fibers' loading in the range of 20% - 40%, the optimum loading was 30%. This led to good tensile strength, while at 40% and also 50% fiber loading, the power deteriorated, as showed in Figure 5. Low fiber loading resulted in low tensile strength attributed to the lack of fibers' ability to transfer the load to one another [12]. Datta & Kopczynska [17] also carried out kenaf as filler in TPU at 10% and 30%. The higher fiber content caused

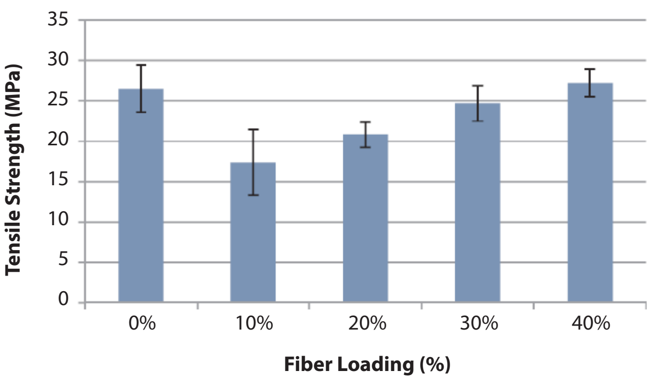

Figure 2 Tensile strength for hemp fiber/TPU composites [9]

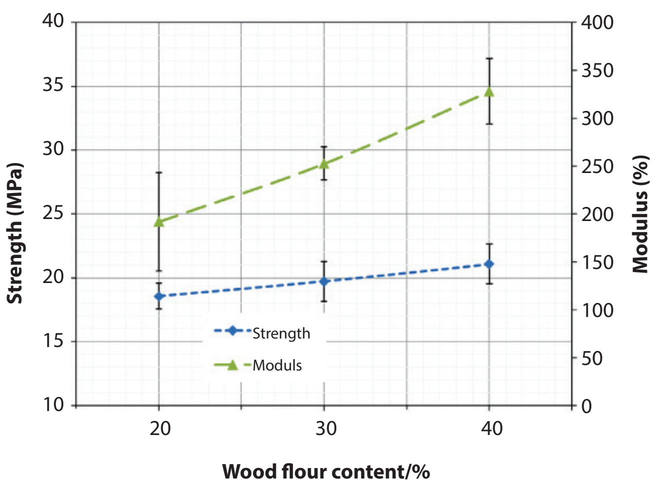

Figure 4 Strength-Modulus plot for TPU/cocoa pod composites [11]

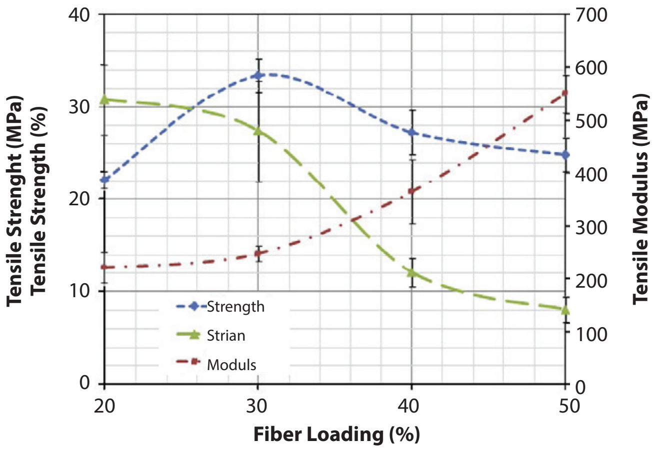

Figure 5 Strength-Modulus-Strain plot for TPU/kenaf composites [12]

the lower tensile strength to be attributable to agglomeration and poor adhesion between fiber and matrix.

The low tensile strength is due to the lack of fibers' ability to transfer the load to one another. At optimum loading, a better fiber-matrix interfacial bonding occurred. In contrast, higher loading related to high population of fibers, accumulation and stress transfer get blocks [19]. Elongation at break decreases when increasing fiber loading, a typical effect of the increased fiber volume, which is having a low strain [20]. Next, kenaf also has been studied by El-Shekeil et al. [11], which verifies loading 20%, 30%, and 40%. The tensile strength decreased when the fiber content increased. This happened due to inadequate fiber/matrix interfacial bonding. The fiber works as carriers of load in the TPU matrix. Good tensile strength depends more on practical and uniform stress distribution. When the fiber content increased, the fiber will be dispersed in the entire composites sheet.

According to Radzi et al. [8] and as presented in Figure 6, 10% loading of roselle in TPU contributed to the lowest tensile strength. As usual, the increase in filler loading leads to higher tensile strength until it reaches optimum percentage. 40% loading of roselle gave the highest tensile strength, among the ratio of 10% to 50% of filler loading. In some research, higher loading will lead

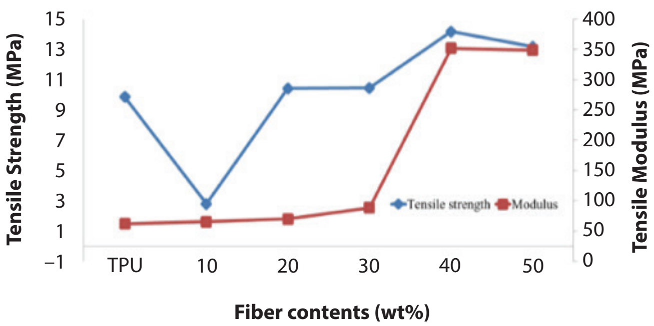

Figure 6 Tensile strength and tensile modulus of TPU/ roselle composites [8]

to accumulation in the matrix, leading to a decrease in mechanical properties, which may be revealed in Figure 7. The matrix cannot wet and fill the fiber because of the higher content.

Balan et al. [18], in the study of coconut shell powder, has utilised it from 5% to 20% as filler in TPU and confirmed the 10% of coconut shell powder as the optimum filler loading. The trend in increasing filler loading will increase the tensile properties is still proved with evidence. The filler loading greater than 10% of coconut shell powder will deteriorate these properties. The addition of filler also enhanced the degree of reinforcements but led to agglomeration, which increases the filler-filler interaction, thus resulted in weak interaction and bonding between coconut shell and TPU matrix. Ahad et al. [13] investigated coconut shell as filler at 5% to 20% and found out that 10% was the optimum content, and the tensile decreased until 20%. The coconut shell particle size and shape vary and can sort into three groups; chromatic, spherical, and fibrous [2]. Besides the type of filler and the loading issues, filler shape also plays a vital role in filler-matrix interaction [13]. This also will be affecting the distribution of the filler inside the TPU matrix.

Ahad and group [3] were interested in sugar palm/TPU with the loading between 10% - 50%, and they focused on the density of the composites vs. filler loading. Therefore, Mohammed [14] discovered the optimum sugar palm in their study, which is about 10%. Table 2 summarizes tensile strength, strain, and modulus of 10% sugar palm loading in TPU.

Tea wastes as a filler also get attention to researchers [15]. The tensile strength deteriorated with the increasing tea waste loading in TPU, which are similar trends in reducing the composite strength. This was the evidence of the strong adhesion between the fiber and the polymer, which is not necessarily enhancing the composite strength. From Figure 8, they observed the optimum filler loading for this composite is 20% and at 40%, tea waste loading is at the worst tensile strength property. Poor filler dispersion is due to excessive fiber loading lead to some accumulation of the filler. The effect of various reinforcing conditions has to consider the proper processing conditions on mechanical performance.

Researchers had been exploring ten types of natural fiber as filler in TPU [6],[13]. The range of filler loading is from 5% to 20%, as appeared in Figure 9. Pineapple, coconut shell, coconut husk, and corn cob showed the highest tensile strength at 5% and even at 10% loading of its fiber. In contrast, other types of fiber (sugarcane bagasse, banana, mangosteen, rambutan, coconut residue, and durian) were considered as low tensile strength fiber. Those natural fibers' superior mechanical properties are associated with their high cellulose and lignocelluloses [6]. Among various natural fibers, pineapples exhibit excellent mechanical properties, and this is also proved by Indra [23]. The mechanical properties of the natural fillers' composites depend on several factors such as the stress-strain behavior of fillers and matrix phases, the phase volume fractions, the fillers concentration, the fillers' distribution and orientation to one another [2].

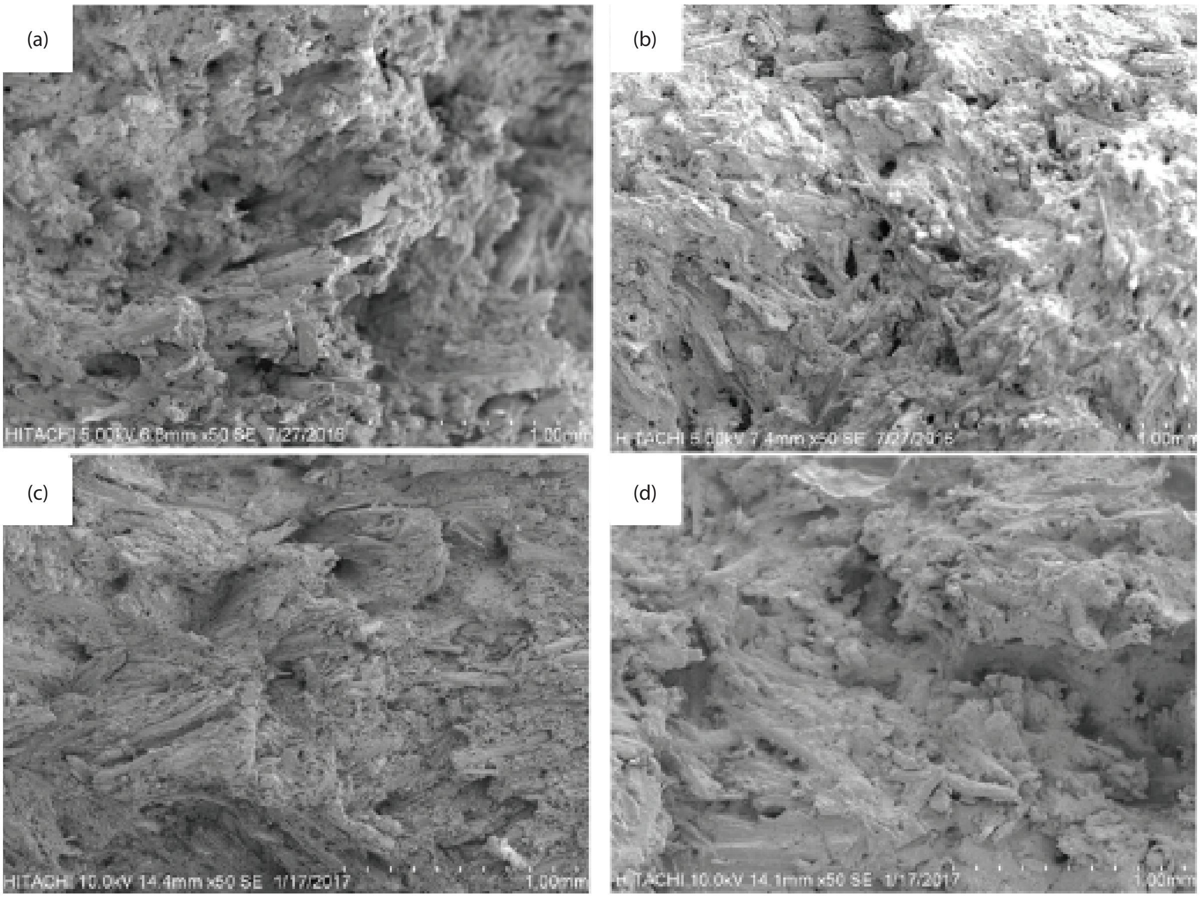

Figure 7 SEM micrograph of TPU composite at different roselle loading (a)10% (b) 20% (c) 30% (d) 40% [8]

Table 2 Mechanical properties of TPU/sugar palm composites [14]

Sample	TPU/SF composites	Tensile strain/%	Tensile strength/MPa	Tensile modulus/MPa
1	Untreated	21.21	10.38	105.53
2	Untreated	18.94	9.13	96.32
3	Untreated	22.98	8.9	90.37
4	Untreated	30.05	11.71	95.84
5	Untreated	23.99	9.36	88.48

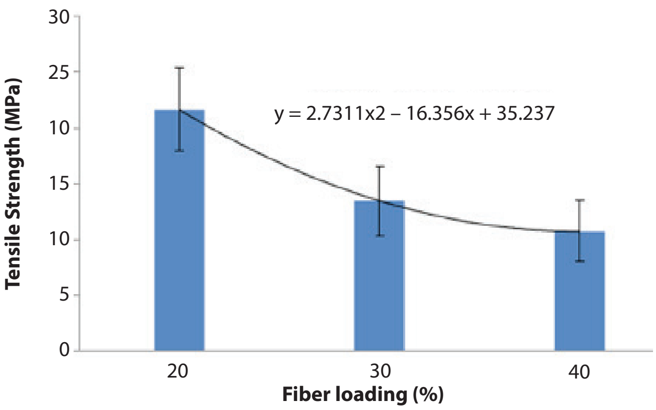

Figure 8 Tensile strength for TPU/tea waste composites at 20%, 30% and 40% loading [15]

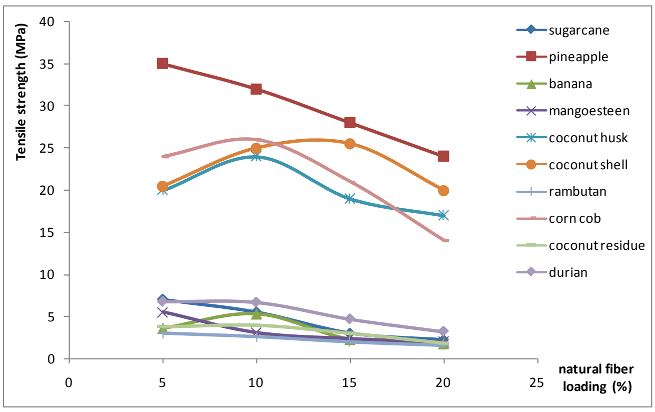

Figure 9 Tensile strength of ten types of natural fiber as filler in TPU composites, with different fiber loading of 5% to 20% [5]

The decreasing of tensile strength with high fiber content may be due to the inability to matrix TPU to hold fiber content that exceeds its optimum value [24].

Generally, we can conclude that the natural fibers present some drawbacks such as incompatibility between fibers and matrices, to form aggregates during the processing, and the poor resistance to moisture [24]. Despite the attractiveness of natural fibers in polymer matrix, they suffer from lower strength [23]. The increment in tensile strength is due to the better-increased surface area of filler in the matrix but up to optimum loading only. Further than that, there was poor interfacial bonding between the hydrophilic fiber and the hydrophobic matrix polymer, leading to decreased tensile strength [2]. The mechanical characteristics of a polymer composite filled with natural fiber mainly result from the quantity and fiber type, besides the interfacial strength between the filler and matrix [25].

CONCLUSION

Tensile testing is the most common testing to determine the properties of natural fiber composites. Furthermore, incorporating natural fiber into TPU is considered a challenge because the chemical structure of both fibers and matrix are various. The properties of TPU composites with varying fiber type and loading as well as fiber structure itself. The hydrophilic nature of the natural fiber and the fiber loading also impact the composite properties. Usually, higher fiber loading is needed to attain the good composite properties. But this review concluded that each type of natural fiber gave different results and they have their optimum loading as filler in TPU. All the types of natural fiber from fruit skin, fruit peel, agricultural residue, and also domestic waste as fillers in TPU have an excellent potential to replace non-biodegradable composites, due to their excellent performance in mechanical properties. Consequently, development in the composites field from waste can be seen in the domain of green composites in the past year.

REFERENCES

[1] L. Ranakoti, M. Pokhriyal, & K. Ankur, "Natural fibers and biopolymers characterization: A future potential composite material," *Journal of mechanical engineering,* 68, 1, pp. 33-50, 2018.

[2] J. Sarki, S.B. Hassan, V.S Aigbodian, & J.E. Oghenevweta, "Potential of using coconut shell particle filler in eco-composites materials," *Journal of alloys and compound,* 509, pp. 2381-2385, 2011.

[3] A. Atiqah, M. Jawaid, M.R. Ishak, & S.M. Sapuan, "Moisture absorption and thickness swelling behavior of sugar palm fiber reinforced thermoplastic polyurethane," *Procedia engineering,* 184, pp. 581-586, 2017.

[4] H. Ku, H. Wang, N. Pattarachaiyakoop, & M. Trada, "A review on the tensile properties of natural fiber reinforced polymer composites," *Composites Part B: Engineering,* 42, 4, pp. 856-873, 2011.

[5] N.A. Ahad, S.H. Ahmad, & J.A. Jalil, "Magnetic and electrical properties of TPU/NR blends filled NiZn ferrite," *Applied Mechanics and Materials*, 754-755, pp. 256-260, 2015.

[6] N.A. Ahad, F.Z. Rozali, N.H. Rosli, N.I.H. Hanif, & N. Parimin, "Oil and water absorption behavior of TPU/natural fibers composites," *Solid state phenomena*, 280, pp. 374-381, 2018.

[7] N.F. Zaaba & H. Ismail, "Thermoplastic /natural filler composites: A short review," *Journal Physical Science,* 30, pp. 81-99, 2019.

[8] A.M. Radzi, S.M. Sapuan, M. Jawaid, & M.R Mansor, "Influence of fiber contents on mechanical and thermal properties of roselle fiber reinforced polyurethane composites," *Fibers and Polymers,* 18, 7, pp. 1353-1358, 2017.

[9] T. Haghighatnia, A. Abbasian, & J. Morshedian, "Hemp fiber reinforced thermoplastic polyurethane composite: An investigation in mechanical properties," *Industrial crops & products,* 108, pp. 853-863, 2017.

[10] H. Bi, Z. Ren, R. Guo, M. Xu, & Y. Song, "Fabrication of flexible wood flour/thermoplastic polyurethane elastomer composites using fused deposition molding," *Industrial crops and products,* 122, pp. 76-84, 2018.

[11] Y.A. El-Shekiel, S.M. Sapuan, & M.W. Algrafi, "Effect of fiber loading on mechanical and morphological properties of cocoa pod husk fibers reinforced thermoplastic polyurethane composites," *Materials & Design,* 64,pp. 330-333, 2014.

[12] Y.A. El-Shekiel, S.M. Sapuan, K. Abdan, & E.S. Zainudin, "Influence of fiber content on the mechanical and thermal properties of Kenaf fiber reinforced thermoplastic polyurethane composites," *Materials & Design,* 40, pp. 299-303, 2012.

[13] N.A. Ahad, Y. Ju Ann, N.A. Norozi, & A.A. Azman, *Tensile strength on seven type of fruits skin fiber thermoplastic polyurethane (TPU),* Proceeding of Advanced Materials Characterization Techniques (AMCT2019), pp. 114-115, 2019.

[14] A.A. Mohammed, D. Bachtiar, M.R.M. Rejab, & J.P. Siregar, "Effect of microwave treatment on tensile properties of sugar palm fiber reinforced thermoplastic polyurethane composites," *Defence Technology,* 14, 4, pp. 287-290, 2018.

[15] Y.A. El-Shekiel, F.M. AL-Oqla, & S.M. Sapuan, "Performance tendency and morphological investigations of lignocellulosic tea waste/polyurethane bio-composite materials," *Polymer Bulletin*, 2019.

[16] S.M. Sapuan, F.L. Pua, Y.A. El-Shekiel, & F.M. Al-Oqla, "Mechanical properties of soil buried kenaf fiber reinforced thermoplastic polyurethane composites," *Materials & Design,* 50, pp. 467-470, 2013.

[17] J. Datta & P. Kopczynska, "Effect of kenaf fiber modification on morphology and mechanical properties of thermoplastic polyurethane materials," *Industrial Crops and Products,* 74, pp. 566-576, 2015.

[18] A. K. Balan, S. M. Parambil, S. Vakyath, J.T. Velayudhan, S. Naduparambath, & P. Etathil, "Coconut shell powder reinforced thermoplastic polyurethane/natural rubber blend-composites: Effect of silane coupling agents on the mechanical and thermal properties of the composites," *Journal of materials science,* 52, 11, pp. 6712-6725, 2017.

[19] Y. A. El-Shekeil, S. M. Sapuan, M. Jawaid, & O. M. Al-Shuja'a, "Influence of fiber content on mechanical, morphological and thermal properties of kenaf fibers reinforced poly (vinyl chloride)/thermoplastic polyurethane poly-blend composites," *Materials & Design,* 58, pp. 130-135, 2014.

[20] S. Diestel & A. Krause, "Wood-based composites with thermoplastic polyurethane as matrix polymer," *Journal of Applied Polymer Science,* 135, 25, p. 46344, 2018.

[21] S. Özturk, "Effect of fiber loading on the mechanical properties of kenaf and fiberfrax fiber-reinforced phenol-formaldehyde composites," *Journal of Composite Materials,* 44, 19, pp. 2265-2288, 2010.

[22] H. Ku, H. Wang, N. Pattarachaiyakoop, & M. Trada, "A review on the tensile properties of natural fiber reinforced polymer composites," *Composites Part B: Engineering,* 42, 4, pp. 856-873, 2011.

[23] M. I. Reddy, M. A. Kumar, & C. R. B. Raju, *Tensile and flexural properties of jute, pineapple leaf and glass fiber reinforced polymer matrix hybrid composites.* Materials today: proceedings, 5, 1, pp. 458-462. 2018.

[24] D. U. Shah, P. J. Schubel, P. Licence, & M. J. Clifford, "Determining the minimum, critical and maximum fibre content for twisted yarn reinforced plant fibre composites," *Composites Science and Technology,* 72, 15, pp. 1909-1917, 2012.

[25] G. P. Otto, M. P. Moisés, G. Carvalho, A. W. Rinaldi, J. C. Garcia, E. Radovanovic, & S. L. Fávaro, "Mechanical properties of a polyurethane hybrid composite with natural lignocellulosic fibers," *Composites Part B: Engineering,* 110, pp. 459-465, 2017.