Effect of birch wood flour on the density, water absorption, thickness swelling, and thermal conductivity of wood thermoplastic polyurethane composites

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Abstract

In this study, some of the important properties of experimentally manufactured wood thermoplastic polyurethane composites were determined. Specimen having 30%, 40%, 50%, and 60% wood flour were mixed with rigid polyurethane foam. Physicalproperties (density, water absorption, and thickness swelling) and thermal conductivity of wood–plasticcomposites (WPC) were investigated. Results indicated that the density of WPCs decreased by increasing wood flour content. The results also revealed that water absorption and thickness swelling of the composites increases with increasing the percentage of wood flour content. It was found thatthermalconductivity of the composites slightly increased with the increasing of wood flour content. Experimental results of thermal conductivity were compared with a theoretical model (Maxwell – Garnett model), the errors associated with the above model, with respect to experimental ones, varies between 40% to 50%.

Key words: Wood polymer composite, Polyurethane;Water absorption, Thickness swelling, Thermal Conductivity.

Introduction

Composite is one of the most advanced and adaptable engineering materials known to men. The processing of plastic composites, using naturalfibers as reinforcement, has increased dramatically inrecent years. A more recent development in compositetechnologies is wood/plastic composites (WPCs).WPCs are a relatively new generation of composite materials and also the mostpromising sector in the field of both composite andplastic industries. Their properties and environmentaladvantages have made them a good choice for manyapplications[8].WPCs are made by combining wood and thermoplastic polymer, which results in a composite that combines the best properties of both components[23]. WPCs are used for many applications, and a largeproportion (about two-thirds) is used for outdoor applications,e.g., decking, railing, fencing, and exterior coveringapplications such as siding and trim [19].The new WPCs products are being designed for applicationswhere long-term performance, consistent appearance,and dimensional stability are important [21].

Wood possesses porous structures consisting of various cell walls, which are mainlycomposed of biopolymers, i.e., carbohydrate polymers of cellulose and hemicelluloses andphenolic polymers of lignin [25]. The cellular structure of wood endows it with high strength-to weightratio. The use of wood particles as filler in the plastic matrix can result in a stiffer and reduced cost for theWPCs.Although the hydrophobic thermoplastic matrix slows the uptake of water and moisture absorption, it remains the important concerns for WPCs [7]. Wood filler is primarily responsible for thickness swelling(TS) and water absorption (WA) of the WPCs [6]. Water absorption and thickness swelling are the most important physical characteristics of wood plastic composites (WPCs) exposed to environmental conditions and, thus, determining their end use applications[4]. Water absorption of WPCs is a significant factor affecting the properties in the final applications [13].The increased moisture content of the compositesreduces their mechanical properties and dimensional stability [2]. WPCs have a thermoplastic-rich surface layer that iscreated during their

processing (through extrusion, compression molding, or injection molding) that produces highlevels of water repellency[5].

It is well known that wood undergoes changes in dimension with changes in moisturecontent. An increase in moisture content causes swelling, and decrease in moisturecontent causes shrinkage The internal stress induced in the composites, during hot pressing, will result in greater thickness swelling of wood composites, compared to normal wood, after exposure to moisture [21]. Thickness swelling of wood composites is a serious and complex phenomenonbecause it can have a deleterious effect on other mechanicaland physical properties of the board [11].

Inrecent days, woodfiber composite materials are widely usedin building components to reduce heat transfer in airconditioned buildings in order to decrease energyconsumption. This is due to the fact that wood has excellent heat insulation properties. The thermal conductivity of a composite depends upon thethermal conductive nature of the fiber, matrix properties aswell as their volume fractions, sizes, shapes, thickness,orientations and perfect bonding between the constituents [18]. Effective thermal conductivity is an important characteristic of heat transfer properties of materials. The temperature field in composite materials cannot be determined unless the thermal conductivities of the media are known [19].

The polyurethanes (PU) foams are widely used as insulating and core materials for furniture, cooling and freezing systems, in housebuilding, shipbuilding etc. The use of rigid foams is resulted from their low heat conduction coefficient, low density, low water absorption, relatively good mechanical strength [23]Although the dosage of a polyurethane cross-linker accounts for only approximately 5% of wood adhesives, its application amount has been quite considerable [25]. The uses of PU adhesives have expanded to include bonding of numerous substrates such as glass, wood, plastics, and ceramics. PU adhesives are sold into an ever-widening array of markets, where they are known for their adhesion, flexibility, low-temperature performance, high cohesive strength, and cure speeds that can be readily tailored to the manufacturer's demands [17].

Experimental work

Rigid polyurethane foamPreparation

The production of rigid polyurethane foam requires two main liquid components; polyoland diisocyanate, in addition to blowing agent. The blowing agent (like water) is usually added to the polyol, together with further auxiliary components such as activators. Polyols are reactive substances, usually liquids, containing at least two isocyanate-reacting groups attached to a single molecule. A large variety of polyols are offered, but most of the polyols used fall into two major categories: hydroxyl-terminated or amino-terminated polyols. Diisocyanate, which are characterized by a (NCO) group, which are highly reactive alcohols. The most widely used isocyanates employed in polyurethane production are toluene diisocyanate (TDI). TDI is produced by chemically adding nitrogen groups on toluene, reacting these with hydrogen to produce a diamine, and separating the undesired isomers.

Polyol

Polyol(CARADOL MD250-10), Shell Chimecals, Germany, was supplied by the National Company for Sponge and Plastic Manufacture, Taiz, Yemen**.** Its viscosity at 25°C is $(300\ mPa.s)$. with hydroxyl value of 250 mg KOH/g.

Toluene Diisocyanate (TDI)

(DURAMOUL 5223 Isocyanates), Dow Chemical Company. (TDI) iscomposed of a mixture of the 2,4 and 2,6 isomers of toluene diisocyanate in a ratio of 80% to 20%. Supplied by the National Company for Sponge and Plastic Manufacture, Taiz, Yemen with NCO content 31%, Its viscosity (at 25°C) is $(5000-9000\ mPa.s)$.

Foaming Process

For foam production, polyoland small amount of distilled water (as blowing agent) were mixed together. After that, TDI was added to the mixture.Themixture was stirred with an electric stirrer to ensuregood dispersion of reagents anda foam ofdesirable cell structure. The reaction starts after a

short period of time andprogresses with heat development. The reactionmix is continually expanded by the blowing gasesreleased, until the reaction product reaches thesolid state as a result of progressive cross linkage,the foam structure being retained. The foam samples produced wereleft to stand for 48 hoursfor full curing under ambient temperature.

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. The wood flour is 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.

Mixing

Dried wood flour and polyethylene foam was mixed based on their weight ratios. A mechanical stirrer was used to mix the fibers and polyol in the foaming process. The power of the stirrer was 600 W, with a rotational speed of 3400 RPM.

Molding

The Casting mold of steel, with a rectangular in shape and dimensions of ($24 \times 3 \times 2$) cm, was used. 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. Evacuate the material that has been previously prepared inside the casting mold, and leveled by hand, then the upper segment of the template is 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 from 5 to 10 minutes. Sampler left in the mold for two hours to complete the reaction. Then the sample out of the mold, and leave to dry without any treatment, to get a sample of dimensions (34 × 12) cm of wood plastic material.

Specimens preparation

Dried wood flour and Polyurethanewas mixed based on their weight ratios. Once the ingredients of each composite formulation were weighed to a (0,01 g) 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 ratio, as shown in Table (1).

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

Physical properties

Specific gravity

The test method covers the determination of the bulk density and specific gravity of the materials in their form, as manufactured, according to Specific Gravity (ASTM D792). Therefore, this test method evaluates a product, not an inherent material property. The test consists of weighing the specimen in air, then immersing it in distilled water at 25°C using a sinker and wire to hold the specimen completely submerged as required, determining a weight of the specimen in water upon immersion, and calculating its bulk specific gravity.

The Specific Gravity of the sample is calculated as follows:

$$SG = \frac{a}{a + w - b}$$

Where: a = apparent mass of specimen, without wire or sinker, in air, b = apparent mass of specimen (and of sinker, if used) completely immersed and of the wire partially immersedin liquid, and w = apparent mass of totally immersed sinker (if used) andof partially immersed wire.

Water Absorption and Thickness Swelling

Early in the history of WPC lumber, it was often suggested that WPCs were resistant to moisture because the wood was completely encapsulated by the plastic. While plastic imparts some resistance to moisture uptake, once moisture enters the matrix, the damage begins. The belief that cellulose fibers or wood particles in composites are encapsulated with plastic is not completely valid [3]. Particularly, it is not valid when composites contain a significant amount of fillers, such as above 40%.

Water uptake tests were carried out according to theASTM D1037-06A, Section 23, method B. The test specimens prepared using the full cross section of the as-manufactured product. Specimens with a dimension of $(152 \times 152\ mm) \times$ the thickness of thematerialwas cut with all four edges smoothly and squarely trimmed. To ensure the constant weight and moisture content for the specimens before each test, all the specimens were put in a conditioning chamber maintained at a relative humidity of $(65 \mp 5\%)$ and a temperature of $(25 \mp 3°C)$.

After conditioning, the weight of the specimenwas measured to an accuracy of $(\mp 0.2\ \%)$. The width, length, andthickness of the specimen was measured to an accuracy of $(\mp 0.3\ \%)$ to compute the volume of the specimen. The specimens then submerged horizontally under (25 mm) of potable water maintained at a temperature of $(25 \mp 1°C)$. Fresh water should be used for each test. After 24 hour submersion,thespecimen suspends to drain for $(10 \mp 2\ min)$. After submersion, the specimens were dried in an oven at $(103 \mp 2°C)$, then the specimen was immediately weighed and the thickness was determined. The moisture content of the specimens, before and after submersion, was calculated based on oven-dry weight.

The values of the water absorption in percentage were calculated according to the equation:

$$WA(t) = \frac{W(t) - W_o}{W_o} \times 100$$

Where $WA(t)$ is the water absorption at time t, W_o Is the oven dried weight and $W(t)$ is the weight of specimen at a given immersion time t.

The values of the thickness swelling in percentage were calculated using the equation:

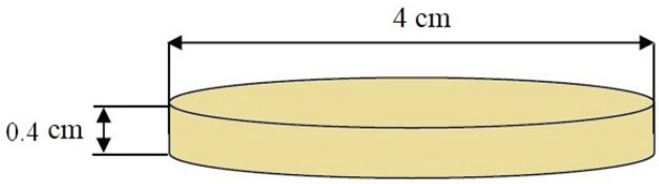
$$TS(t) = \frac{T(t) - T_o}{T_o} \times 100$$

Where $TS(t)$ is the thickness swelling at time t, T_o is the initial thickness of specimens and $T(t)$ is the thickness at time t.

Thermal Conductivity

Samples preparation

The composite samples have been prepared by using hand-lay-up technique to measure the thermal conductivity of wood flour with average size 425 microns reinforced in thermos plastic polyurethane. A mold of 40 cm diameter and 0.4 cm thickness was made from a stainless steel sheet. It was coated with wax an as releasing agent for easy removal of the sample. The composite was cured under a load of about 40kg for 24 hours before it was removed from the mold. Cylindrical specimens, according to the standard specification of the Lee's disk instrument, with dimension of $(4 \times 0.4)\ cm$ [diameter× thickness], was fabricated, as shows in Figure (1).



Fhgure (1): Standard dimensions of specimen of thermal conductivity test

Experimental Setup

The apparatus used was a modification of the standard Lee's disk method for the measurement of thermal conductivity. Schematic diagram of the apparatus is represented in Figure (2). This consists of three copper plates (A–C) drilled to accept liquid-in-glass thermometers and a 6 W electrical plate heater of the same diameter as thecopper plates. The sample to be studied is approximately at the same diameter as the copper discs A & B, which are 4cm. The heat is supplied from the heating coil inserted between disc B and C.The specimen was then placed between copper plates A and B. The heater was sandwiched between plates B and C and, after tightening the clamp screw to hold all the discs together, the power to the heater was switched on.

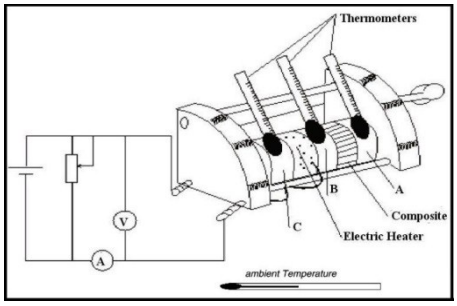


Figure (2): Schamtic diagram of Lee's disk apparatus

The temperature of each disk independently rising from the other discs, and at different rates with the time, according to its position from the heat source.When the Voltage and Current becomes steady, heating process began until the temperatures of B and C stop fluctuating, which means that the heat flowing through the insulator each second is the same as the heat lost by C each second. We assume that there is no heat loss out of the sides of the sample through covering the device with glass wool. At the beginning of each determination, the power from astabilized DC supply was turned on full until the average temperature of the sample reached the desired value. The power was then adjusted to allow the temperatures of the plates to stabilize. As equilibrium was reached, readings were taken every 10–15 min. When the temperature of all parts of the apparatus had been stable to with in $(\mp 1°C)$ for 30 min, a value for the thermal conductivity of the specimen (k) of thickness (d) and radius (r) was calculated, using the following equations [Oluyamoand Adekoya 2015]:

$$k = \frac{ed}{2\pi r^2}\left[a_s\left(\frac{T_A + T_B}{2}\right) + 2a_A T_A\right]$$

Where e is the loss of heat through the unit time (second) and through the area (m^2), which is given by:

$$e = \frac{IV}{a_A T_a + a_s\left(\frac{T_A + T_B}{2}\right) + a_B T_B + a_C T_C}$$

Where a_A , a_B , a_C and a_s are the exposed surface areas of discs A, B, C and the wood polymer composite sample respectively. T_A , T_B and T_C are the temperatures of the discs A, B and C above ambient. V is the potential difference across the heater and I is the current which flows through it.

Results and Discussion

Specific Gravity

The results of the specific gravity of wood polyurethane composites are presented in Table (2).

Figure (3) shows the effect of wood flour content on the specific gravity of wood polyurethane composites. The figure shows that the specific gravity decreases with increasing the proportion of wood flour content. This reduction may be due to moisture content. Polymers, which are used in composite materials, practically do not absorb water (hundredth or thousandth percent by weight) [3].

Table (2): Values of density of different composites

Specimen Code	Wood Flour(%)	Density $\left(\frac{kg}{m^3}\right)$	Specific gravity (Experimental)	Specific gravity (Theoretical)
A	30	956.71	0.959	1.009
B	40	876.90	0.879	0.906
C	50	810.06	0.812	0.845
D	60	752.20	0.754	0.767

Incorporation of wood flour into polymer significantly increases moisture uptake. Wood flour content, wood particle size, processing method, and additives influence the amount of water absorbed by the WPC [15]. Moisture sorption in wood is complex and the final equilibrium moisture content is affected by temperature and humidity [14].

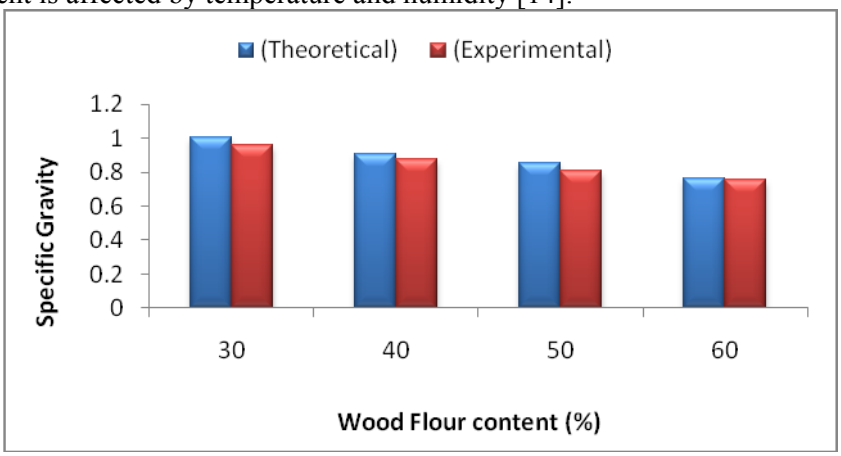


Figure (3): Specific Gravity of wood flour polyurethane composites

Though moisture could potentially be used as a foaming agent to reduce density, this approach is difficult to control and is not common industrial practice [16]. Depending on ambient conditions, wood flour can absorb several weight percents of moisture within hours. On the other hand, moisture in wood flour is converted to steam when the composite is subjected to a hot press while processing, which adds a micro-bubbling to the composites. Steammake the material foamed, with non-controlled porosity. This noticeably decreases the density of the final WPC product.

Water Absorption and Thickness Swelling

Figure (4) illustrates the effect of wood flour content on the water absorption of the WPCs. From the figure, it is clear that water absorption, increased slightly with wood flour content up to 50%.

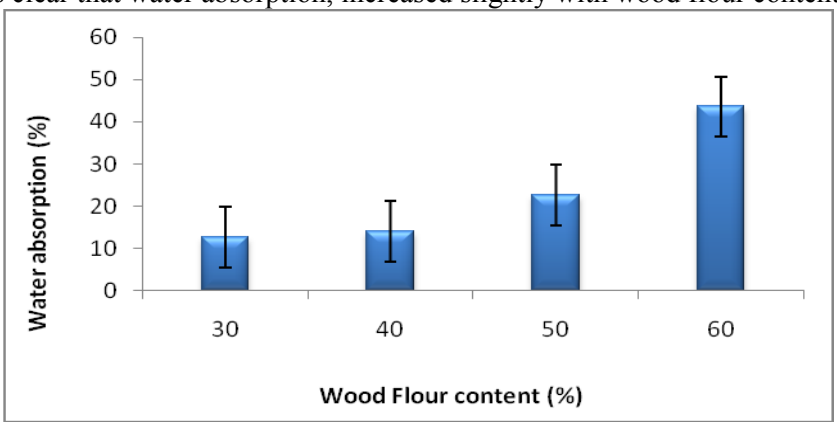


Figure (4): Water absorption of wood flour polyurethane composites

A significant increase of water absorption was observed when the wood flour content became 60%, (with increments of about 92% from 50% to 60% wood flour content). These results mainly attributed to the hydrophilic nature of wood. Wood is a hydrophilic porous composite which consists of cellulose, lignin, and hemicelluloses polymer that are rich in functional groups, such as hydroxyls, which are mostly responsible for the high water absorption. And due to this reason, the WPCs have the potentialityto uptake water under humid condition. Similar results for increasing pattern of water absorption were also reported by [8], which study the effects of wood properties on the behaviors of wood particle reinforced polymer matrix composites. In composites with higher wood flour contents, water absorption, increased more rapidly. Because higher wood content will causestresses within the material and create voids and microcracks, due to the poor interfacial bonding between the wood flour and the polymer matrix. When the composites have been immersed in water, thecapillary action conducts the water molecules in the material and fills in the voids andcracks in the composites [1].

Thickness swelling diagram of different wood flour content is illustrated in Figure (5). Thickness swelling of the wood flour polyurethane composites has a similar trend as the water absorption, where composites with high water absorption also showed higher thickness swelling. As it is clearly seen, thickness swelling increases with increasing wood flour content. By increasing the wood flour content from 40% to 50%, and 60% there was a significant increasing in thickness swelling of the composite.

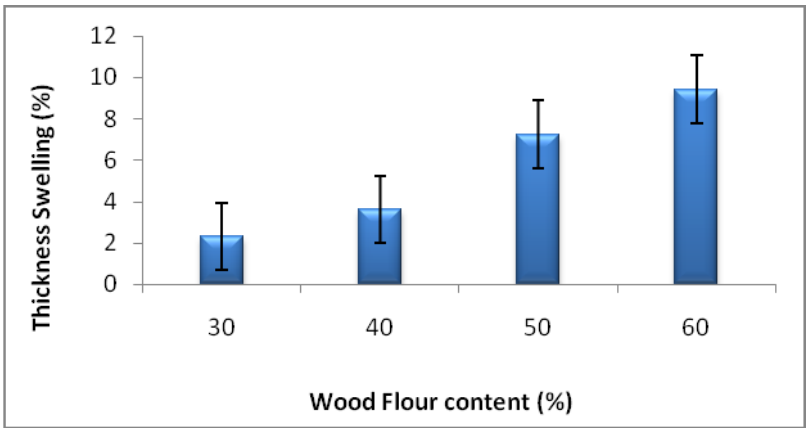


Figure (5): Thickness Swelling of wood flour polyurethane composites

Natural fiber-based polymer composites are poorly water resistant due to the presence of polar groups, which attract water molecules through hydrogen bonding. The hydrophilic nature of wood flour causes the thickness swelling in wood plastic composites manufactured. It is well established that the thickness swelling in natural fiber thermoplastic composites is mainly due to the presence of hydrogen bonding sites in the natural fibers [4]. Cellulose and hemicelluloses are mostly responsible for the high water absorption of natural fibers, since they contain numerous accessible hydroxyl groups. As cellulose fiber is the main component in the wood flour, the absorbedwater mostly resides in the regions such as the flour lumens, the cell wall, and the gaps at the interface between the wood flour and the polymer matrix. As the wood flour loading increased, the cellulose content increased which in turn resulted in the absorption of more water and then increased the thickness swelling.

Thermal Conductivity

The effect of wood flour fraction on the thermal conductivity of WPCs is shown in Figure (6). The thermal conductivity of a composite depends upon the thermal conductive nature of the fiber, matrix properties as well as their volume fractions, sizes, shapes, thickness, orientations and perfect bonding between the constituents.

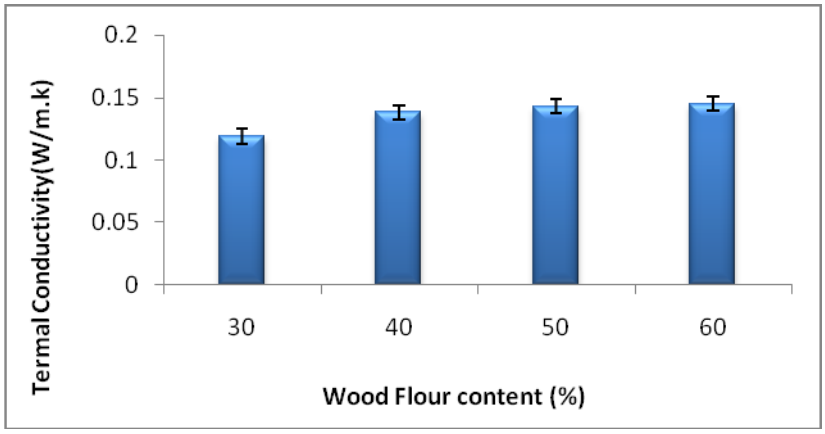


Figure (6): Thermnal conductivity of wood flour polyurethane composites

From Figure(6), The thermal conductivity is increased slightly with the increasing of the wood flour content, due to the increasing of the natural fiber incomposite sample which has a larger thermal conductivity than polyurethane foam. This phenomenon occurs due to the fact that at high concentration ratios, the filler particles become closer to each other and this ends up with the thermalbridges along the path of the heat flow, which causes an increase in the effectivethermal conductivity of the composite. The thermal conductivity of wood is affected by a number of basic factors: moisture content, temperature, grain direction, and structural irregularities. As the proportion of wood flourincreases, the moisture content will increase, leading to an increase in the thermal conductivity of the composite, becausewater is moreconductive than wood and polyurethane, so the thermal conductivity increases proportionally as the moisture level is increased.

Figure (7), represents a comparison between experimental and theoretical results of thermal conductivity varying with filler volume fraction. Theoretical results for effective thermal conductivity of WPCs are calculated using Maxwell – Garnett Model [10]. Fiber volume fraction is calculated according to ASTM D2584 [12]. From Figure (7), the experimental results show that the measured thermal conductivities of WPCs are higher than the predicted values by the theoretical model. It has been found that the errors associated with the theoretical model, with respect to experimental ones, lie between 40% to 50%. This maybe due to some of the assumptions taken of models that are not practical. Furthermore, in the theoretical model's orientation of the

fillers was assumed to be perfect, but in actual practice, when the fillers are added to the foam matrix, some of the fillers may be misaligned.

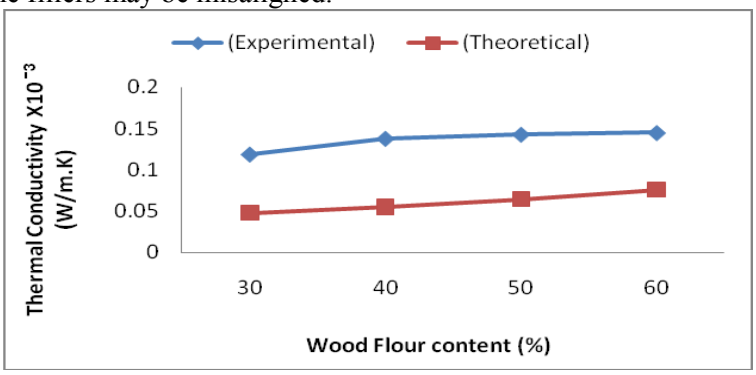


Figure (7): comparision between experimental and theoretical thermal conductivity

Conclusions

The following conclusions could be drawn from the results of the present study:

1-Density of the composites is reduced with the increasing of wood flour content. A slight difference (about 5%) between the experimental and theoretical values of density was observed.

2-Addition of wood flour content as a filler increases the water absorption of WPCs. A significant increase in water absorption was observed as the proportion of the wood flourcontent became 60%.

3-Thickness swelling of the composite increased with increasing wood flour content. This can be attributed to the high hemicellulose and high lignin content in wood flour.

4-Thermal conductivity of WPCs increases with the increasing concentration of wood flour content. A comparison between the experimental values of thermal conductivities results from this study with Maxwell – Garnett Model, which have found that it's bigger than theoretical expectations within a range of (40% to 50%).

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الملخص

تناولت الدراسة الحالية بيان تأثير إضافة نشارة الخشب الناعمة على الخصائص الفيزيائية (الكثافة، امتصاص الماء، الانتفاخ العرضي) و على معامل التوصيل الحراري لمتراكبات الخشب- البولي يوريثان اللدن. تم تصنيع عينات من المتراكب بنسب وزنية مختلفة من نشارة الخشب الناعمة (30%، 40%، 50%، و 60%). تم دراسة الخواص الفيزيائية ومعامل التوصيل الحراري لهذه المتراكبات وفق المقاييس العالمية المعتمدة. بيَّنت الدراسة الحالية تناقص كثافة المتراكبات عند زيادة نسبة نشارة الخشب المضافة.يُعَزى هذا النقصان في الكثافة إلى زيادة امتصاص الكثافة من قبل نشارة الخشب. اوضحت النتائج أيضاً أن زيادة نسبة نشارة الخشب الناعمة أدت إلى زيادة كمية امتصاص الماء وكذلك زيادة الانتفاخ العرضي للمتراكبات. السبب في هذه الزيادة يعود إلى طبيعة مادة الخشب السليلوزية التي تسمح بامتصاص كمية أكبر من الماء عند زيادة تركيزها في المتراكبات. من ناحية أخرى، أظهرت النتائج زيادة معامل التوصيل الحراري للمتراكبات عند زيادة نسبة نشارة الخشب الناعمة. هذه الزيادة سببها مرة أخرى زيادة كمية الرطوبة الممتصة من قبل نشارة الخشب التي تؤدي إلى زيادة معامل الامتصاص الحراري للمادة المتراكبة.

الكلمات المفتاحية: متراكبات الخشب-البولي يوريثان، الكثافة، امتصاص الماء، الانتفاخ العرضي، معامل التوصيل الحراري.