



A review on the tensile properties of natural fiber reinforced polymer composites

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ABSTRACT

This paper is a review on the tensile properties of natural fiber reinforced polymer composites. Natural fibers have recently become attractive to researchers, engineers and scientists as an alternative reinforcement for fiber reinforced polymer (FRP) composites. Due to their low cost, fairly good mechanical properties, high specific strength, non-abrasive, eco-friendly and bio-degradability characteristics, they are exploited as a replacement for the conventional fiber, such as glass, aramid and carbon. The tensile properties of natural fiber reinforced polymers (both thermoplastics and thermosets) are mainly influenced by the interfacial adhesion between the matrix and the fibers. Several chemical modifications are employed to improve the interfacial matrix–fiber bonding resulting in the enhancement of tensile properties of the composites. In general, the tensile strengths of the natural fiber reinforced polymer composites increase with fiber content, up to a maximum or optimum value, the value will then drop. However, the Young's modulus of the natural fiber reinforced polymer composites increase with increasing fiber loading. Khoathane et al. [1] found that the tensile strength and Young's modulus of composites reinforced with bleached hemp fibers increased incredibly with increasing fiber loading. Mathematical modelling was also mentioned. It was discovered that the rule of mixture (ROM) predicted and experimental tensile strength of different natural fibers reinforced HDPE composites were very close to each other. Halpin–Tsai equation was found to be the most effective equation in predicting the Young's modulus of composites containing different types of natural fibers.

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1. Introduction

A fiber reinforced polymer (FRP) is a composite material consisting of a polymer matrix imbedded with high-strength fibers, such as glass, aramid and carbon [2]. Generally, polymer can be classified into two classes, thermoplastics and thermosetting. Thermoplastic materials currently dominate, as matrices for bio-fibers; the most commonly used thermoplastics for this purpose are polypropylene (PP), polyethylene, and poly vinyl chloride (PVC); while phenolic, epoxy and polyester resins are the most commonly used thermosetting matrices [3]. In the recent decades, natural fibers as an alternative reinforcement in polymer composites have attracted the attention of many researchers and scientists due to their advantages over conventional glass and carbon fibers [4]. These natural fibers include flax, hemp, jute, sisal, kenaf, coir, kapok, banana, henequen and many others [5]. The various advantages of natural fibers over man-made glass and carbon fibers are low cost, low density, comparable specific tensile properties, non-abrasive to the equipments, non-irritation to the skin, reduced energy consumption, less health risk, renewability, recyclability and bio-degradability [3]. These composites materials are suitably

applicable for aerospace, leisure, construction, sport, packaging and automotive industries, especially for the last mentioned application [3,6]. However, the certain drawback of natural fibers/polymer composites is the incompatibility between the hydrophilic natural fibers and the hydrophobic thermoplastic matrices. This leads to undesirable properties of the composites. It is therefore necessary to modify the fiber surface by employing chemical modifications to improve the adhesion between fiber and matrix [3].

There are many factors that can influence the performance of natural fiber reinforced composites. Apart from the hydrophilic nature of fiber, the properties of the natural fiber reinforced composites can also be influenced by fiber content/amount of filler. In general, high fiber content is required to achieve high performance of the composites. Therefore, the effect of fiber content on the properties of natural fiber reinforced composites is particularly significance. It is often observed that the increase in fiber loading leads to an increase in tensile properties [7]. Another important factor that significantly influences the properties and interfacial characteristics of the composites is the processing parameters used. Therefore, suitable processing techniques and parameters must be carefully selected in order to yield the optimum composite products. This article aims to review the reported works on the effects of fiber loading, chemical treatments, manufacturing techniques and process parameters on tensile properties of natural fiber reinforced composites.

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2. Tensile properties

Generally, the tensile properties of composites are markedly improved by adding fibers to a polymer matrix since fibers have much higher strength and stiffness values than those of the matrices as shown in Tables 1–3 [3,8].

Consider the tensile strength of S-glass from Table 1, and that of polypropylene (PP) from Table 2 and that of polyester resin from Table 3, it can be found that the tensile strength of the fiber (S-glass) is 75–150 times higher than those of the matrices (PP and polyester resin). It can also be found that the Young's modulus of the fiber (S-glass) is 80–160 times higher than those of the matrices (PP and polyester resin) [3–8].

In general, higher fiber content is desired for the purpose of achieving high performance of short fiber reinforced polymer composites (SFRP) [7]. It is often observed that the presence of fiber or other reinforcement in the polymeric matrix raises the composite strength and modulus [5]. Therefore, the effect of fiber content on the tensile properties of fiber reinforced composites is of particular interest and significance for many researchers [7].

Nonwoven mats from hemp and polypropylene fibers in various proportions are mixed and hot pressed to make composite materials. The effect of hemp fiber content and anisotropy are examined on the basis of tensile properties of the resultant composite materials. The tensile strength, with fibers in the perpendicular direction, tended to decrease with increasing hemp fiber content (a maximum decrease of 34% at 70% of hemp) as depicted in Fig. 1. Whereas, the tensile strength, with fibers in the parallel direction, showed a different trend and a maximum value was found with increasing fiber loading. It was found that the tensile strength of composites with fibers in the perpendicular direction was 20–40% lower than those of composites with fibers in parallel direction. Since the fibers lay perpendicular to the direction of load, they cannot act as load bearing elements in the composite matrix structure but become potential defects which could cause failure. As expected, better tensile properties are found in the specimens cut from the composite sheets parallel to the direction of carding as depicted in Fig. 1 [9].

In general, the Young's modulus of the composite materials increase with an increase in fiber content, reaching a maximum value at 50% hemp fiber loading and then decreasing slightly at 70% hemp fiber content. The Young's modulus was almost two and a half times higher at 50% hemp fiber loading than at 0% fiber content, i.e. pure PP as depicted in Fig. 2 [9].

Fig. 3 illustrated the tensile strength of 20-mesh hardwood, 40-mesh hardwood, flax and rice hull fibers reinforced HDPE composites. Li et al. [5] reported that flax fiber content from 10% to 30% by mass was mixed with high-density polyethylene (HDPE) by extrusion and injection moulding to produce bio-composites. The re-

sults showed that increasing fiber content resulted in increasing tensile properties initially as depicted in Fig. 3. It peaked at 20% by volume; it then dropped. However, the elongation at break of the composites showed the reverse trend as depicted in Fig. 4 [5].

The tensile strengths of 40-mesh hardwood fibers reinforced HDPE composites increased gradually, and up to a maximum at 25% of fiber loading by volume, and then dropped back as depicted in Fig. 3 [11]. On the other hand, the tensile strengths of 20-mesh hardwood fibers reinforced HDPE composites reduced with increasing fiber loading [11]. This is totally different from that of 40-mesh hardwood fibers. The tensile strengths of rice hull fibers reinforced HDPE composites were shown in Fig. 3 [10]; the behaviour of the curve was more or less the same as those found in 20-mesh hardwood but it has a maximum tensile strength at 5% by volume of fiber content [10]. The tensile strengths decreased with increasing particulate loading slightly [10].

Fig. 5 showed the Young's modulus of 20-mesh hardwood, 40-mesh hardwood, flax and rice hull fiber reinforced HDPE composites with varying percentage by volume of fiber loading. It can be found that the Young's modulus of 20-mesh and 40-mesh hardwood fibers reinforced HDPE composites with fiber loading of 0–40 wt.% [11]. The value increased with increasing fiber loading. Up to 30% volume fraction of hardwood, the Young's moduli of 20-mesh hardwood fiber composites were lower than their counterparts. After 35% volume fraction of hardwood, the Young's moduli of 20-mesh hardwood fiber composites were higher than their counterparts. Fig. 5 also illustrated the Young's modulus of flax fibers reinforced HDPE composites with fiber loading of 0–40% vol. [5]. It can be found that the Young's modulus increased with increasing fiber content [5]. The Young's modulus of rice hulls fibers reinforced HDPE composites with fiber loading of 0–40% vol. was depicted in Fig. 5 [10]. The trends of all the curves for Fig. 5 were more or less the same as, i.e. the values of the Young's modulus increased progressively with increasing fiber loading. However, the largest increase with increasing fiber content was for flax fiber reinforced composites, while the least increase was for rice hull fiber reinforced composites.

The dependence of tensile properties of micro winceyette fiber reinforced thermoplastic corn starch composites on fiber contents was studied. Fig. 6 illustrated that with the increase fiber content from 0% to 20 wt.%, the tensile strength was approximately trebled to 150 MPa [12]. The increase was progressive. However, the elongation of the composites decreased with increasing fiber loading as depicted in Fig. 7. The elongation dropped significantly between fiber loading of 0–10% by weight; after this the decrease was very slightly. On the other hand, the energy at break of the composites decreased slightly from neat resin to 5 wt.% of fiber and dropped significantly from 5% to 10% by weight of fiber as depicted in Fig. 8; after this there was a slight increase [12].

Table 1
Properties of selected natural and manmade fibers [adapted from 3, 8].

Fiber	Density (g/cm ³)	Elongation (%)	Tensile strength (MPa)	Elastic modulus (GPa)	Refs.
Cotton	1.5–1.6	7.0–8.0	400	5.5–12.6	[6,7]
Jute	1.3	1.5–1.8	393–773	26.5	[6]
Flax	1.5	2.7–3.2	500–1500	27.6	[4]
Hemp	1.47	2–4	690	70	[4]
Kenaf	1.45	1.6	930	53	[4]
Ramie	N/A	3.6–3.8	400–938	61.4–128	[8]
Sisal	1.5	2.0–2.5	511–635	9.4–22	[8]
Coir	1.2	30	593	4.0–6.0	[9]
Softwood kraft pulp	1.5	4.4	1000	40	[9]
E-glass	2.5	0.5	2000–3500	70	[9]
S-glass	2.5	2.8	4570	86	[9]
Aramid (Std.)	1.4	3.3–3.7	3000–3150	63.0–67.0	[9]
Carbon (Std. PAN-based)	1.4	1.4–1.8	4000	230–240	[9]

Table 2

Properties of typical thermoplastic polymers used in natural fiber composite fabrication [adapted from 3, 8].

Property	PP ^a	LDPE	HDPE	PS	Nylon 6	Nylon 6,6
Density (g/cm ³)	0.899–0.920	0.910–0.925	0.94–0.96	1.04–1.06	1.12–1.14	1.13–1.15
Water absorption-24 h (%)	0.01–0.02	<0.015	0.01–0.2	0.03–0.10	1.3–1.8	1.0–1.6
T _g (°C)	–10 to –23'	–125	–133 to –100'	N/A	48	80
T _m (°C)	160–176	105–116	120–140	110–135'	215	250–269
Heat deflection Temp (°C)	50–63	32–50	43–60	Max. 220	56–80	75–90
Coefficient of thermal expansion (mm/mm/°C × 10 ⁵)	6.8–13.5	10	12–13	6–8	8–8.86	7.2–9
Tensile strength (MPa)	26–41.4	40–78	14.5–38	25–69	43–79	12.4–94
Elastic modulus (GPa)	0.95–1.77	0.055–0.38	0.4–1.5	4–5'	2.9	2.5–3.9
Elongation (%)	15–700	90–800	2.0–130	1–2.5	20–150	35–>300
Izod impact strength (J/m)	21.4–267	>854	26.7–1068	1.1	42.7–160	16–654

^a PP = polypropylene, LDPE = low density polyethylene, HDPE = high-density polyethylene and PS = polystyrene.**Table 3**

Properties of typical thermoset polymers used in natural fiber composites [adapted from 3, 8].

Property	Polyester resin	Vinyl ester resin	Epoxy
Density (g/cm ³)	1.2–1.5	1.2–1.4	1.1–1.4
Elastic modulus (GPa)	2–4.5	3.1–3.8	3–6
Tensile strength (MPa)	40–90	69–83	35–100
Compressive strength (MPa)	90–250	100	100–200
Elongation (%)	2	4–7	1–6
Cure shrinkage (%)	4–8	N/A	1–2
Water absorption (24 h@20 °C)	0.1–0.3	0.1	0.1–0.4
Izod impact strength (J/m)	0.15–3.2	2.5	0.3

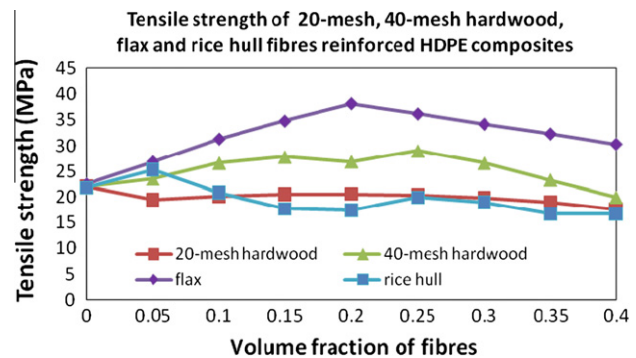
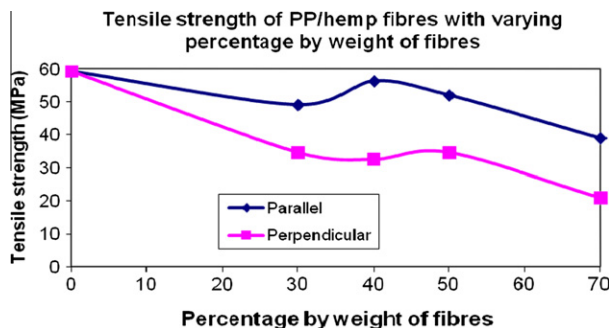
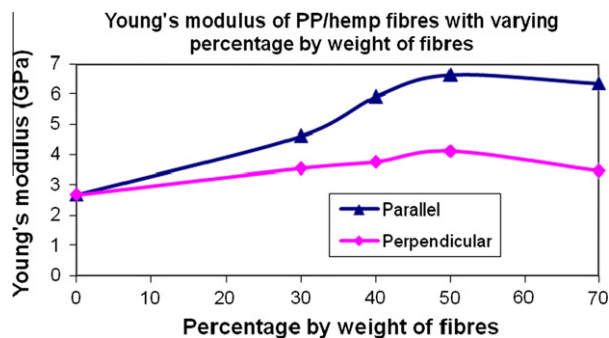
**Fig. 3.** Tensile strength of 20-mesh hardwood, 40-mesh hardwood, flax and rice hull fibres reinforced HDPE composites [adapted from 5, 10 and 11].**Fig. 1.** Tensile strength of polypropylene/hemp fibres with varying percentage by weight of fibres [adapted from 9].**Fig. 2.** Young's modulus of polypropylene/hemp fibres with varying percentage by weight of fibres [adapted from 9].

Fig. 9 illustrated that with the increase of fiber content from 0% to 20 wt.%, the Young's modulus was approximately trebled to 140 MPa [12]. From 0% to 10% by weight of fiber loading, the

Young's modulus was steady but increased progressively after that [12].

Khoathane et al. [1] found that increasing the amount of bleached hemp fiber (0–30 w/t%) resulted in the initial increase of tensile strength of the fiber reinforced 1-pentene/polypropylene (PP1) copolymer composite at 5% fiber content to 30 MPa from 20 MPa for the neat resin as depicted in Fig. 10. The tensile strength then dropped to a low 23 MPa at 20% fiber loading [1]. After this, the tensile strength increased again and its value was about at par with that of 5% fiber content when the fiber was 30% [1]. Fig. 11 illustrated the effect of fiber contents on Young's modulus of bleached hemp fiber reinforced PP1 composites [1]. The value of the Young's modulus increased by over twice from 1.3 GPa (neat resin) to 4.4 GPa (30 w/t) [1].

Long-discontinuous natural fibers of kenaf and of jute reinforced polypropylene (PP) composites fabricated by carding and hot pressing process with fiber weight fraction varying from 10% to 70% in steps of 10% were studied [13]. The experimental results illustrated that the tensile and modulus strength of both kenaf and jute fiber reinforced PP composites increased with increasing fiber loading and a maximum was reached before falling back at higher fiber weight fraction. These were illustrated in Figs. 12 and 13 [13].

From the above citations and discussions, it can be found that the values of the tensile strength of natural fiber reinforced composites increased with increasing fiber loading up to a maximum or optimum value before falling back. However, it is generally true that the values of the Young's modulus increased progressively with increasing fiber loading. On the other hand, some researchers found totally the opposite trend to the increase of composite strength with increasing fiber content. This can be attributed to many factors such as incompatibility between matrix and fibers, improper manufacturing processes, fiber degradation and others.

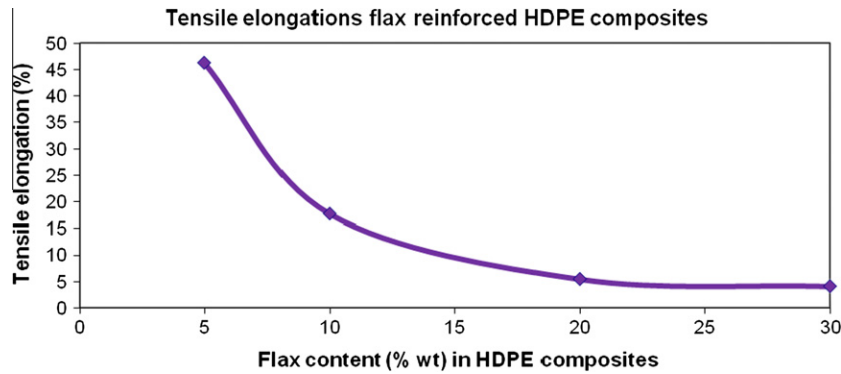


Fig. 4. Tensile elongation of bio-composites vs. fiber mass concentration [adapted from 5].

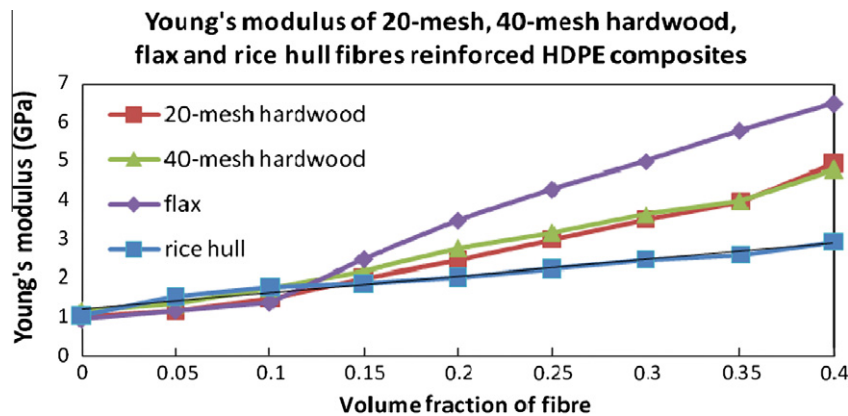


Fig. 5. Young's modulus of 20-mesh hardwood, 40-mesh hardwood, flax and rice hull fibre reinforced HDPE composites with fibre loadings of 0–40% vol. [adapted from 5, 10 and 11].

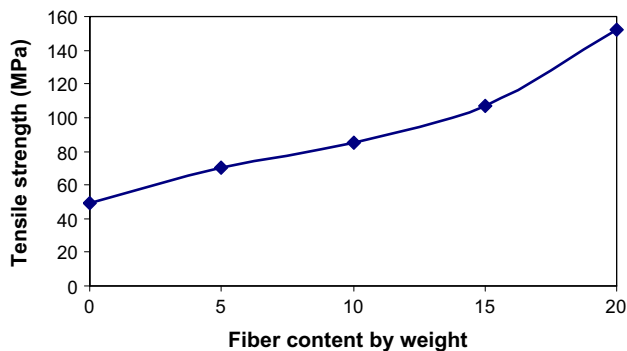


Fig. 6. The effect of fiber content on the tensile strength of micro winceyette fiber reinforced thermoplastic corn starch composites [adapted from 12].

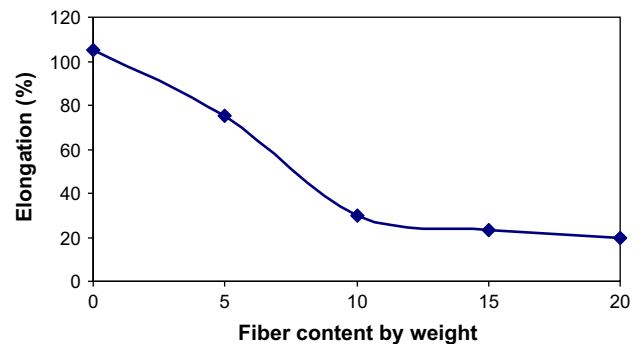


Fig. 7. The effect of fiber content on the elongation of micro winceyette fiber reinforced thermoplastic corn starch composites [adapted from 12].

The hydrophilic nature of natural fibers is incompatible with hydrophobic polymer matrix and has a tendency to form aggregates. These hydrophilic fibers exhibit poor resistant to moisture, which lead to high water absorption, subsequently resulting in poor tensile properties of the natural fiber reinforced composites. Moreover, fiber surfaces have waxes and other non-cellulosic substances such as hemi-cellulose, lignin and pectin, which create poor adhesion between matrix and fibers. Therefore, in order to improve and develop natural fiber reinforced polymer composites with better tensile properties, it is necessary to increase fibers hydrophobicity by introducing the natural fibers to surface chemical modification (surface treatment). The fiber modification is attempted to improve fibers hydrophobic, interfacial bonding between matrix

and fiber, roughness and wettability, and also decrease moisture absorption, leading to the enhancement of tensile properties of the composites [13–17].

The different surface chemical modifications, such as chemical treatments, coupling agents and graft co-polymerization, of natural fibers aimed at improving the tensile properties of the composites were performed by a number of researchers. Alkali treatment, also called mercerization, is one of the most popular chemical treatments of natural fibers. Sodium hydroxide (NaOH) is used in this method to remove the hydrogen bonding in the network structure of the fibers cellulose, thereby increasing fibers surface roughness [13]. This treatment also removes certain amount of lignin, wax and oils covering the external surface of the fibers cell wall,

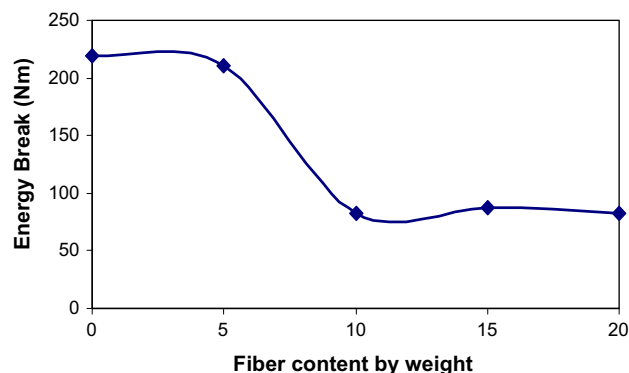


Fig. 8. The effect of fiber contents on the energy at break of micro winceyette fiber reinforced thermoplastic corn starch composites [adapted from 12].

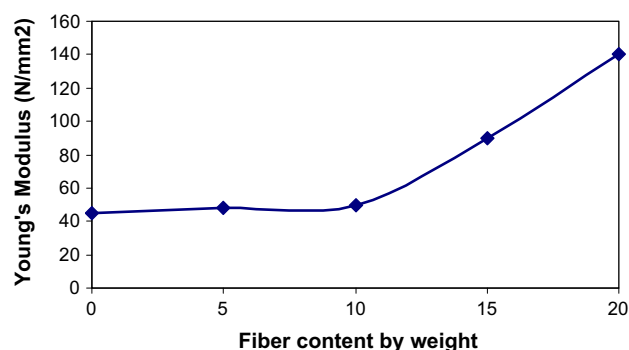


Fig. 9. The effect of fiber contents on the Young's modulus of micro winceyette fiber reinforced thermoplastic corn starch composites [adapted from 12].

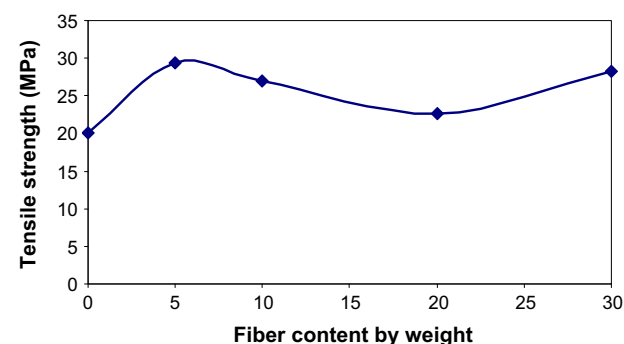


Fig. 10. The effect of fiber contents on tensile strength of bleached hemp fiber reinforced PP1 composites [adapted from 1].

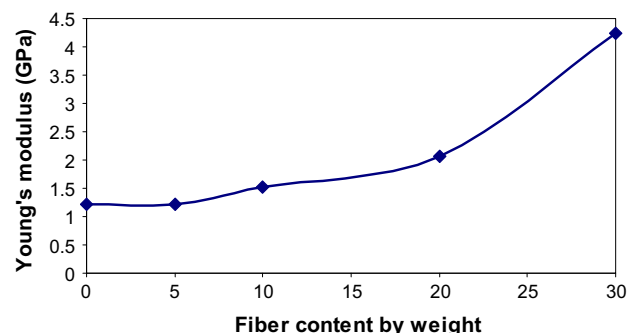


Fig. 11. The effect of fiber contents on Young's modulus of bleached hemp fiber reinforced PP1 composites [adapted from 1].

depolymerises the native cellulose structure and exposes the short length crystallites [14]. Acrylic acid treatment was also reported to be effective in modifying the natural fibers surface. A study on flax fibers-reinforced polyethylene bio-composites by Li et al. found that the efficiency of such a treatment was higher than alkali and silane treatment [14].

The chemical coupling method is also one of the important chemical methods, which improve the interfacial adhesion. In this method the fiber surface is treated with a compound that forms a bridge of chemical bonds between fiber and matrix. The chemical composition of coupling agents allows them to react with the fiber surface forming a bridge of chemical bonds between the fiber and matrix. Most researchers found these treatments were effective and showed better interfacial bonding [13]. Among different coupling agents, maleic anhydride is the most commonly used. In general, the literature reports improvements in tensile strength and elongation at break when maleic anhydride grafted matrices are used as compatibilizers (coupling agent) [15].

Hu and Lim [18] investigated that alkali treatment significantly improved the tensile properties of hemp fiber reinforced polylactic acid (PLA) compare to those untreated. Figs. 14 and 15 showed that the composites with 40% volume fraction of alkali treated fiber have the best tensile properties. The tensile strength and tensile modulus of the composites with 40% treated fiber are 54.6 MPa and 85 GPa respectively, which are much higher than neat PLA, especially for the tensile modulus which is more than twice of that of neat PLA (35 GPa).

Fuqua and Ulven reported that fiber loading of treated (alkali and bleached) and untreated flax fiber without compatibilizer (maleic anhydride grafted polypropylene or MAPP) in PP composites caused inferior tensile strength (even compared with pure PP) [19]. However, treated fiber loading with compatibilizer resulted in favourable tensile strength as depicted in Fig. 16 [19]. Fig. 17 illustrated that the continuously increased trend of composite modulus can be found in all cases (untreated, bleached and treated) and reached a maximum value at 65/5/30 (wt.% PP/MAPP/fiber loading) [19]. This can be argued that the introduction of alkali treatment with 5% MAPP in the natural fiber reinforced plastic composites helped to improve both tensile strength and Young's modulus of the composites compare to those without MAPP.

Liu et al. evaluated the effects of different fiber surface modifications, 2%NaOH, 2 + 5%NaOH (Note that 2 + 5% NaOH treatment is a continuation treatment from 2%NaOH process and then soaked with 5% NaOH) and coupling agent, on jute/polybutylene succinate (PBS) bio-composites [20]. The experiment results showed that surface modifications could remove surface impurities, increased surface roughness and reduced diameter of jute fiber, subsequently, significantly increased the tensile strength and modulus of the composites but decreased breaking elongation as depicted in Figs. 18–20. It was observed that the bio-composites of jute fibers treated by 2%NaOH, 2 + 5%NaOH or coupling agent, obviously had their tensile properties increased when compared to those untreated and yielded an optimum value at fiber content of 20 wt.%. The results also showed that the strength and stiffness of composites were dependent on the types of treatment. In Figs. 21 and 22, the 100/0/0 referred to w/t% of PP (100%), MAPP (0%) and fiber loading (0%); while 65/5/30 referred to w/t% of PP (65%), MAPP (5%) and fiber loading (30%).

Li et al. [14] studied flax fiber reinforced polyethylene bio-composites. In the study, flax fibers, containing 58 w/t% of flax shives were used to reinforce polyethylene (high-density polyethylene and linear low density polyethylene). The composites contained 10 w/t% of fiber and processed by extrusion and injection moulding. Five surface modification methods, alkali, silane, potassium permanganate, acrylic acid, and sodium chlorite treatments, were employed to improve the interfacial bonding between fibers and matrix. Fig. 21

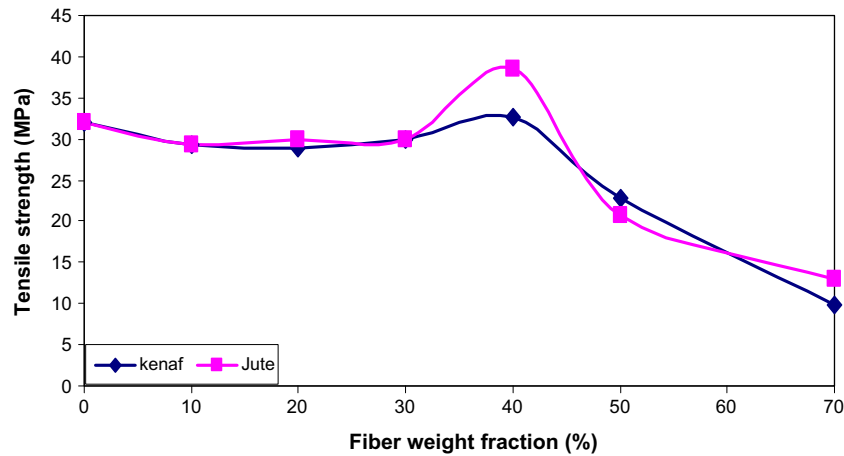


Fig. 12. Tensile strength of bio-composites of PP vs. fibre weight fraction [adapted from 13].

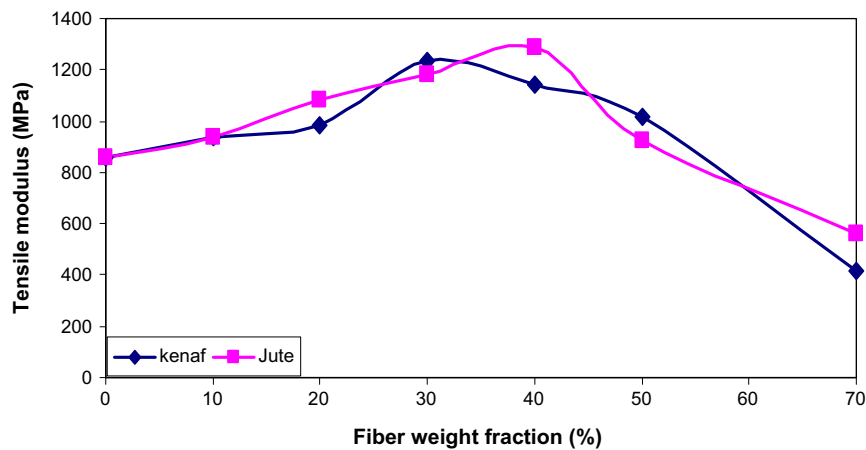


Fig. 13. Tensile modulus of bio-composites of PP vs. fibre weight fraction [adapted from 13].

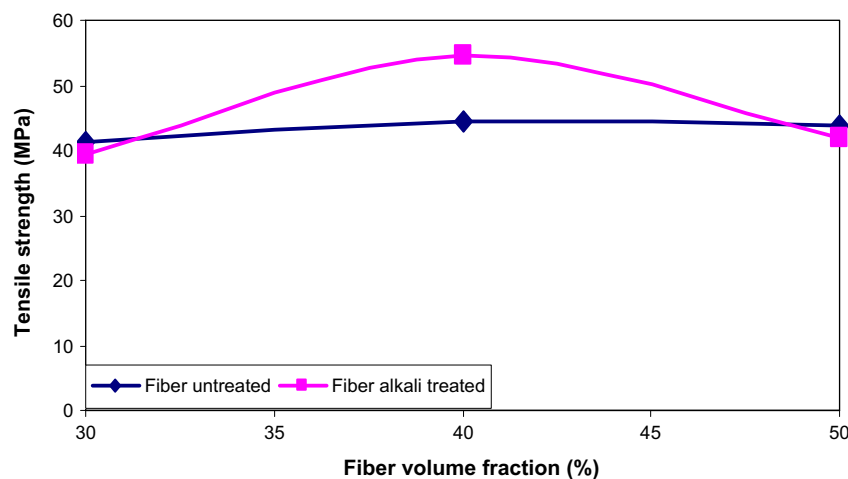


Fig. 14. Tensile strength of treated and untreated hemp-PLA composites vs. fibre content [adapted from 18].

(LLDPE) and Fig. 22 (HDPE) showed that the biocomposite tensile strengths were increased after surface modifications. Among these surface modification techniques, acrylic acid was found to be a relatively good method in enhancing tensile properties of both flax/HDPE and LLDPE bio-composites [14].

Fuqua and Ulven investigated the different MAPP loading (0, 5 and 10 w/t%) effects on tensile properties of corn chaff fiber reinforced polypropylene composites [19]. They also investigated the effect of various treatments, silane z-6011, silane z-6020 and 5 w/t% MAPP, on corn chaff fiber & distilled dried grains (DDGS)

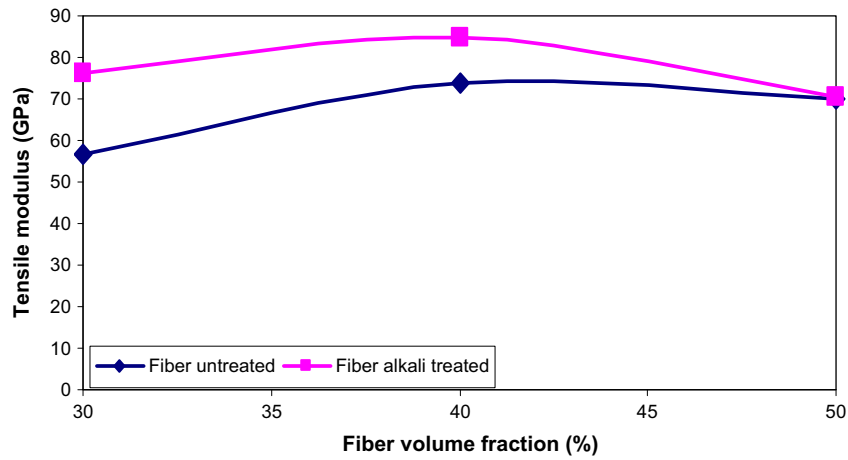


Fig. 15. Tensile modulus of treated and untreated hemp-PLA composites vs. fibre content [adapted from 18].

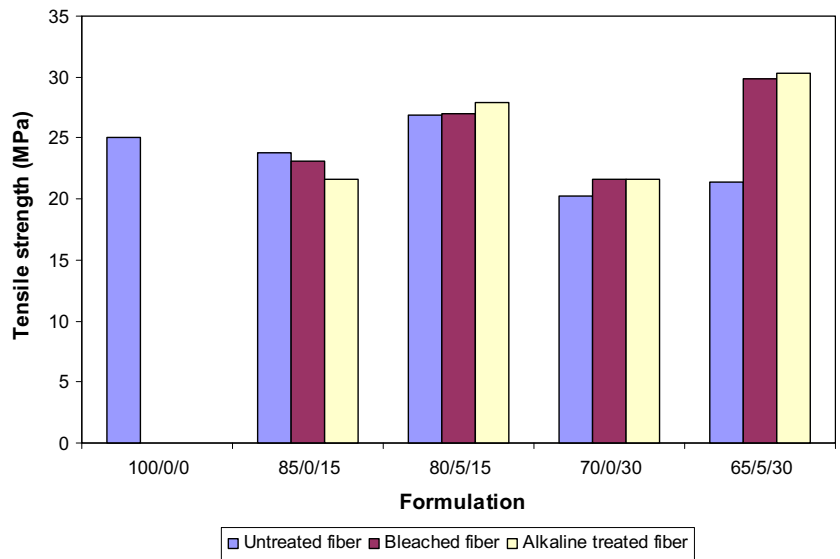


Fig. 16. Effect of coupling agent concentration on tensile strength of PP composites with 10% w/t coir fibre [adapted from 19].

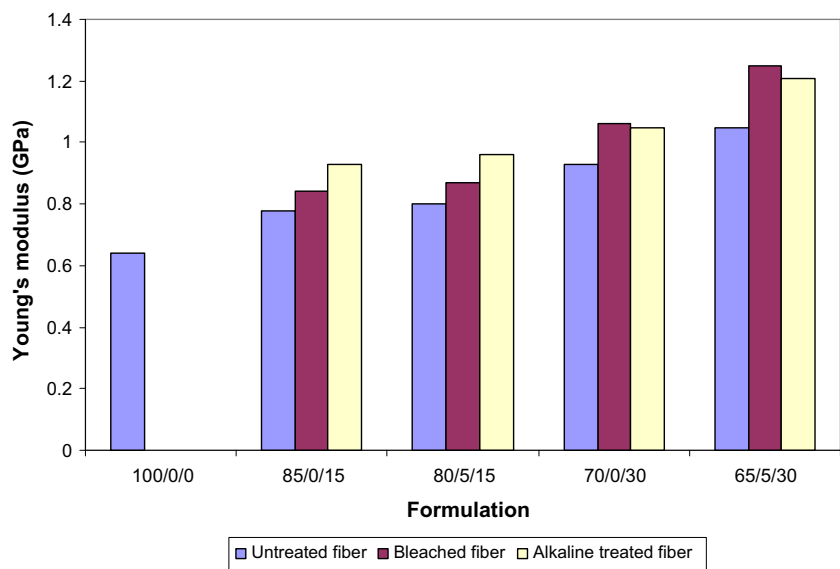


Fig. 17. Effect of coupling agent concentration on Young's modulus of PP composites with 10 w/t% coir fibre [adapted from 19].

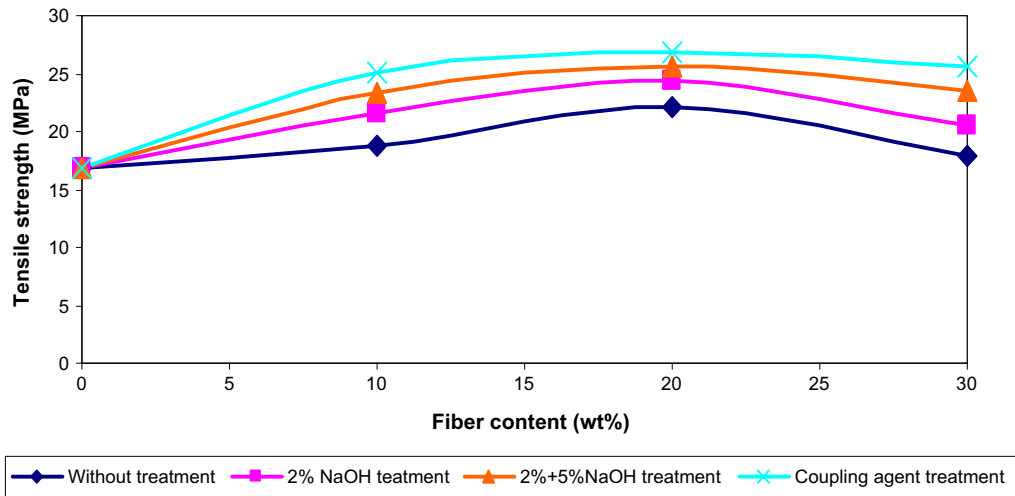


Fig. 18. Effect of surface modification on tensile strength of PBS/jute bio-composites with different fibre loading [adapted from 20].

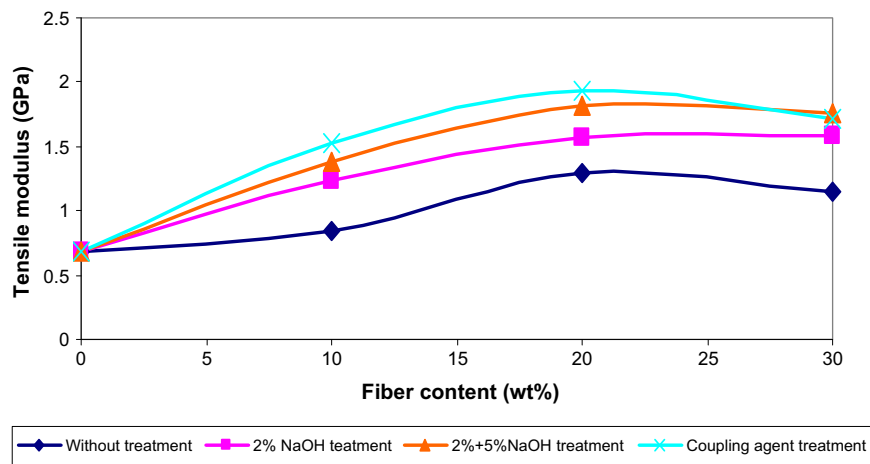


Fig. 19. Effect of surface modification on tensile modulus of PBS/jute bio-composites with different fibre loading [adapted from 20].

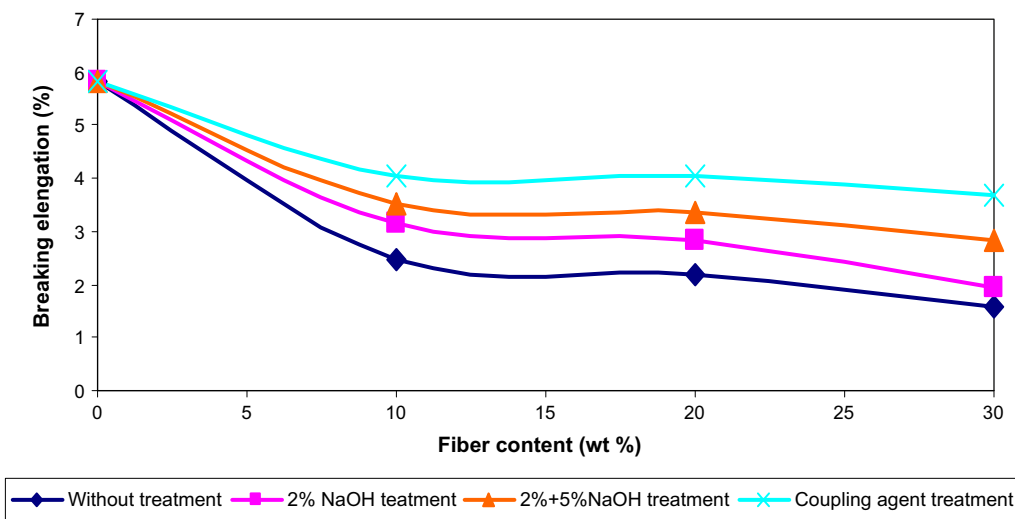


Fig. 20. Effect of surface modification on breaking elongation of PBS/jute bio-composites with different fibre loading [adapted from 20].

reinforced polypropylene composites [19]. It was found that 5 w/t% MAPP yielded the optimum value for the composites in term of

tensile strength and modulus as shown in Figs. 23 and 24 respectively [19]. The strength reduction observed with high MAPP

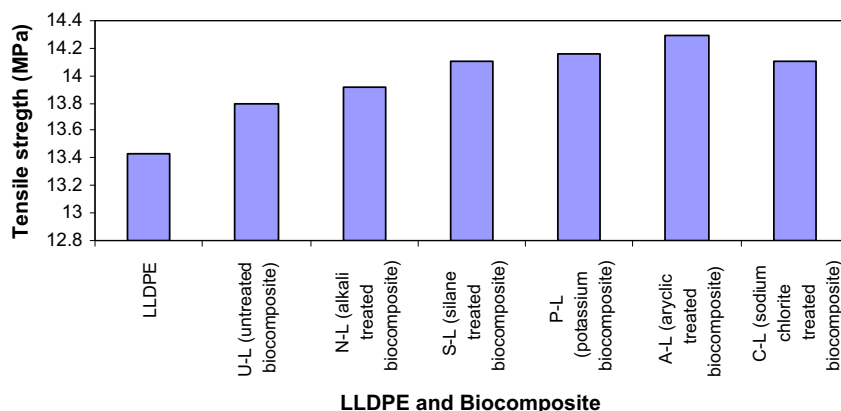


Fig. 21. Tensile strength of LLDPE and fibre-LLDPE bio-composites after surface modifications [adapted from 14].

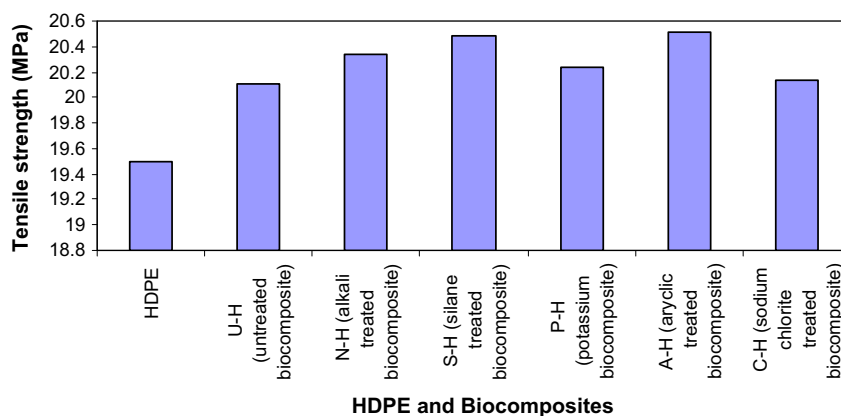


Fig. 22. Tensile strength of HDPE and fibre-HDPE bio-composites after surface modifications [adapted from 14].

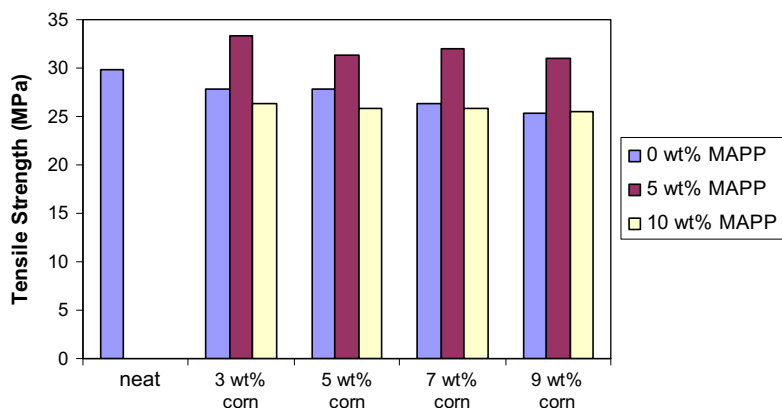


Fig. 23. Effects of MAPP loading on tensile strength of corn chaff fibre reinforced PP composites [adapted from 19].

loading was caused by the interaction between the compatibilizer (MAPP) and the fiber/matrix system. The anhydride units of MAPP maintain loop confirmations within the composite systems, since they all can act with equal probability with the cellulose in the corn fibers. Coupled with MAPP's low average molecular weight, the interaction between the PP matrix and MAPP becomes dominated principally by Van der Waals forces; since chain entanglement of PP and MAPP is virtually impossible. MAPP that is not utilized for fiber/matrix adhesion and is therefore mechanically

harmful to the composites, which leads credence to the significant performance variation between 5 and 10 w/t% loadings. However, through the use of 5 w/t% MAPP, it was found that the tensile properties of the composites increase, especially tensile strength compared to neat resin and those untreated.

Sain et al. investigated the effect of a low-molecular weight MAPP on tensile properties of polypropylene reinforced with the varieties of natural fibers such as old newsprint, kraft pulp and hemp [20]. Figs. 25 and 26 showed that the optimum level of the

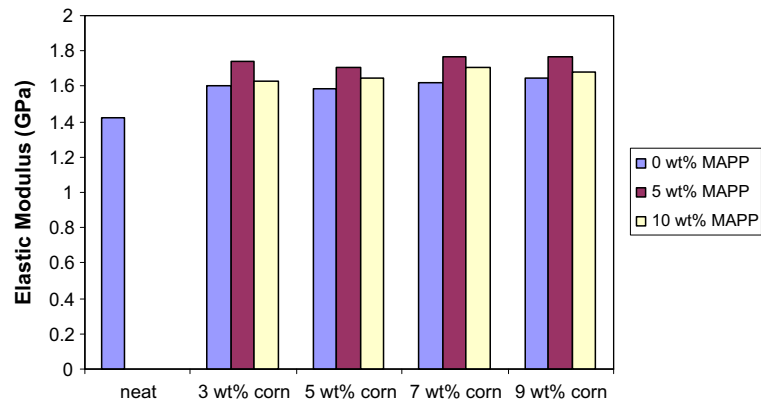


Fig. 24. Effects of MAPP loading on tensile modulus of corn chaff fibre reinforced PP composites [adapted from 19].

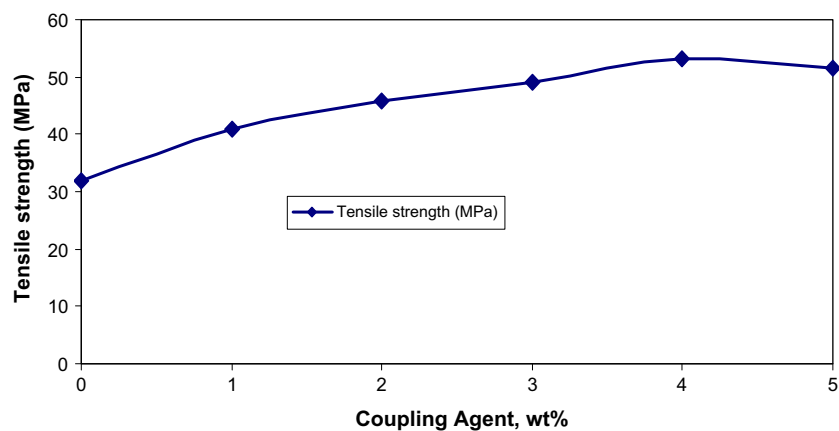


Fig. 25. Tensile strength of MAPP loaded old newsprint-filled PP composites [adapted from 20].

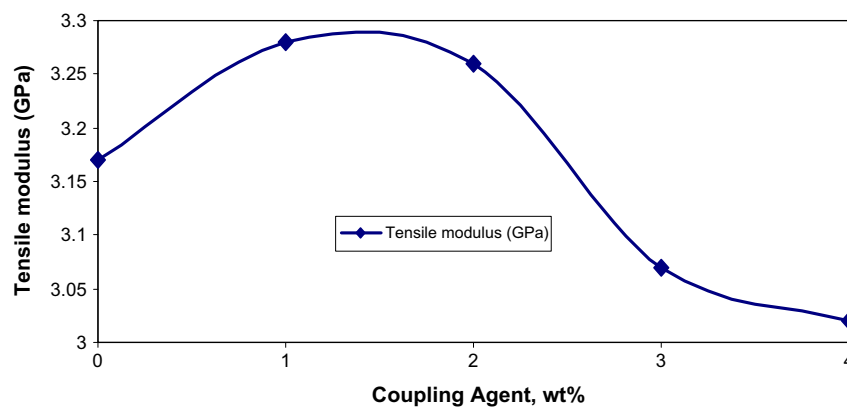


Fig. 26. Tensile modulus of MAPP loaded old newsprint-filled PP composites [adapted from 20].

coupling agent (MAPP) by weight of the old newsprint-filled PP composites was 4% for tensile strength and 1.5% for tensile modulus respectively [20].

Herrero-Franco and Valadez-Gonzalez studied the tensile behaviour of HDPE reinforced with continuous henequen fibers, which were treated by the optimum concentration (0.015 wt.%) of silane coupling agent concentration [21]. The results indicated that silane increased tensile strength of the composite. It was noticed, however, that none of the fiber–matrix interface improve-

ments had any significant effect on the value of Young's modulus of continuous henequen fiber reinforced HDPE composites [21].

Another important factor that significantly influences the properties and interfacial characteristics of the composites is processing techniques and parameters used. Common methods for manufacturing natural fiber reinforced thermoplastic composites are extrusion-injection moulding and compression moulding. Tungjitpornkull and Sombatsompop researched on the difference in the tensile properties of E-glass fiber (GF) reinforced

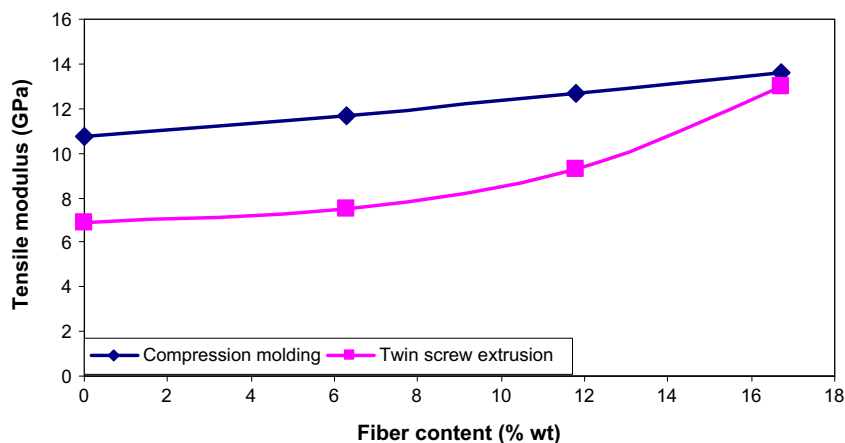


Fig. 27. Tensile modulus of glass fibre reinforced WPVC composites manufactured by twin screw extrusion and compression moulding processes [adapted from 22].

wood/PVC (WPVC) composites, manufactured by twin screw extrusion and compression moulding processes respectively [22]. The experimental results suggested that the GF/WPVC composites produced from compression moulding gave better tensile modulus than those from their counterparts as depicted in Fig. 27. The shear stress in compression moulding was lower than that in twin screw extrusion, as a result there was less thermal degradation of PVC molecules and less breakage of glass fiber, resulting in longer fiber length in the composites manufactured by compression moulding. The composite manufactured by compression moulding would have higher specific density, which resulted in less void and air and was then stronger than its counterpart [22].

The study by Siaotong et al. aimed to determine the optimum values for fiber content by mass (0%, 12.5% and 25%), extrusion barrel zone temperatures (75–110–120–130–140 °C and 75–120–130–140–150 °C) and extrusion screw speed (110 and 150 rpm) for the production of flax fiber reinforced polyethylene (HDPE and LLDPE) composites [23]. Response surface methodology was applied as optimization technique over three response variables: density deviation (%), tensile strength (MPa) and water absorption (% mass increase) of the composites. According to statistical analysis, the optimum values that yield the highest tensile strength (17.09 MPa for LLDPE composite and 21.70 MPa for HDPE composite) were: fiber content of 6.25%, barrel zone temperatures of 75–116–126–136–146 °C and screw speed of 118 rpm for LLDPE composites, and fiber content of 5%, barrel zone temperatures of 75–118–128–138–148 °C and screw speed of 128 rpm for HDPE composites. The optimum values of temperatures (T) were closer to the higher levels (75–120–130–140–150 °C) because lower temperatures result in inconsistent melt of resin that can lead to non-uniform dispersion of the fibers in the composites and eventually lower the tensile strength. The optimum values of screw speed were closer to the lower level (110 rpm). This was because the higher screw speed led to shorter residence time, non-uniform dispersion of fibers, high porosity, and consequently, lowers tensile strength. However, the unexpected result was the very low optimum level of the fiber content. Theoretically, an increase of flax fibers should improve the mechanical properties of the composites, yet, the results of tensile strength negated this [23].

Li et al. determined the appropriate value of injection temperature and pressure for flax fiber reinforced high-density polyethylene bio-composites. The results showed that higher fiber content in composites led to higher mechanical strength [24]. Injection temperature of lower than 192 °C was recommended for better composite quality because at higher temperature, fiber degradation (fiber degradation temperature \approx 200 °C) might have oc-

curred, therefore, lead to inferior tensile properties. However, the injection temperature should not be lower than 160 °C in order to ensure adequate melting of matrix. In comparison with injection temperature, the influence of injection pressure was not obvious. However, higher injection pressure is preferred to obtain better composite tensile properties [24].

The optimum pressure was determined for the natural fibre mat (hemp and kenaf) reinforced acrylic resin manufactured by high-tech vacuum compression process. Fig. 28 showed that the maximum pressure for the composites was at 60 bars. Above this value, there was a decrease in tensile properties of the composites due to the damage of the fiber structure. The advantages of using vacuum technology are to allow a reduction of the press time to a minimum without decreasing the performance of the cured materials. In addition, the work conditions were significantly improved when the vacuum chamber process was used. [25].

Khondker et al. studied the processing conditions of unidirectional jute yarn reinforced polypropylene composites fabricated by film stacking methods [26]. From optical micrographs obtained, they suggested that there must be an optimum processing temperature for which this composite might perform better in tensile properties. According to the optical microscopy results, they showed that the composites moulded at a temperature of 160 °C for 15 min and under 2.0 MPa moulding pressure, would have the PP matrix films fused and the PP melted completely and penetrated into the fiber bundles. This temperature was considered favourably ideal for the processing of composites that used lignocellulosic fibers as reinforcement, as most lignocellulosic fibers cannot withstand processing temperatures higher than 175 °C for longer duration, and hence limiting their ability to be used with some thermoplastic resins [26].

The effect of the melting-mixing technique parameters on the tensile properties of sisal fiber reinforced polypropylene composites were optimised by varying the 29 through 32, mixing time of 10 min, rotor speed of 50 rpm and a mixing temperature of 170 °C were found to be the optimum mixing conditions. For mixing times (Figs. 29 and 30), below the optimum value, the tensile strength and Young's modulus were low because of ineffective mixing and poor dispersion of the fiber in PP matrix. As the mixing time was increased, melting of PP resin became extensive and resulted in better fiber distribution into the matrix. When mixing time was more than 10 min, fiber breakage and degradation would happen, leading to a decrease in tensile properties. For mixing temperatures (Fig. 31), the performance of short fiber composites was controlled directly by fiber aspect ratio, quality of dispersion and

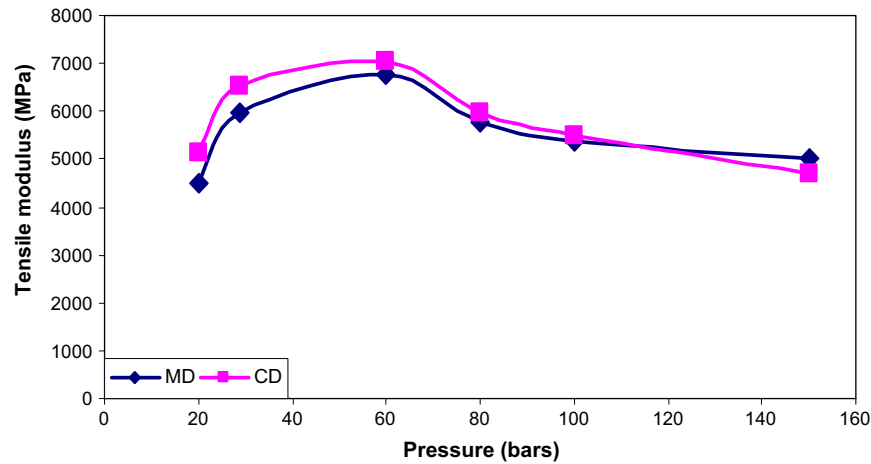


Fig. 28. Tensile modulus of natural fiber mat (hemp and kenaf) reinforced acrylic composites in machine direction (MD) and cross direction (CD) under varying pressures [adapted from 25].

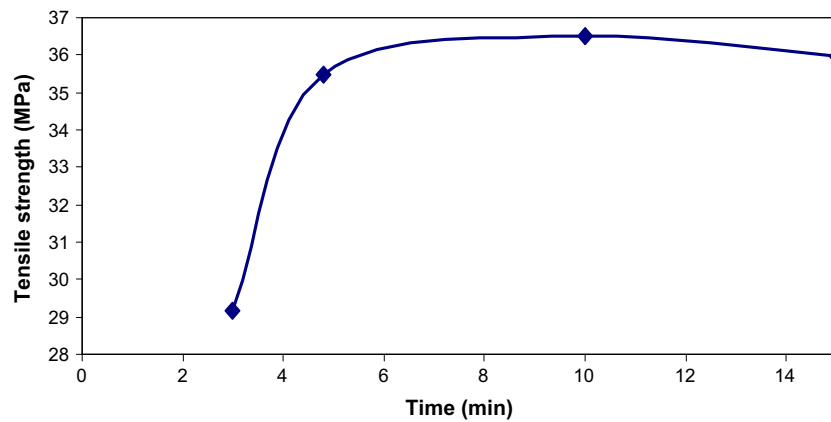


Fig. 29. Tensile strength of melting mixing of PP/sisal composites with varying mixing times; fibre content 30%, fibre length 10 mm [adapted from 27].

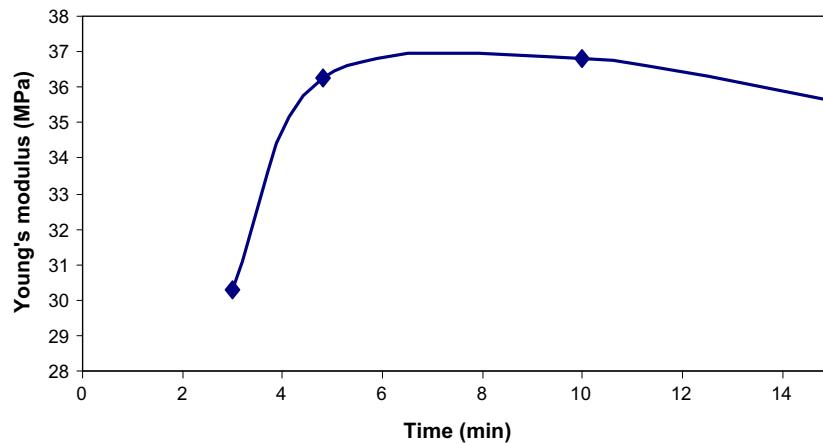


Fig. 30. Tensile modulus of melting mixing of PP/sisal composites with varying mixing times; fibre content 30%, fibre length 10 mm [adapted from 27].

interface between fiber and polymer. Below the optimum value, viscosity as well as shear stress generated in the mixture was very high, resulting in the break down of fibers to shorter lengths during mixing, leading to a lower tensile strength of the composites. On

the other hand, if mixing temperature was above the optimum, the thermal degradation of fibers would occur, leading to the decrease of tensile properties. For mixing speeds (Fig. 32), low tensile strength was observed at speeds lower than the optimum value

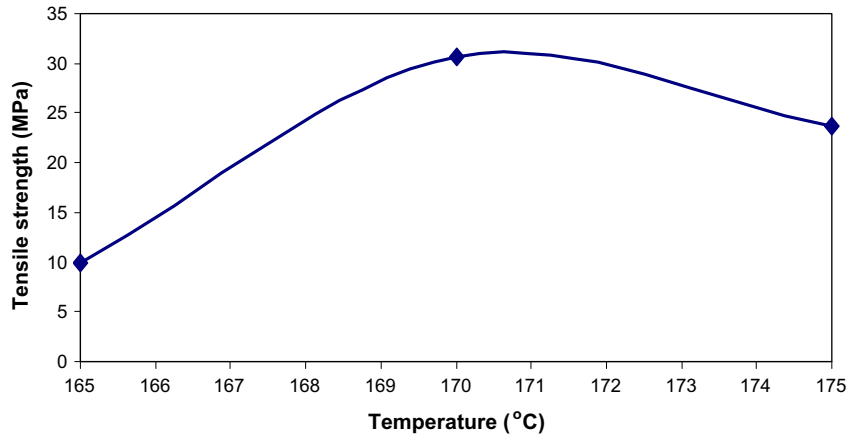


Fig. 31. Tensile strength of melting mixing of PP/sisal composites with varying mixing temperatures; fibre content 30%, fibre length 10 mm [adapted from 27].

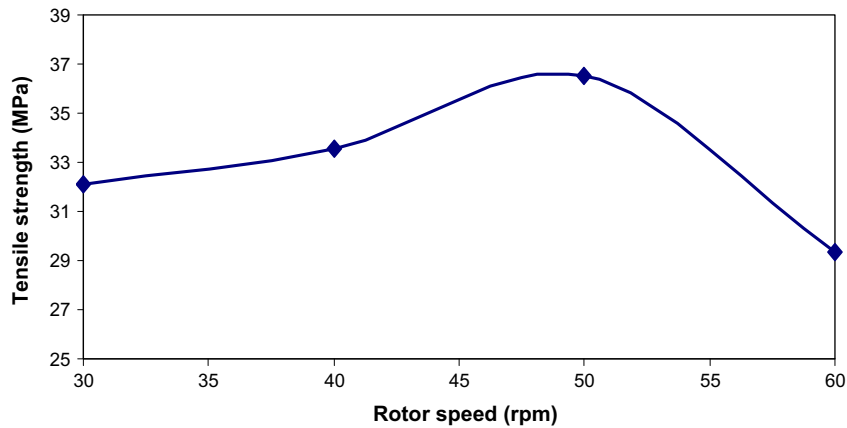


Fig. 32. Tensile strength of melting mixing of PP/sisal composites with varying rotor speeds; fibre content 30%, fibre length 10 mm [adapted from 27].

due to poor dispersion of fibers in molten PP matrix. Above the optimum rotor speed, there was a reduction in strength because of fiber breakage at high rotor speed [27].

3. Mathematical modelling

Facca et al. exploited a micromechanical model which was a semi-empirical modification of the rule of mixtures (ROM) strength Eq. (10):

$$\sigma_{1U} = \sigma_{FU} \left(1 - \frac{l_c}{2l} \right) V_F + \sigma_M^* (1 - V_F), l \geq l_c \quad (1)$$

The modified equation for cylindrical fibers was

$$\sigma_{1U} = \alpha \tau_i V_F \frac{l}{d} + \sigma_M^* (1 - V_F), l \leq l_c \quad (2)$$

The modified equation for rectangular fibers was

$$\sigma_{1U} = \alpha \tau_i V_F \left(\frac{l}{2} \right) \left(\frac{W + T}{WT} \right) + \sigma_M^* (1 - V_F), l \leq l_c \quad (3)$$

where σ_{1U} , α , τ_i , σ_M^* , l , l_c , V_F , d , W , T are composite tensile strength, the clustering parameter, interfacial shear strength, maximum stress evaluated at the peak composite strength, fiber length, critical fiber length, fiber volume fraction, cylindrical fiber diameter, rectangular fiber width, rectangular fiber thickness, respectively.

All of the above-mentioned parameters are available from literature to predict the tensile strength of HDPE reinforced with a variety of natural fibers (hemp, hardwood flour and rice hulls) and synthetic (E-glass) fibers [10].

Note that, the direction of short fiber is assumed to be perfectly aligned and fiber curvature is negligible. Also, experimental approaches are required to determine the interfacial shear strength (τ_i) of the fiber; either fiber pullout or fragmentation test can be used. Figs. 33–37 showed the predicted and experimental tensile strength of different natural fiber reinforced HDPE composites [10]. It was found that for most cases the tensile strength of the predicted and experimental results were at par. It can be argued that Eqs. (1)–(3) gave a good prediction of the experimental results except those shown in Fig. 37, where, the experimental tensile strength of HDPE composites reinforced with rice hulls fibers initially increased to a maximum value of 24.88 MPa at 5 vol.% of rice hulls fiber; it then gradually dropped to a minimum value of 17.11 MPa at 40 vol.% of filler. On the other hand, the predicted tensile strength of the composites initially decreased to a minimum value of 21.78 MPa at 5 vol.% of rice hulls fiber; it then gradually increased to a maximum value of 28.78 MPa at 25 vol.% of filler before dropping back to 24.11 MPa at 40 vol.% of filler [10].

Facca et al. also found that the increase by weight of natural short fibers like hemp, hardwood, rice hulls in high-density polyethylene manufactured by twin-screw brabender mixer compounding and compression moulding, increased the tensile modulus of all composites [11]. Again, in order to reduce cost

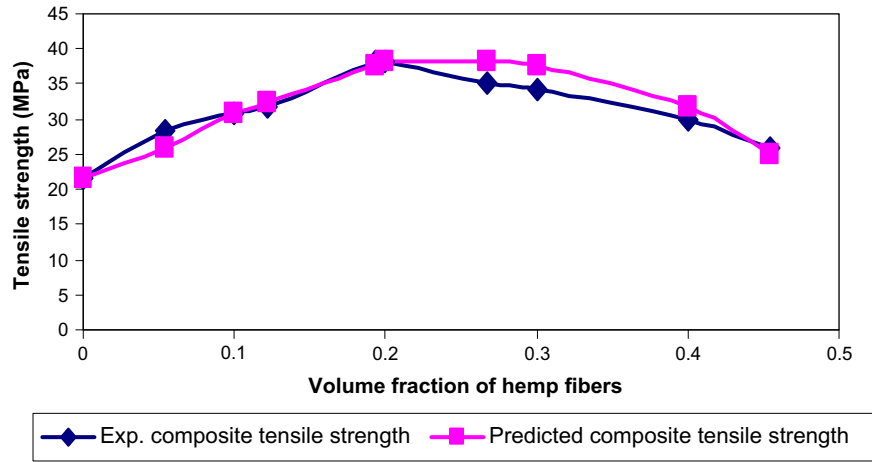


Fig. 33. Predicted and experimental tensile strengths of HDPE composite reinforced with hemp fibers between fiber loadings of 10–60 wt.% [adapted from 10].

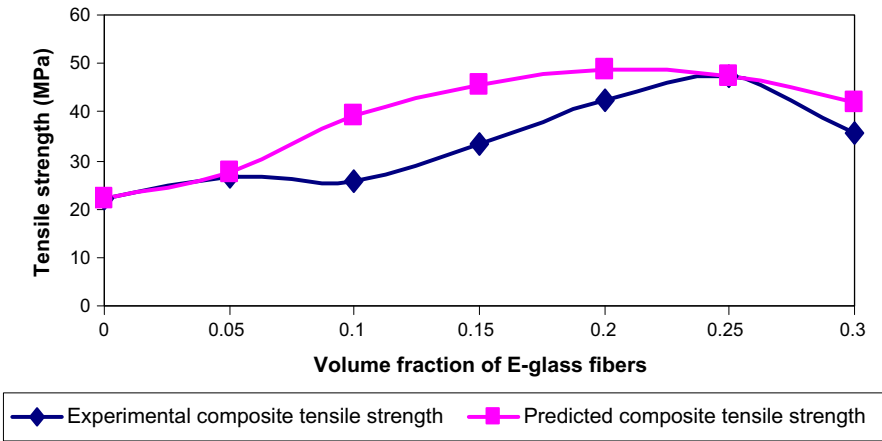


Fig. 34. Predicted and experimental tensile strengths of HDPE composites reinforced with E-glass fibers between fiber loadings of 10–60 wt.% [adapted from 10].

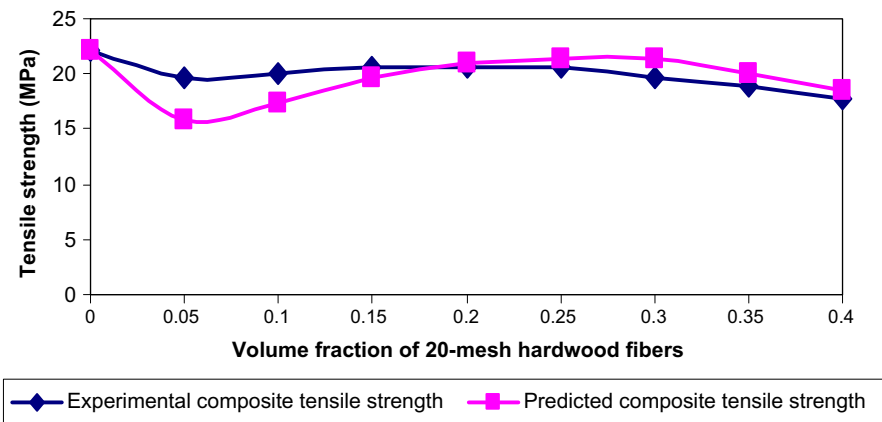


Fig. 35. Predicted and experimental tensile strengths of HDPE composites reinforced with 20-mesh hardwood fibers between fiber loadings of 10–60 wt.% [adapted from 10].

and time consuming experiments, the experimental results of the tensile modulus of the composites were compared with the theoretical values obtained from various mathematical models shown in Eqs. (4)–(9):

(1) Rule of mixture (ROM) [11]:

$$E = E_F V_F + E_M V_M$$

(4)

where E_F , V_F , E_M and V_M are the moduli and volume fractions of the fiber and matrix respectively.

(2) Inverse/transverse rule of mixtures (IROM) [11]:

$$E = \frac{E_F E_M}{V_M E_F + V_F E_M} \quad (5)$$

where E_F , V_F , E_M and V_M are the moduli and volume fractions of the fiber and matrix respectively.

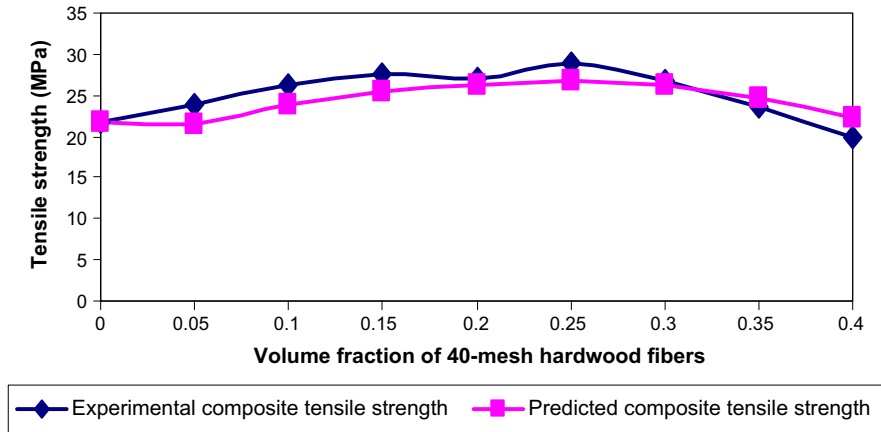


Fig. 36. Predicted and experimental tensile strengths of HDPE composite reinforced with 40-mesh hardwood fibers between fiber loadings of 10–60 wt.% [adapted from 10].

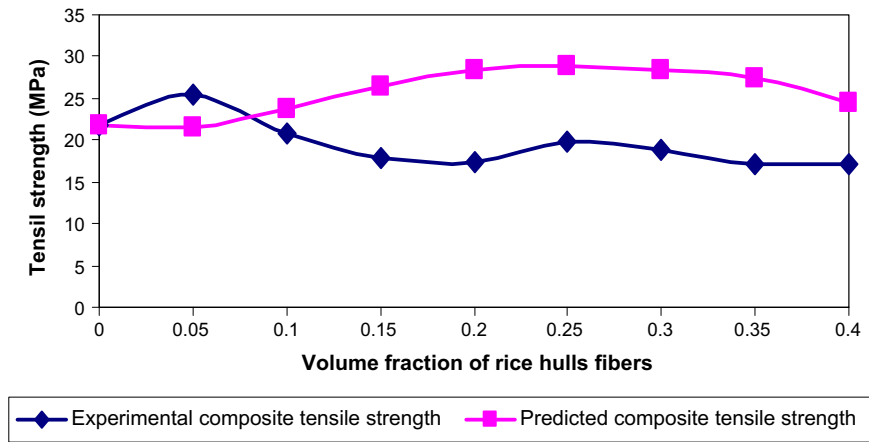


Fig. 37. Predicted and experimental tensile strength of HDPE composites reinforced with rice hulls fibers between fiber loadings of 10–60 wt.% [adapted from 10].

(3) Halpin–Tsai Eq. (11):

$$E = E_M \left(\frac{1 + \xi \eta V_F}{1 - \eta V_F} \right) \quad (6)$$

In Eq. (6) the parameter η is given as:

$$\eta = \frac{(E_F/E_M) - 1}{(E_F/E_M) + \xi} \quad (7)$$

where ξ in Eqs. 6 and 7 is a shape fitting parameter to fit Halpin–Tsai equation to experimental data. The significance of the parameter ξ is that it takes into consideration the packing arrangement and the geometry of the reinforcing fibers.

A variety of empirical equations for ξ are available in the literature, and they depend on the shape of the particle and on the modulus that is being predicted. If the tensile modulus in the principal fiber direction is desired, and the fibers are rectangular or circular in shape, then ξ is given by the following equations:

$$\xi = 2 \left(\frac{L}{T} \right) \quad \text{or} \quad \xi = 2 \left(\frac{L}{D} \right) \quad (8)$$

where L is the length of a fiber in the one-direction and T or D is the thickness or diameter of the fiber.

(4) Shear-lag theory:

$$E = E_F \left(1 - \frac{\tanh(\frac{\eta L}{2})}{(\frac{\eta L}{2})} \right) V_F + E_M V_M \quad (9)$$

the parameter η for shear-lag analysis is available on the literature [10].

$$E = \frac{3}{8} E_1 + \frac{5}{8} E_2 \quad (10)$$

where E is the elastic modulus of the composite. E_1 and E_2 are the elastic moduli of randomly oriented fiber reinforced composites given by Halpin–Tsai equations (Eq. (12)) [14]:

$$E_i = E_M \left(\frac{1 + \xi_i \eta_i V_F}{1 - \eta_i V_F} \right), \quad \eta_i = \frac{(E_F/E_M) - 1}{(E_F/E_M) + \xi_i} \quad (11)$$

where $\xi_i = 2(l_f/d_f)$ for $i = 1$ or $\xi_i = 0.5$ for $i = 2$

Figs. 38–42 showed the Young's modulus of natural fibers (hemp, hardwood, rice hulls and E-glass) reinforced high-density polyethylene composites containing different types of natural fibers at different volume fraction of the fibers. It was found that, Halpin–Tsai model was the most accurate amongst others to predict tensile modulus of natural fiber reinforced thermoplastics used in the study made by Facca et al. [11].

Lee et al. found that the tensile moduli of the kenaf or jute reinforced PP composites increased with increasing fiber contents up to 40% fiber weight fraction. Furthermore, the study employed Tsai and Pagano's model (Eq. (11)) in predicting the tensile modulus of randomly oriented long-discontinued fiber reinforced composites. It was found that the model predictions agreed well with experimental results for the volume fraction of less than 30–40% by weight of kenaf and jute respectively, where the void content were not high as illustrated in Figs. 43 and 44 respectively [13].

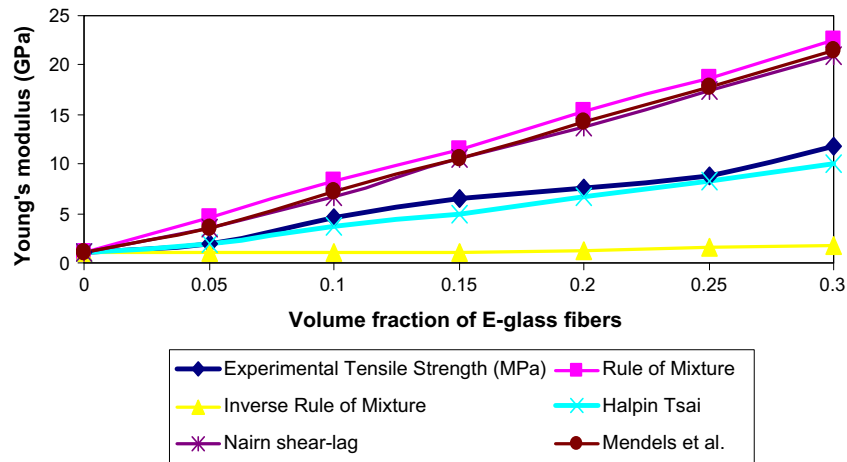


Fig. 38. Young's modulus of HDPE composites containing E-glass fibers [adapted from 11].

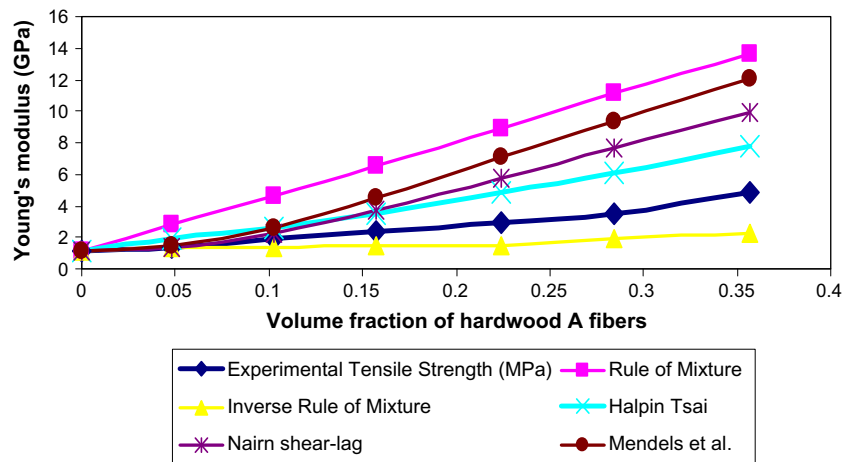


Fig. 39. Young's modulus of HDPE composites containing hardwood A [adapted from 11].

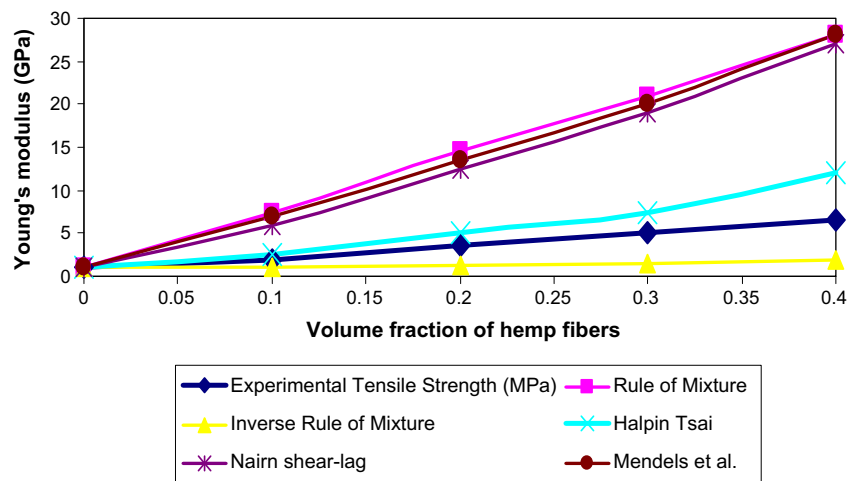


Fig. 40. Young's modulus of HDPE composites containing hemp fibers [adapted from 11].

4. Discussions and conclusions

The scientific world is facing a serious problem of developing new and advanced technologies and methods to treat solid wastes, particularly non-naturally-reversible polymers. The processes to

decompose those wastes are actually not cost-effective and will subsequently produce harmful chemicals. Owing to the above ground, reinforcing polymers with natural fibers is the way to go. In this paper, most of the natural fibers mentioned were plant-based but it should be noted that animal fibers like cocoon

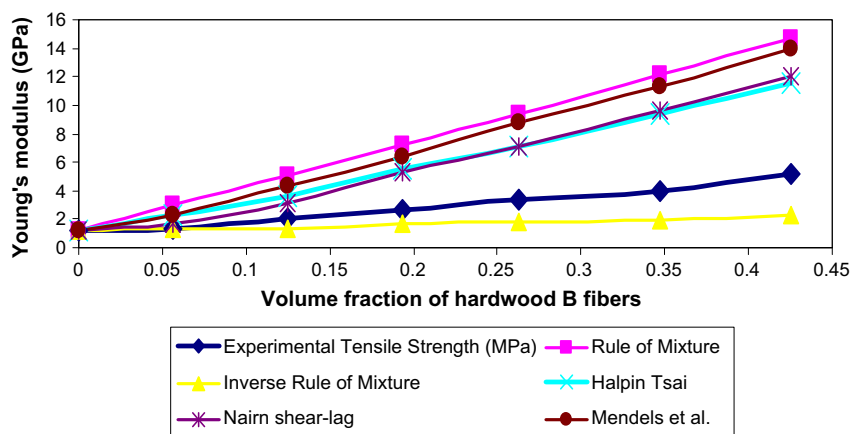


Fig. 41. Young's modulus of HDPE composites containing hardwood B [adapted from 11].

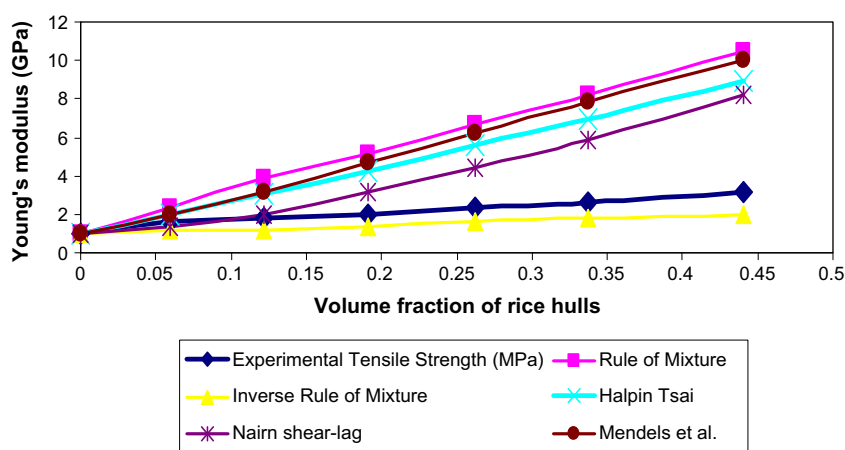


Fig. 42. Young's modulus of HDPE composites containing rice hulls [adapted from 11].

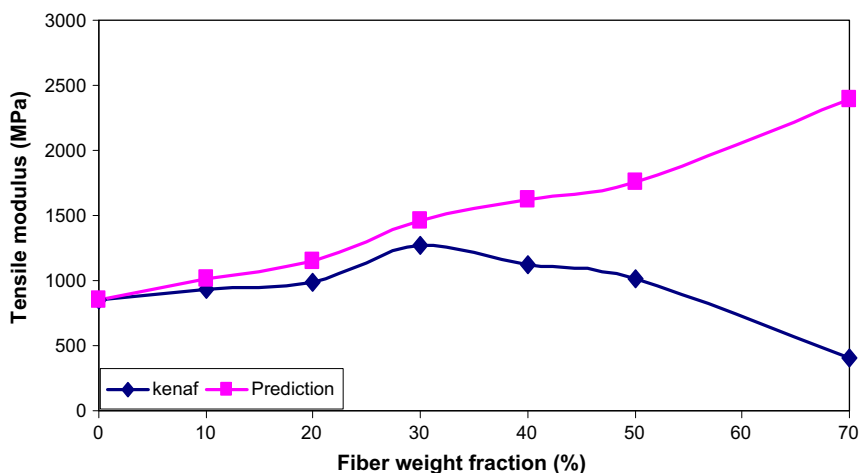


Fig. 43. Predicted and experimental result of tensile modulus of kenaf reinforced PP composites [adapted from 13].

silkworm silk, chicken feather and spider silk have also been used and the trend should go on. Those fibers, both animal- and plant-based have provided useful solutions for new materials development, in the field of material science and engineering. Natural fibers are indeed renewable resources that can be grown and made within a short period of time, in which the supply can be unlimited as compared with traditional glass and carbon fibers

for making advanced composites. However, for some recyclable polymers, their overall energy consumption during collecting, recycling, refining and remoulding processes have to be considered to ensure the damage of the natural cycle would be kept as minimal.

On top of it, Natural fibers are low cost, recyclable, low density and eco-friendly material. Their tensile properties are very good

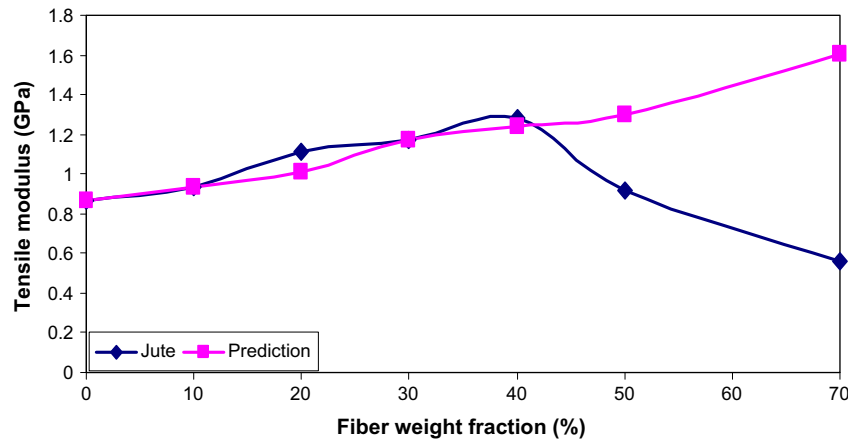


Fig. 44. Predicted and experimental result of tensile modulus of jute reinforced PP composites [adapted from 13].

and can be used to replace the conventional fibers such as glass, carbon in reinforcing plastic materials. A major drawback of using natural fibers as reinforcement in plastics is the incompatibility, resulting in poor adhesion between natural fibers and matrix resins, subsequently lead to low tensile properties. In order to improve fiber–matrix interfacial bonding and enhance tensile properties of the composites, novel processing techniques, chemical and physical modification methods are developed. Also, it is obviously clear that the strength and stiffness of the natural fiber polymer composites is strongly dependent on fiber loading. The tensile strength and modulus increase with increasing fiber weight ratio up to a certain amount. If the fiber weight ratio increases below optimum value, load is distributed to more fibers, which are well bonded with resin matrix resulting in better tensile properties. Further increment in fiber weight ratio has resulted in decreased tensile strength as described in the main text. Mathematical models were also found to be an effective tool to predict the tensile properties of natural fiber reinforced composites.

Finally, it can be found that the main weakness to predict the tensile properties of plant-based natural fiber composites by modelling was giving too optimistic values like results in Figs. 33–37. The modelling has to be improved to allow improvements in the prediction of tensile properties of composites reinforced with both plant- and animal-based fibers.

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