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Analysis and development of recycled materials for wood plastic composite product

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ABSTRACT

This study is to find out the best optimum ratio of the mixed waste materials of high-density polyethylene and wood's flour by comparing the mechanical properties of recycled product (WPC). In this study, the fabrication process of wood plastic composite (WPC) contained of virgin material, post-consumer high-density polyethylene, and wood's flour is through a twin-screw extruder and hot compression machine. The WPC product with four different ratio filler content based on weight percentage, *i.e.* 0 wt %, 20 wt %, 30 wt % and 40 wt % , were tested using universal tensile machine and impact tester according to ASTM D 3039, ASTM D 790, and ASTM D 6110 . The results of experiment showed that tensile and flexural properties of the composite increased with the adding of the wood's flour material. The testing showed, however, totally opposite to the result of the impact test. In overall, the results of observations showed that recycled WPC have better mechanical properties compared to non-recycled WPC.

Introduction

By the increasing amounts of garbage or wastes day by day are, as undeniable facts, aligned with the growth of world's population against waste originated from daily consumption/usage. In this context, the wastes of plastic that are naturally as non-biodegradedable and indecomposable materials will end up in the dumping site without knowing the real benefits on how to use its further. To overcome this problem, therefore, the study is performed to develop the composite that called as wood plastic composite or WPC from recycled material such as wood flour and plastic. The product made contains of the virgin and post consumer materials (*e.g.* the High Density Polyethylene orHDPE) that will be functioned as the polymer matrix material and wood are flour as the reinforcement material.

The Wood Plastic Composites (WPC) or known as wood fibre-plastic composites and/or green composites are a new group of materials that were since two decade ago has generated the interest of the industry for many applications. The WPC products (as composite materials) used are made in a range of polymers, such as polyethylene (PE), polypropylene (PP) or polyvinylchloride (PVC) with the various proportions along wood or other natural fibres, in which they will be used to produce profiles or mould objects with the structural integrity and workability of wood and the durability of polymers.

Specifically, the recycled materials used are post consumer materials such as crushed HDPE and the wood flours produced from furniture manufacturing or sawmill plant. In order to make WPC, in this study we use the matrix variation of processes contains of the second generation of materials - known as recycled material - and the first generation material as non-recycle or virgin material. On this, to find out the better performances of product, the test carried out are through mechanical testing such as tensile test, flexural test, and pendulum impact test, while against the behaviour of wood plastic composite through the tests such as the tensile strength, yield point, and modulus of elasticity (MOE).

Material and Preparation

Thermoplastic Material

The most common polymers used for WPC products are the recycled and virgin material, such as PE and PP. The reason behind of its is to such material due to the cost consideration in which price of PE-based products in the market are relatively cheaper, besides the availability and the characteristic of a higher heat distortion temperature compared to PVC-based products. In this study, PE-based plastics materials used would be processed below 200^{0}C.

The choice of plastic types depends on the particular application requirements [4]. The virgin plastics and any recycled plastic which will be melted and processed below the degradation point of wood, however, require the suitable materials for manufacturing WPC. In this study, the plastics type is the recycled HDPE in the form of fine granules (i.e. the post-consumer products such as milk plastic bottles) that having the melt flow index 0.7 gram per 10 minutes.

Fig. 1. Recycled HDPE (Milk Plastic Bottles)

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Wood Flour

The wood material used must be free of any non-wood contaminants, such as nails or dirt, even though the contaminants are only affects to the appearance properties rather than performance.

There were two main options of wood material, i.e. wood flour and wood fibre. In facts, wood’s flour is much more widely available than wood fibre, beside it's reportedly cheaper. In addition, wood’s flour is relatively free flowing and much easier to be processed (such as feeding and handling process) rather than wood fibrewood, even though fibre type gives somewhat better performance.

The wood’s flour is however available in many sizes, from 20 mesh (coarse) to 400 mesh (extra fine), but the 40 mesh size is the most common one. For most of applications, a satisfactory result composition based on criteria between price, performance, and ease of processing were the wood’s flour with size of 40 mesh of (Table 1).

Table 1. Effect of Pine Wood Flour Particle Size on Performance of Wood-filled Polypropylene Composite [3]

Mesh Size	Notched Izod Impact (J/M)	Tensile Strength MPa	Bending Stiffness GPa
20	27	23.3	2.98
40	21	24.4	3.10
80	19	24.8	3.12
120	17	24.2	2.84

According to Stark [3], despite the wood species have some effects on composite processing and performance, they are however having differences are small. Therefore, it is very practical to make the composite by the wood species that is readily available at the least cost. What actually the critical concern in this study is the moisture of wood. Ideally, the moisture content should be 5 % to 8 % or less. In addition, the fine mesh size of wood flour which will affect to product performance, such as to increase the stiffness and reduce the impact strength. The longer size of wood fibres contributed to strength and also the bind difficulties with the polymer. For most applications, however, the 0 mesh wood flour gave satisfactory performance and ease of processing [5].

Figure 2 shows the virgin wood flour from the timber process plant/ sawmill and the recycled wood flour of the post consumer wood furniture. The screening of wood's flour of recycled wood is required in order to shift out the strange particles. By a vibrating inclined process, the particles required must be passes the mesh 80 or 0.25 mm in which they were air-dried and oven-dried at 100^0C to reduce the moisture content until to less than 2% [2].

(a)

(b)

Fig. 2. Wood flour (a) Virgin (b) Recycle

Additives

The different types of additive used for WPCs product are to aid the processing operation, such as lubricants that provide processing stability and preservation in long-term service (i.e. heat and light stabilizers), and also to improve mechanical properties such as coupling agents. The function of additive as lubricants are as following:

a. Internal. This act in the resin phase to increase the melting flow, prevent the shear burning, and resist the melting fracture by reducing viscosity at high shear rate.

b. External. This act at the interface between resin and other materials to improve the releasing of the composite, promote the dispersion of fillers, resisting the melted fracture, and reducing friction between resin and process equipment. Primary phenolics (free-radical scavengers) or secondary hosphate (hydroperoxide decomposer) might be used as heat stabilizers for WPC, while light stabilizers that were commonly used in WPC includes UV absorbers (benzotriazole or benzophenone), radical scavengers or hydroperoxide decomposers, and hindered amine light stabilizers (HALS).

Furthermore, for the function as compatibilizers or coupling agents, the additive (polymers) contains of both polar functional groups that can react or interact with the hydroxyl groups of cellulose and non-polar chain sections that are more compatible with the hydrocarbon chains of WPC polyolefin [7]. The compatibilizers promote the adhesion and dispersion of the wood component in the polymer matrix, besides improving the mechanical properties. The compatibilizers used as WPC additives, as an example, is Maleic Anhydride (MAH) that is grafted from polyethylene (PE-g-MAH) and polypropylene(PP-g-MAH), Trimethoxyvinylsilane that is grafted from Polyethylene and Analogous Copolymers, and Polymethylene Polyphenylene Isocyanate and Methylol Phenolic that is grafted from Polyolefins [8].

Specimen Preparation

The ratio levels of wood's flour applied in this study is 20 wt. %, 30 wt. % and 40 wt. % based on the total weight, while the rates of HDPE are 100 wt. %, 80 wt. %, 70 wt. % and 60 wt% as stated in Table 2. The extruder co-rotating twin-screw type is used to blends the thermoplastic composites reinforced with wood’s flour. The purpose of using the twin-screw extruder is to produce the homogeneous composite pellets.

Normally, the blend material will be in the bulk shapes. However, this shape is not appropriate for the pressing process. Instead of using the crushing machine, all the 6 ratios of blended materials should be crushed into pallet form. The process applied is repeatedly carried out for few times until the form of materials become the appropriate pallet size. The blending temperature is set up to 190^0 C with screw speed at 50 rpm. HDPE and silane put into mixer and let it melt for 3 minutes, followed by feeding the wood flour based on composition design in Table 2 and then let it mix uniformly in 5 minutes.

After completed the compounding process, the mixed raw material in bulk shape then crushed into pallet form. To control the thickness of panel materials, the panel produced should be formed into the flat panel. To fabricate this panel, all of materials in pallet form were pressed by using hydraulic hot press machine with fixed set up at 200^0 C for upper and lower plate, the pressing pressure at 25 tons for 6 minutes followed by cooling until room temperature. For the panels testing, the specimens were taken from the plate and cut into the dimensions according to ASTM standard.

WPC Type	Wood Flour (wt.%)	HDPE (wt.%)	Additive Agent (wt.%)
A	0	100	0
B	20	80	2
C	30	70	2
D	40	60	2

Table 2: Ratio of the raw material and additive agent of the both recycled and virgin material for WPC fabrication

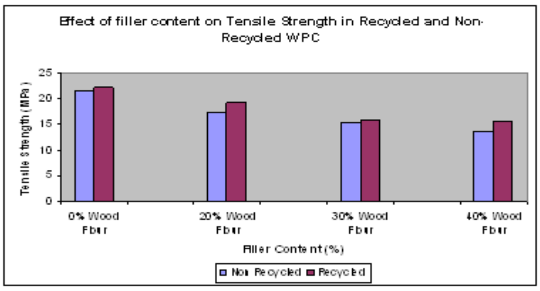
Results and Discussion

Tensile Testing

Figure 3 shows the test results of tensile modulus versus filler content of wood plastic composite. From this tensile test showed that the value of tensile strength is decreased, while the filler content is increased. Besides, the value of tensile modulus is increased gradually when the filler content increased. The results showed some differences of the without-filler specimen compared to others 3 types formulation as follows:

a. The modulus value for non-recycle HDPE material is higher than the value of recycled HDPE material, but the modulus value of recycled material for the rest of 3 formulations is higher than the virgin HDPE material.

b. Without mixing any filler in pure HDPE will show the better modulus result compared to the recycled HDPE. Through wood fibre variation, the result showed that the using of either virgin or recycled HDPE is significantly affects to the modulus and tensile values.



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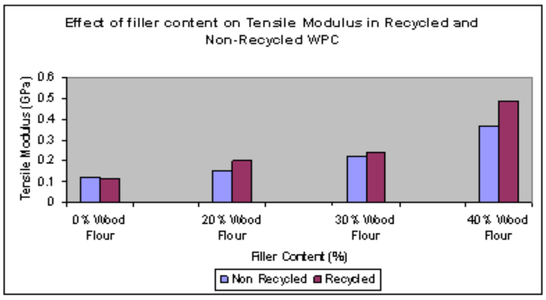
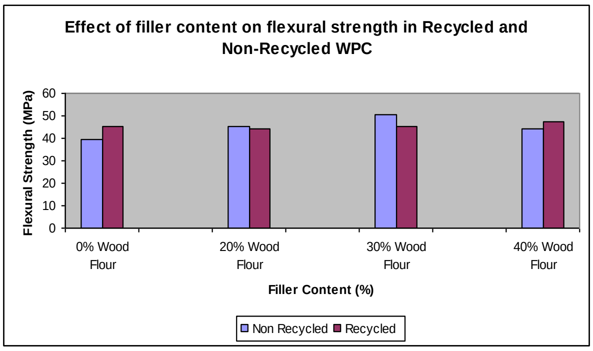


(b)

Fig. 3. Recycled and non-recycle WPC (a) tensile strength (b) tensile modulus

Flexural Testing

Figure 4(a) shows that the value of flexural strength for non-recycled WPC is increased when the filler content increased for 0 wt. % to 30 wt. %, but it is decreased for 40 wt. %. Theoretically, the strength value must increase when the filler content is 40 wt. %. However, in this experiment, the value shown lower than filler content 30 wt. %. Most probably reason is because the material was not properly blended when mixing process carried out. The same problem occurred on the recycled material when the filler is added to 20 wt. % content, where the strength value of samples is supposed to be higher than pure PE [9].



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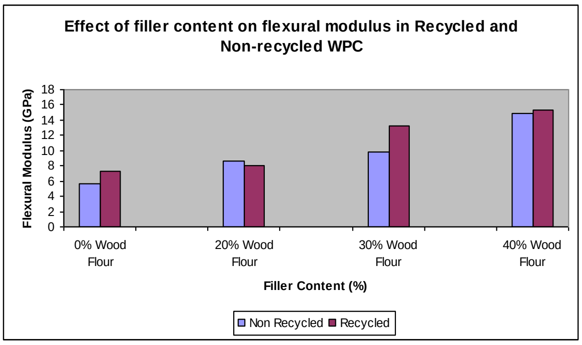

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Fig. 4. Recycled and non-recycled WPC (a) Flexural strength (b) flexural modulus

Refers to the flexural modulus graph as is shown in Figure 4(b), the modulus' values are gradually increased when the filler content is increased. Normally, the modulus values specimens of PE content with wood fibers are better than neat PE as were proved in Figure 4(b). For example, the virgin material filler content with 20 wt. % is 8.6265 GPa and it increased by 14.3 % for 30 wt. % is 9.8574 GPa. Theoretically, the flexural properties are influenced by quantity of the fillers [10].

Impact Testing

The charpy-impact strength test of un-notched composite specimens is carried out according to ASTM D6110. This test is corresponded to the energy lost by the pendulum during the breakage of the specimens. It is the sum of the energies required to produce fracture initiation, fracture propagation, bending of the specimens, production of vibration, friction loss in the arm bearing and on the face of the sample after failure. Figure 5 shows that there are no significant differences of the impact strength test results between recycled material and non-recycled material. There were only slightly drop when the filler content is high, even though the impact strength for filler content formulation for 30 % filler (that is 0.92 KJ/m2) is less than 40 %.

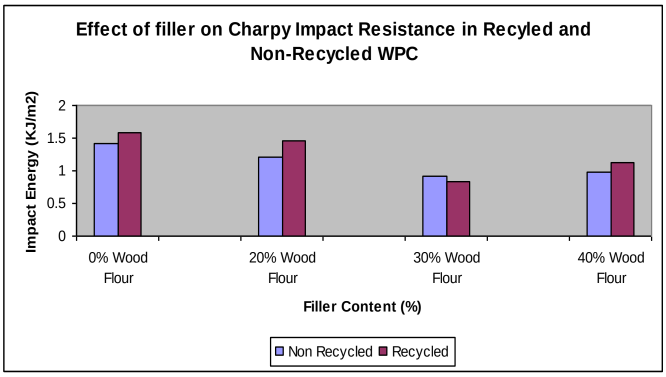


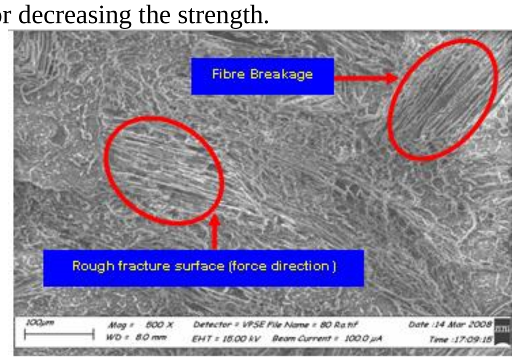
Fig. 5. Impact Energy (KJ/m^2) of the Recycled and Non-Recycled WPC

Mechanical Properties of WPC

The mechanical interlocking between wood's fibre and HDPE matrix are significantly affected to the mechanical properties. When wood fibres used were by high percentage, then it makes micro-voids between matrix and fibre in the composite content. This is means that when the micro-voids are increased, then the tensile modulus is increased. This is an indication that finer wood particle structure increases the mechanical properties [11]. The reasons are:

a. By using the coarse particle and the high quantity of wood flour will make the air-trapped in the composite between the bonding areas. This air became the micro-voids foam after the compacting process and content at bonding area.

b. By using the coarse size wood flour, make the debonding within the matrix (Figure 6). Actually, some of wood particles were not properly bonded with the molten thermoplastic where they make some gap areas. This defect makes the composite failure or decreasing the strength.



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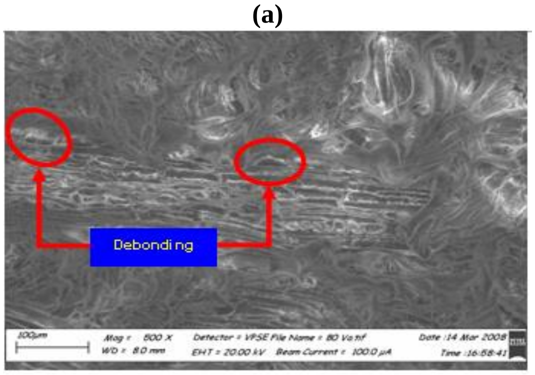


(b)

Fig. 6. Fracture surface of tensile specimen for 20% filler in (a) recycle WPC, and (b) non-recycled WPC

The size of filler influenced the tensile and flexural properties as shown in Figure 4. The size of the filler is divided into coarse particles, such as above 20-150 mesh, large particle size for 150-200 mesh, and fine particle size around 0.1 µm. The using of coarse filler will affect to decreasing the strength, but increasing the modulus value. While fine particle provides better flow of molten composite where at that moment is the good bonding between matrix and fibre. The type of filler used on matrix is very important. The strength value of this formulation is better than the wood flour [10]. The reasons are as following:

a. Most of the filler types in powder form. So, it will well blend with matrix and gives better properties.

b. A wide distribution of particles of mineral filler can beneficial because it can provide a better packing density of particles in matrix [10].

c. By reinforcing of wood fibre with high aspect ratio and glass fibre, strength of the filled plastic will increases. It shows that effect of the wood's flour on tensile properties of matrix are, typically, depending on particle size of the filler and type of mineral filler used. Finally, the strength of composite is improved by size of filler particles, instead by the percentage of the filler added on matrix.

This study showed that micro-voids effecting the WPC properties. Whenever the fibres were added or bond with HDPE matrix, then it tends to absorb the moisture. Compared to the fine particle wood flour, the coarse size tends to absorb the moistures. Theoretically, when fibre ratio is increased, then the un-notch impact energy is decreased [12]. This is as shown in Figure 5. In addition, when the matrix content is high, then the energy used to break the specimen is also high. On this,

a. When the energy applied is easy to break the specimens/ panels, then this is due to the filler effect to the decreasing the properties of composite. Therefore, the size and shape of filler content is very important against the tensile and flexural properties.

b. If the composite product use coarse wood fibre, then the composite will break easily compared to the using of fine particles. The fine particles in which they are well mixed or blended by matrix will result the composite with high strength.

Morphology Analysis

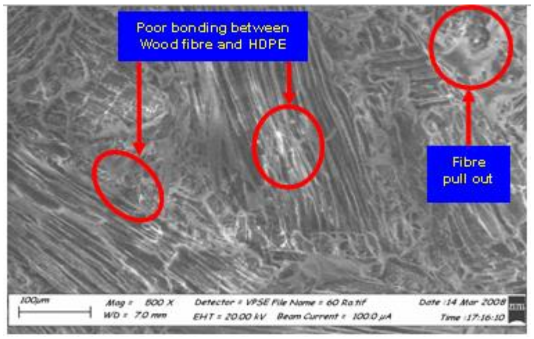
SEM Examination on Tensile Fracture Surface

Generally, there were two ways of WPC might be fractured, that are:

a. If the WPC is stressed with parallel force direction, then the fractures occurred with cracks running on the interfacial area and pulling out the fibre from the matrix as is shown in Figure 6.

b. Alternatively, if the WPC is stressed in the perpendicular direction to the force, the intermolecular cleavage occurred with the cracks running across fibre, interfacial area, and polymer as well as break the fibres.

The fractures surface along the longitudinal direction had fairly tight matrix against the wood particles and were relatively free of voids (Figure 8). According to Figure 7, some of wood fibres had pulled out away from the matrix and a few had fractured in the transverse direction. This is because when the transverse direction specimens hit by the load head, less dissipation energy occurred is eventually resulting in lower impact of strength [13]. The transverse direction specimens had more number of voids and had rough surface as is depicted in Figure 8. When the load head hit the transverse direction specimens, the impact wave had to meet the different phases were more often i.e. fibre, interfacial area, polymer and voids.



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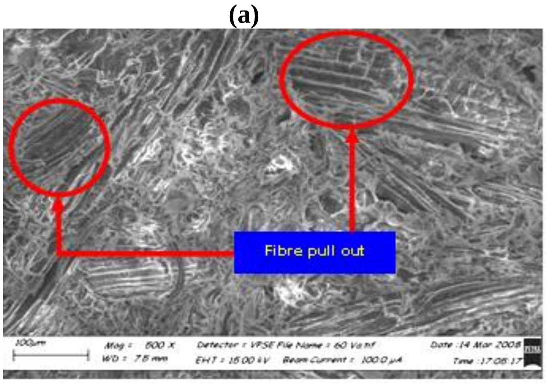

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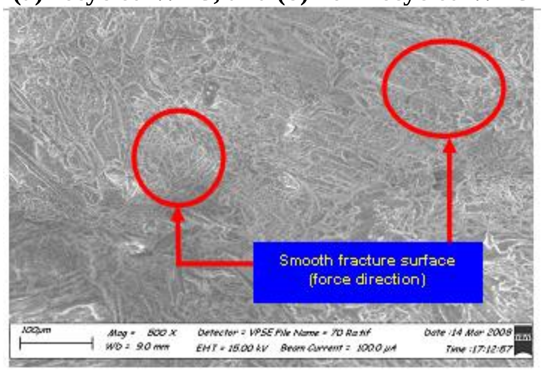
Fig. 7. Fracture surface of tensile specimen for 40% filler in (a) recycled WPC, and (b) non-recycled WPC



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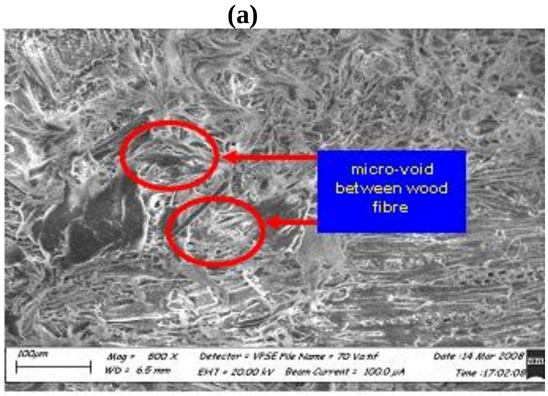


(b)

Fig. 8. Fracture surface of tensile specimen for 30% filler in (a) recycled WPC, and (b) non-recycled WPC

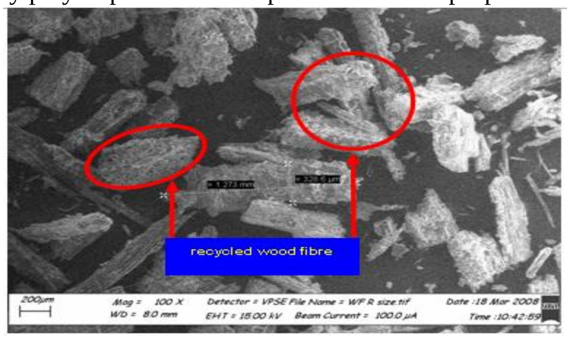
Dissipation energy occurred more often on along the longitudinal direction where the micro voids between fibre and matrix are assumed to consume the energy impact. The main fractured mechanism along the longitudinal direction was due fibre pullout and interfacial bond failures, while the main fractures mechanism in the transverse direction was due to fibre breakage.

SEM Examination on Wood Flour Particle Structure

According to the SEM micrograph as is shown in Figure 9, the particles are anisotropic. The average size of the particular recycled wood's flour is around 328 µm and the virgin is 481 µm. The average initial fibre length is 1.8 ± 0.8 mm, but they had been reduced during processing. Actually, the increasing of the wood contents causes the melt viscosity increase that leads to thermo-mechanical degradation of the fibres. Figure 9 shows that the average fibre length is 2.1 ± 0.8 mm and 1.3 ± 0.4 mm for virgin and recycled wood content respectively. This value must be treated with caution, because anisotropy biases somewhat the results of the laser diffraction measurements.

The large particles debonding were easily under the effect of external load event at very small loads induced the separation of the interfaces at 500 to1000 µm size. Extensive debonding leads to the decreasing of strength with the increasing of filler content. The processing at high temperatures may change its structures and properties. Besides, cellulose fibres, as well as processing conditions may also influence the crystalline structure of the matrix polymer [12].

The two different types of material composites contain of different sizes of wood flour are presented in Figure 9. The fine mesh wood flour increases the stiffness, but reduces the impact strength. The longer fibres contribute to strength, but more difficult to bind with the polymer [8]. From the observation, the composite using recycled wood flour produced the good mechanical test results. This is because, perhaps, the particle size of recycled wood's flour is a bit smaller than virgin wood's flour. However, we cannot conclude that the filler is a best function in this composite. Even though the filler is best function for WPC samples, the HDPE functioned as matrix is also actually play important role to produce the best properties.



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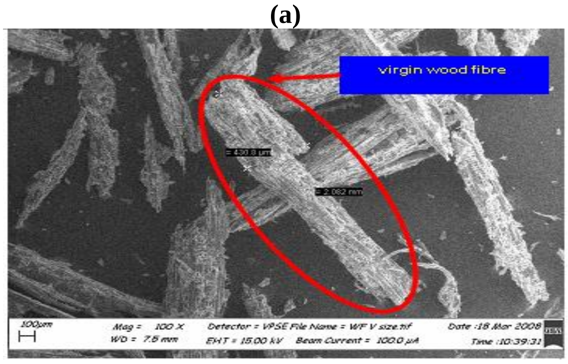

(b)

Fig. 9. SEM micrograph showing (a) recycled wood fibre, and (b) non-recycled wood fibre

Influence of Aspect Ratio

Aspect ratio in this context is a ratio of fibre length to fibre thickness. For wood flour, the ratio is about 328 µm and 481 µm for recycled and the virgin respectively. Generally, long fibres and oriented along the flow render a composite material with improved mechanical properties compared to short-fibre-filled composite material. In the other words, a higher aspect ratio leads to better flexural properties. As a result, in WPCs, the fibre is the main load bearing component. The more of fibre are oriented along the flow, the higher of the flexural properties of the material will be. The length and width of wood fibre typically is 2 mm and 0.5 mm, with aspect ratio of 15 or a length of 10 mm and width of 4 mm with aspect ratio of 25.

The mixing processes resulted the decreasing f aspect ratio of fibre due to they were shorten the fibre thickness. For example, the processing of blanched wood fibre along with polyethylene in a co-rotating twin screw extruder at 100 and 300 rpm resulted the fibre length became shorter,in which a fraction of the smallest particles which around 50 µm in length were increased to 3% ~ 5%.

Influence of Coupling Agents

Based on the this study, the coupling agent used for WPC panels has no effects to the structure produced since the structure of the composite depends on the composition of material as well as types of material used such as recycled material and virgin material. By adding the solid coupling sealant agent to the melt or applied emulsified sealant directly to the fibre, there was only little effect found on mechanical performance of composites [9]. They are not significantly increasing the flexural and tensile modulus, only insignificantly existed. This is contrary to what was expected, where the strong covalent bonds formed between the wood filler and the HDPE.

Influence of Matrix

The changing of the melt flow index (MFI) of virgin polyethylene had a significant effect on viscosity, but there are a little effect on mechanical performance over the range studied (0.7g/10 min). The result suggested that MFI could be used to compensate the increased viscosity of the higher performance fibrous composites. The matrix will act to protect the individual fibres from surface damage as a result of the mechanical abrasion and chemical reactions towards the environment.

The virgin WPCs composites are stronger and stiffer than the recycled WPCs composites, but had lower flexural and tensile strength values. Similar trends were also seen on composites made by recycled material. For example, through the using of recycled polyethylene (a block copolymer with higher impact strength) instead of the virgin polyethylene, resulting the composites with higher impact strength, although the difference in this property was not nearly as great as for the unfilled polymers [10].

In other words, although the choice of plastic can affect composite properties, by the addition of reinforcement, actually, can also affect the mechanisms in which the plastic achieve its performance. This may offer some practical utilities if the problems associated with the high viscosity of the recycled system can be overcome.

Conclusion

In this study, the second generation of materials, which is recycled WCP, showed better for strength and modulus value. This is due to the content of recycled WPC contains of more than one additive or due to the other chemical agent contributes to maintain the properties.

For the mechanical properties, 30 wt. % is the optimal filler content for both types of WPC. Since the recycled WPC is equally competitive to the non-recycled WPC, by using the second generation material can reduce the manufacturing cost as well as save the natural resources from depletion.

Overall, the effects of adding filler material into a HDPE matrix would result in the decreasing of tensile and flexural strength, but it increase the Young's and flexural modulus.

- Filler content $\propto$ Decrease in Strength, Increase in Rigidity

The ductility of composite is reduced when the filler increased. In this regard, the amount of filler content affects the toughness of the material (as the ability to the plastically deformed is drastically reduced at high filler contents).

- Filler content $\propto$ Decrease in Ductility

The conclusion for the mechanical properties, as follows:

a. It is shown that 30 wt. % is the optimal filler content for wood's flour.

b. While the filler content below 30 wt. %, anyhow, it can be still used for the applications considered (trade-off the performance between strength, rigidity, and resin usage).

c. The performance of composites with filler content above 30 wt. % does not justify the amount of resin usage reduction.

The future study is required for melt-blended composites made from recycled wood fibre and plastics in order to improve the properties and processing materials as well as to increase the potential of applications. This is to corresponds the potential of the composite materials made with other major components of the waste stream, such as low-density polyethylene, polystyrene, and mixed waste plastics. Beside, the using different types of additive between wood fibre and plastic matrix to enhance physical and mechanical properties, as well as the impact energy and the resistances to relatively extreme environments (especially fire) and in turn, biodegradation.

In general, the mechanical and dimensional stability properties of second-generation of WPC samples made from recycled material are an essentially equivalent to or better than properties obtained from first-generation [13]. Panels made by recycled materials are more favourably compared to those made of virgin materials. On this, the mechanical and physical properties of panels made with recycled polyethylene fibre or high density polyethylene and waste wood are similar to those of panels made by non-recycled materials. The second-generation composites, or possibly with higher one, can be produced by using recycled materials without reducing property values as a consequence [13].

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