Influence of wood flour on the mechanical properties of birch flour reinforced thermoplastic polyurethane composite

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DOI: https://doi.org/10.47372/uajnas.2015.n2.a20

Abstract

The focus of this study is to characterize the mechanical properties of a composition prepared fromwood flour and thermoplastic Polyurethaneat different ratios. Wood flour was mixed with polyurethane at 30%, 40%, 50%, and 60%. Mechanical properties of the composites have been measured intension, compression, and impact tests. Analyseshave shown an increase in tensile strength with by increasing wood flour content up to 40%, then it decreases when the proportion of wood flourbecomes 50% and 60%. Tensile modulus showed an increasing trend with the increase of fiber content up to 50%, then it drops slightly when the reinforcement ratio becomes60%. Results of compression test showed that 40% reinforcement loading have the peak of compressive strength. High impact resistance was observed with 40% wood flour content. A significantdecrease in impact resistance was observed when the wood flour content ratio exceeded 40%.

Key words: composites; mechanical properties; polyurethane; wood flour; polyol

Introduction

Since 1980s, wood-polymer composites (WPC) have been extensively used in automotive, building products, packaging materials, and other applications[14]. In recent years, wood fibers have gained a significant interest as reinforced material for commercial thermoplastics. They are now fast evolving as a potential alternative to inorganic fillers for various applications. These days, various synthetic polymers are being prepared, combined with various reinforcing fillers, in order to improve the mechanical properties and to obtain the characteristics demanded in actual application [13]. Thermoplastic composites that fortified by Lignocellulosic fillers are favored as a new generation of reinforcing materials.These commodities became popular because of their low cost, biodegradability, ease of processing and the absence of toxic byproducts [36]. However, Cellulosic fibers have some disadvantages, such as lower processing temperature and incompatibility between the hydrophilic fibers and hydrophobic polymers. In addition, moisture absorption of the cellulose fibers has undesirable effects on the properties of wood plastic composites [7]. Synthetic fibers, such as glass and carbonfibers, are brittle and are often broken into smaller fragments during processing. This potentially makes thefracture resistance of the composite poor during processing [30]. In contrast, Lignocellulosic fibers are flexible and will not fracture when processed over sharp curvatures. This enables the fibers to maintain the desired aspect ratio for good performance of the composites[16].Wood porous structure, composed of lignin, cellulose and hemicellulose, is filled with a solid, plastic and fairly hard substance. In principle, WPCs should display superior mechanical properties, dimensional stability, greater resistance to chemical and biological degradation, and less moisture absorption than non-impregnatedwood [12]. Wood is a complex substance that contains cellulose, hemicellulose, lignin and extractives. The quantity of these components varies from one species to another and affects the properties of wood [5]. Mechanical properties of WPC depend on many factors such as a kind of a used polymers, wood species, filler content or a size of a used wood particle [11].The composites properties depend on those of the individual components and on their interfacial compatibility [8]. Several research works have been carried out to identify the parameters that govern the mechanical behavior of particulate composites. Generally, it has been found that the reinforcement effect increases with decreasing particle size and with the increasing of adhesion to

the matrix [19]. Additionally, one observes a reduction in composite strength with increasing filler amount due to poor adhesion between the hydrophilic wood and the hydrophobic polymer [27]. These problems are typically overcome through the use of processing aids and coupling agents [6]. All parameters influencing the characteristics of WPC are related either to their structure, or to the relations between phases. The individual phases influence the resulting characteristics of the material of their own characteristics and by the mutual interaction of the matrix and the filler. And it is the interaction between the fractions that enables us to contribute to materials with new qualities. Strength of interfacial phase is an important criterion in propertiesof WPCs. Due to polarity mismatching of natural fibers and plastics, their interracial boundaries are often weak and lead to undesirable mechanical properties [29]. The mechanical properties of polymers determine its response to an applied stress or strain. These properties manifest in the ability of the material to resist deformation characteristics. Many studies have been carried out to improve the adhesion between two phases, including incorporation of coupling agent into the compound and modification of the mechanical and physical properties of the composites. Bledzki et al. [9], and Maldas et. al. [20], have shown that fiber polymer compatibility can be enhanced by selecting suitable coupling agents. An investigation on the influence of the aspect ratio and interfacial adhesion on the mechanical properties of WPC by Renner [31], has shown the principal applicability of this concept to such composite materials. The influence of the content of chemically modified wood upon the mechanical properties of wood/ thermoplastic polymer composites has been evidenced by Ruxanda et. al. [32]. Han et al. [15], examined the effect of using nanoclay and coupling agent on mechanical and thermal properties of composites obtained from bamboo fibers-heavy polyethylene. The effects of liquefied tropical wood as reinforcing fillers in the thermoplastic polymer composite on the mechanical and physical properties were investigated by Mohd Idrus et. al. [22]. The effects of hydroxyl value of polyols, wood flour particle size, wood flour content, isocyanateindex, and water amount on the compressive property of the foam were investigated by Yuan, and Shi [35]. A number of studies have addressed the influence of wood fibers have on the tensile properties of various thermoplastics [26, 17, 23, 33, 34]. The results are quite variable,depending on the polymer matrix used, the filler type and quantity.

Materials and Method

Basic materials

(i) Polyol:

Polyols are compounds with multiple hydroxyl functional groups available for organic reactions, and they react with isocyanates to make polyurethanes used to make mattresses, foam insulation for appliances and adhesives. Polyol, produced by Shell Company, Germany, was supplied by the National Company for Sponge and Plastic Manufacture, Taiz, Yemen.

(ii) Toluene Diisocyanate (TDI)

Toluene Diisocyanate (TDI) is an organic compound with the formula $H_3C_6H_3(NCO)_2$. Two of the six possible isomers are commercially important: 2, 4- TDI, and 2, 6- TDI. 2, 4- TDI is produced in the pure state, but TDI is often marketed as $\frac{80}{20}$ and $\frac{65}{35}$ mixtures of the 2, 4 and 2, 6 isomers respectively. The isocyanates functional groups in TDI react with a hydroxyl groups to form urethane linkages. 2, 4-Toluene Diisocyanate is primarily used as a chemical intermediate in the production of polyurethane products. It is extremely toxic from acute and chronic exposures. TDI is produced by DOW Company, Germany, and supplied by the National Company for Sponge and Plastic Manufactures, Taiz, Yemen**.**

(iii) Stannous-2-ethylhexoate (T9)

Stannous Octoate Catalyst (T9) is primarily used to promote crosslinking in the production of rigid, semi-rigid, and flexible polyurethane foams, and as a catalyst in the production of polyurethane, silicone, and other polymers. It is considered insoluble in water, toxic to aquatic

organisms, and may cause long-term adverse effects in the aquatic environment. The chemical formula of (T9) is:

$$Sn\ [OCO\ CH\ (C_2H_5)\ (CH_2)_3CH_3]_2$$

Polyurethanes

Polyurethanes are formed by reacting a polyol (an alcohol with more than two reactive hydroxyl groups per molecule) with a diisocyanate or a polymeric isocyanate in the presence of suitable catalysts and additives. Because of the a variety of diisocyanates and the wide range of polyols that can be used to produce polyurethane, a broad spectrum of materials can be produced to meet the needs of specific applications**.**

Additive materials

Birch flour was used as raw materials for obtaining fillers. Wood flour was supplied by a local workshop. The wood flour particles of 425 microns (~ 40-mesh) in size were procured from local workshops, as shows in Figure (1). The wood flour was dried before manufacturing in an oven for 24 h at 100℃ in order to remove moisture, The dried wood flour was stored in a sealed plastic container to prevent the absorption of water vapor.

Figure (1): Wood flour used in the study

Mixing

Dried wood flour and polyethylene powder was mixed based on their weight ratios. A mechanical stirrer, purchased from Rayder Company (China), was used to mix the fibers and polyol in the foaming process. The power of the stirrer was 600 W, witha rotational speed of 3400 RPM.

Foaming Process

Catalyst (T9) was added to the polyol and blended for one minute, by using a mechanical stirrer. Birch flour was added to the previous mixture, then mixed from 5 to 10 minutes, depending on the amount of flour (until homogeneity is obtained). Polymeric material (TDI) was adds to the polyol and flour, then mixed for 5 to 10 minutes until homogeneity was reached.

Molding

The Casting mold of steel was used with rectangular in shape with dimensions of ($24 \times 3 \times 2$) cm. It was manufactured in local workshops; in the form of pistonopensides, so that it can close one of its ends by plate of steel, which fasten with eight screws. The Casting molds were covered from inside with a layer of polyethylene (PE) to prevent the adhesion of the material with the internal walls of the mold. The material that has been previously prepared inside the casting mold was evacuated and leveled by hand, then the upper segment of the template was placed. The composite bars were produced using a hot press, compression molding by a hydraulic piston of 0.9 bars. Pressing temperature was 160ºC and pressing time was from 5 to 10 minutes. The sampler was left in the mold for two hours to complete the reaction.Then the sample was taken out of the mold and left to dry without any treatment, to get a sample of dimensions (34 × 12) cm of wood plastic material.

Specimens preparation

Dried wood flour and Polyurethane was mixed, based on their weight ratios. Once the ingredients of each composite formulation were weighed to a (0.01g) precision, they were mixed by hand lay up process and again kept in plastic bags before the compression molding process. Four different types of specimens have been fabricated with four different weight ratios, as shows in Table (1), and Figure (2). The composites were then cut into dumbbell-shape, and two bars-

shapes by using special molds to produce the samples for tensile strength, compression, and impact specimens

Table (1): Composition of the wood polymer composites

Specimen Code	Mesh Size	Wood Flour (%)	Polyurethane (%)
A	40	30	70
B	40	40	60
C	40	50	50
D	40	60	40

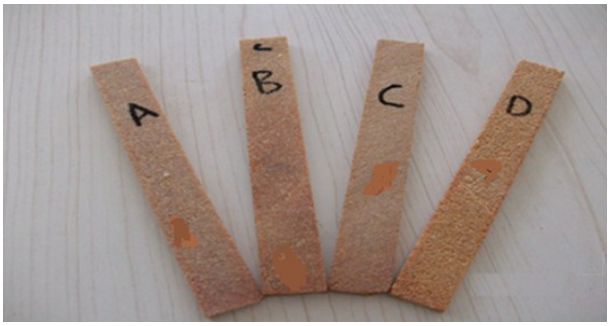

Figure (2): Different types of specimens fabricated with four different weight ratio

Mechanical testing

In order to evaluate the effect of flour content on the mechanical properties of the composite, the testing of the tensile,compressive strength, and impact strength was performed. Ten samples for each group were cut from the manufactured composites**.**

Tensile strength

Vertical tensile specimens resistance was measured according to the American Standard (ASTM - D638)[4]. For the purpose of measurement, the measuring machine type (Zwick / Roell), model Z005, and with a range of (5000) Newton was used. To measure the tensile strength, the standard dimensions of the samples were used, as shown in Figure (3).

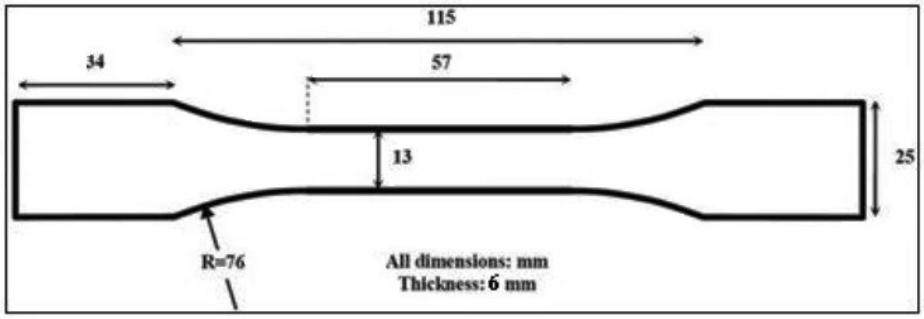

Figure (3): Standard dimention of tensile test according to ASTM D638

Compressive strength

The goal of a compression test is to determine the behavior or response of a material while it experiences a compressive load by measuring fundamental variables, such as strain, stress, and deformation. Compressive strength of wood polymer composites in longitudinal direction was measured according to the American Standard (ASTM - D695)[2]. Measuring machine type (TREBEL) and with a range of (5000) Newton was used. For this testing, the block compression specimens were cut with an electric saw into standard dimensions. The typical blocks are 12.7 x 12.7 x 25.4mm.

Impact resistance

Impact test is used to determine the amount of impact energy required to break the specimen. The most common impact tests (Charpy and Izod) employ a swinging pendulum to strike the bar; heights before and after impact are used to compute the energy required to fracture the bar and, consequently, the bar's impact strength. For the impact resistance, an impact test was carried out using an impact pendulum test machine type (Zwick / Roell), model PSW-750 up to 750 joules. An un-notched Izod Impact test is conducted to study the impact energy according to (ASTM-D256)[3]. An un-notched specimen with dimensions $(5 \times 1 \times 0.5)cm$ (length ×width × thickness) were cut and kept in a cantilever position.Ten replicates were run for each test.

Results and Discussion

Tension Test

Tensile strength was made at room temperature and humidity (75 RH), and with tensile velocity of (5mm / min).The results of the analysis of the ultimate tensile strength, elongation to break, and 0.2% offset yield stress of the tested WPCs specimens are given in Table(2).

Table (2): Tensile properties wood flour reinforced thermoplastic polyurethane

Specimen Code	Wood flour (%)	Ultimate tensile strength (MPa)	Elongation to break (%)	Modulus of elasticity (GPa)	0.2% offset yield stress (MPa)
A	30	10.19	6.11	26.9	3.18
B	40	15.27	5.52	40	5
C	50	10.62	4.70	57.64	3.68
D	60	5.95	2.08	44.54	2.3

Figures (4) a, b, c, and d presents the stress–strain behavior of different samples under the influence of tensile test.

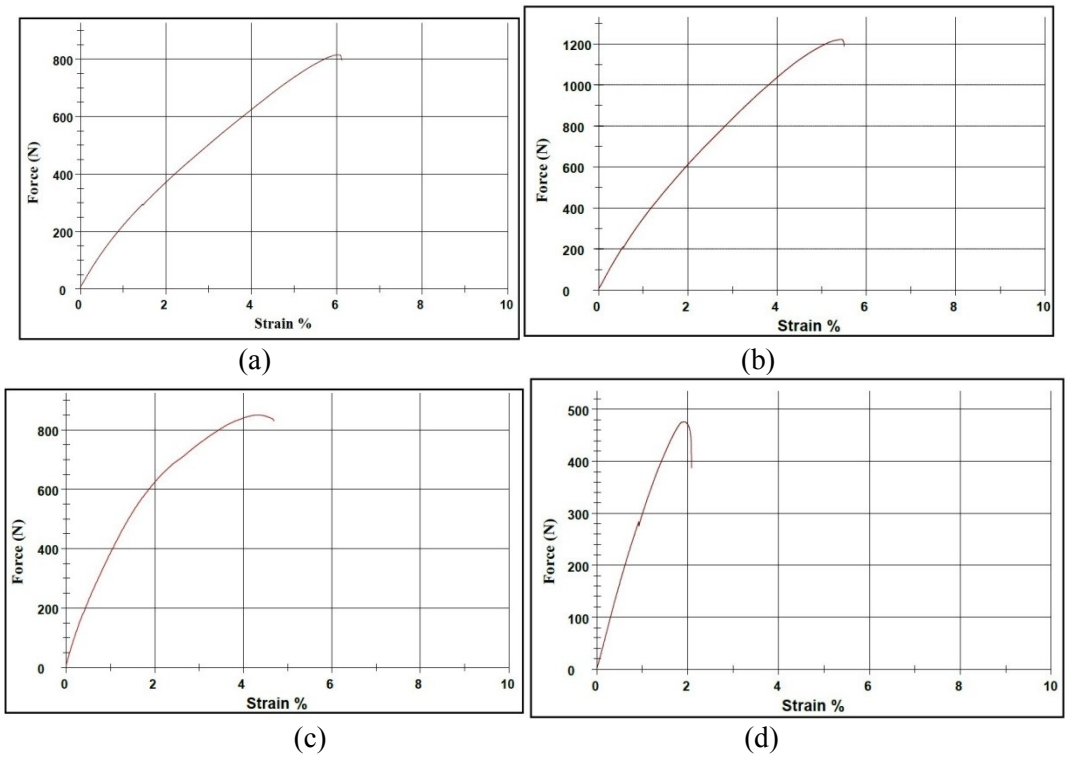

Figure (4): Stress-Strain behavior of the different wood flour content of wood polyurethane composites. (a) 30%. (b) 40%. (c) 50%. (d). 60%

Figure (5) shows the effect of wood flour content on tensile strength of wood polymer composites. It can be observed that the tensile strength increases nonlinearly with the flour content.

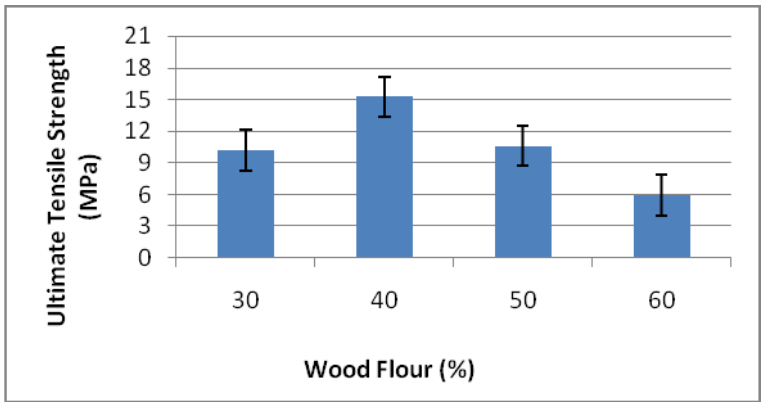

Figure (5): Shows the effect of wood flour content on tensile strength of wood polymer composites

The tensile strength of the WPCs was increased with the increasing of wood flour content up to (40%) but further increment in the filler content decreased the tensile strength. The tensile strength was increased from $(10.19\ MPa)$ for (30%) flour content to $(15.27\ MPa)$ for (40%) content. Tensile strength, then was decreased to $(10.62\ MPa)$ and $(5.95\ MPa)$ for (50%), and (60%) flour content, respectively. Several factors affect the strength of composites, such as, poor dispersion of the fibers in the matrix,strengths of fibers and matrix, fiber content, the interfacial bondingbetween fibers and matrix, and moisture pick-up[10].The reduction in the tensile strength of the WPCs beyond (40%) flour was mainly attributed to the poor compatibility between polar flour and nonpolar polyurethane, which formed the weak interfacial regions. The weak interfacial regions resulted in the reduction in the efficiency of stress transfer from the polymer matrix to the reinforcement component. Similar observations have been reported byAlperen Kaymakci and Nadir Ayrilmis[1].

On the other hand, it can be clearly observed that the elongation, at break, decreased steadily with the wood–fiber content, as shown in Figure (6). The steep decline in elongation immediately on filler addition is obvious, because the wood–fibers have low elongation at break and restrict the polymer molecules flowing past one another. When the percentage of flour filler increased, the ductility of the polyurethane wood flour composites was greatly decreased. This demonstrates that the filler had hardened the composites and reduced their ductility. These results were consistent with the results ofMay et.al.[21], and Nadir et.al.[24].

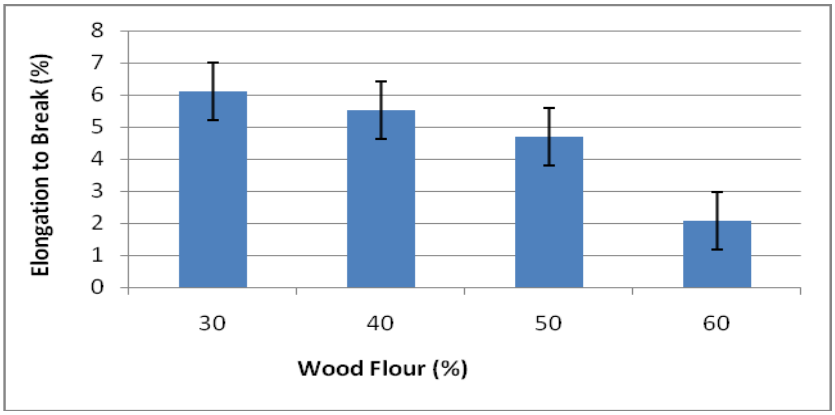

Figure (6): The effect of wood flour content on the elongation to break of wood plymer composites

Figure (7) shows the Modulus of elasticity (tensile modulus) of the composites. It can be clearly observed that the increasing of the wood flour percentage increases the tensile modulus. The addition of wood flour increased the tensile modulus of the composites; this is because the wood flour is stiffer than polymer.

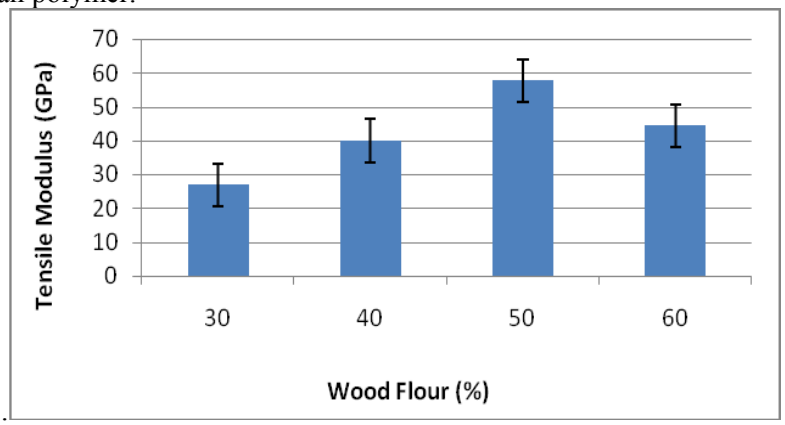

Figure (7): The effect of wood flour content on tensile modulus of wood polymer composites

The maximum tensile modulus was found at 50% wood flour loading. Then, the tensile modulus decreased when the wood flour content reached 60%. One might also expect to obtain a greater tensile modulus as the wood flour content was increased because the flour usually retains most of its lignin[25]. However, The issue here is different.

The tensile modulus was found to reduce when the flour ratio increases to more than 50%. This decline could be due to two reasons, one being a poor dispersion of the flour particles throughout the polyurethane matrix, and the other the moisture pick-up in the flour. The former reason explained that the flour fibers tended to cling together, due to strong interferehydrogen bonding, and resisted dispersion of the individual fibers as the fiber content was increased. In the latter case, since the flour used in this work was hydrophilicin nature and was chemically untreated, the flour may have picked up moisture during storage, processingand testing.

Compressive strength

This is a mechanical test to measure how easily a matter can undergo deformation under stress. Compressive strength was made at room temperature and humidity (75 RH), and with compressive velocity of (10mm / min). The effect of increasing flour content on the compressive strength of the wood polymer composites is listed in Table (3).

Figure (8) shows the relationship between compressive strength and wood flour content in wood polymer composite samples. Compressive strength was found to increase significantly as the wood flour content increased up to 40%, which is found to be the maximum. A significant decrease in the compressive strength was observed when the wood flour content increased to more than 40%.

Table (3-2): The effect of wood flour content on the compressive sterngthof wood polymer composites

Compressive Strength (MPa)	Strain	Stress (MPa)	Local force at ends (N)	Specimen Code
29.74	0.2233	6.64	664	A
42.52	0.2166	9.21	921	B
35.75	0.1933	6.91	691	C
19.19	0.16	3.07	307	D

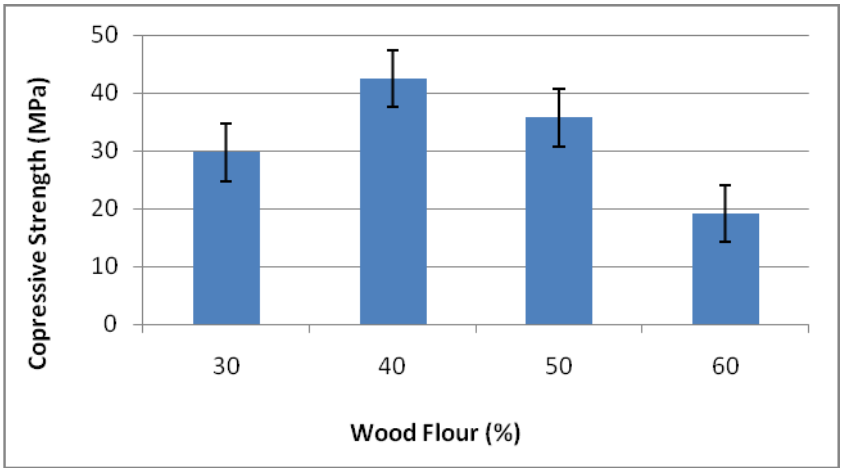

Figure (8): Relationship between wood flour content and compressive strength

The decrease in the compressive strength after the optimum are attributed to different reasons. First, it may refer to weak interfacial bonding between wood and the polymer, due to formation of agglomeration of wood flour which causes the reduction of adhesion in the composite interface. Secondly, it was found out that a linear relationship between the density and the compressive strength. However, as the wood flour concentration increases, the density of wood polymer composites became lower.

On the other hand, the poordispersion of the wood flour in the matrix increases the interface defects or depending between polymer and flour content which reduces the ductile portion of the composite, thus decreasing the composite compressive strength. The result is in good agreement with Homkhiew et.al.[18].

Impact Resistance

One of the most critical properties of composite materials is the residual strength after impact: How much degradation has resulted from a tool strike, a collision, or some sort of impact. Figure (9) illustrates the effect of wood flour content on impact resistance of wood polymer composite.
Reports in the literature indicate that impact resistance often decreases as an effect of wood reinforcement, similarly to many particulate filled composites. From the figure, the ultimate value of impact resistance was found in 40% of wood flour content. A significant drop in the impact resistance was recorded when wood flour content increased to 50%, and 60%.

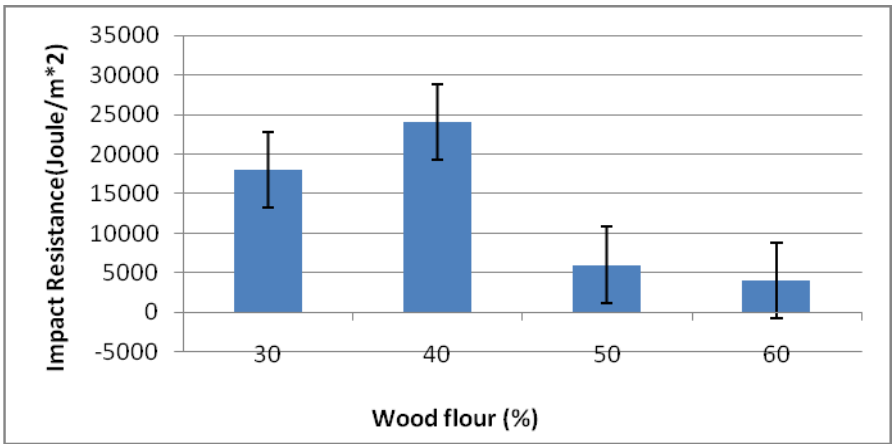

Figure (9): The effect of wood flour content on impact resistance of wood polymer composites

The poor interfacial bonding between wood flour and the matrix polymer causes microcracks to occur at the point of impact, which cause the crack to easily propagate in the composite without any coupling agent. These microcracks cause a decreased impact resistance of the composites. It may be stated that wood flour filler does not provide the energy dissipation mechanism in polyurethane necessary for impact toughening. On the other hand, the addition of wood flour into polymer matrices reduces the ductile behavior of the matrix by making the composites more brittle.The higher degree of brittleness introduced by the addition of wood flour into the polymer matrix causes this decrease in impact resistance. However, the increase of wood flour fraction causes lack of energy absorbent, which is usually observed when combining short natural fibers with thermoplastics. Nurshamila et. al. [28], observed a similar trend with wood fiber reinforced polypropylene composites.

Strain of the composite also plays a major role in the impact resistance. It is noticed from Figure(10) that strain is decreasing with the increase of wood flour content. Thus, the impact resistance is also decreased with the increase of wood flour content.

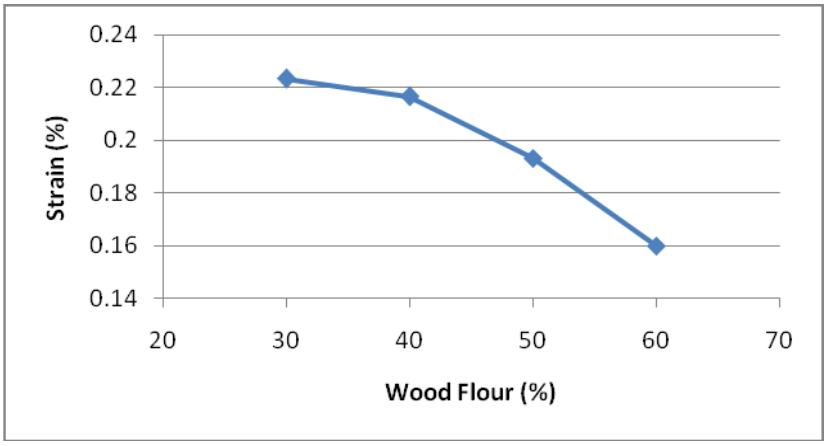

Figure (10): The effect of wood flour content on the strain of wood polymer composites

Conclusions

This study examined the effect of wood flour as reinforcing agent on the mechanical properties of wood thermoplastic polyurethane. From the above presented results and discussion, the following specific conclusions can be drawn:

1-In general, the mechanical properties of the tested wood polymer composites substantially depend on wood flour content. In tension test, composites have shown lower tensile strength when the proportion of wood flour exceeds 40%, which have the maximum tensile strength. Tensile modulus is increased with the increase in wood flour content up to 50%, but then droped when wood flour content becomes 60%. The tensile elongation at break values is declined sharply with the addition of the wood flour.

2- The compressive strength is observed to increase with the increase in wood flour content up to 40%.

3-Impact resistance of wood flour reinforced polyurethane of 40% content shows the maximum impact energy absorption. However,impact resistance is decreased by increasing wood flour content more than 40%.

4- Finally, The best of mechanical properties for wood flour conent reinforced polyurathene stands between (40 to 50 %). Therefore, 40% by weight of wood flour is the overall equilibrium point for mechanical efficiency of the wood polymer composites compounded.

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دراسة تأثير إضافة نشارة الخشب الناعمة على الخصائص الميكانيكية لمتراكبات الخشب- البولي يوريثان اللدن

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DOI: https://doi.org/10.47372/uajnas.2015.n2.a20

الملخص

يركز هذا البحث على دراسة الخواص الميكانيكية للمتراكبات المحضرة من نشارة الخشب الناعمة كمضاف معزز للبولي يوريثين اللدن وبنسب وزنية مختلفة. صنعت عينات من المتراكب بنسب وزنية من نشارة الخشب الناعمة 30٪، 40٪، 50٪، و60٪. تم قياس الخواص الميكانيكية للمركبات في اختبارات متانة الشد والضغط ومقاومة الصدم. أظهرت النتائج زيادة في متانة الشد مع زيادة نسبة نشارة الخشب الناعمة إلى حين الوصول إلى نسبة 40٪، ثم تنخفض عندما تصبح نسبة نشارة الخشب 50٪ و60٪ . لقد أوعز هذا الانخفاض في قيمة متانة الشد إلى ضعف انتشار نشارة الخشب داخل البوليمر، بالإضافة إلى قلة التوافق بين نشارة الخشب القطبية والبوليمر غير القطبي. أظهرت نتائج اختبار متانة الضغط أن أعلى قيمة للمتانة كانت عند نسبة تعزيز مقدارها 40%، ثم ينخفض عند زيادة نسبة نشارة الخشب. أنَّ سبب هذا الانخفاض يعود إلى ضعف الترابط الداخلي بين الخشب والبوليمر بسبب تكتل نشارة الخشب وعدم انتظام انتشارها مما يؤدي إلى ضعف التصاقها بالبوليمر. بينت نتائج فحص مقاومة الصدم أنّ أعلى قيمة للمقاومة كانت عند نسبة تعزيز مقدارها 40%، ثم انخفضت عند زيادة نسبة نشارة الخشب. تم تعليل هذا الانخفاض إلى وجود التشققات الدقيقة داخل المتراكب بسبب ضعف الترابط بين الخشب والبوليمر مما يؤدي إلى انتشار هذه التشققات الدقيقة وتسببها في إضعاف مقاومة الصدم.

الكلمات المفتاحية: البولي يوريثين، الخصائص الميكانيكية، نشارة الخشب، متانة الشد، مقاومة الصدم .