Wood Plastic Composites from Waste Plastic Bags: Formula Optimization Using Extreme Vertices Mixture Design

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Abstract. Waste plastic bags (WPB) from high density polyethylene (HDPE) have been reprocessed for many applications. However, it is limited to be used as wood plastic composites (WPC) for building applications as the decrease of mechanical properties. This research examined the optimal composition of WPB that can be added to WPC and complied with Indonesian Standard (SNI) 8154 – 2015. Samples were produced according to extreme vertices mixture design using twin screw extruder (TSE). The materials were WPB bounded in the range of 0 to 20 wt%, sawdust at 50 to 70 wt%, neat HDPE at 30 to 50 wt%, and HDPE grafting maleic anhydride (PE-g-MA) at 0 to 10 wt%. The compositions were optimized to obtain desired flexural modulus and strength as required on SNI standard. The optimized formula was then reproduced and characterized to analyze its water content, density, swelling, and formaldehyde content. The results showed that the optimum formula was predicted at 3 wt% of WPB, 50 wt% of sawdust, 37 wt% of neat HDPE, and 10 wt% of PE-g-MA, giving 2111 MPa and 27.3 MPa of predicted values that above the minimum requirement for WPC material (2000 MPa for modulus, 20 MPa for strength). Meanwhile, the modulus and strength of verification sample were 2173 MPa and 25.7 MPa. Further analysis on the other properties showed 0.6 ± 0.1% of water content, 1.7 ± 1.4% of swelling, and 0.015 ± 0.000 mg/L of formaldehyde that lower than maximum specifications, and 1.13 ± 0.01 g/cm³ of density that above minimum the requirement.

1. Introduction

Wood plastic composites (WPC) are composed of wood and plastic, thus reduce wood consumptions. It has capability to replace traditional material such as metal and wood due to mechanical properties improvement [1]. It has been applied in building, such as panels, roofing, fencing and outdoor structure [2]. However, despite its benefit, the use of virgin plastics as matrix should also be reduced as it is non-renewable character. Therefore, utilization of waste plastics is an alternative that can be done.

In asia, Indonesia become one of the second largest producer of waste plastic after China [3]. It was predicted that one of contributor to the huge amount of waste plastic in Indonesia was waste plastic bag (WPB) which was produced almost more than 10 million annually [4] from high density polyethylene (HDPE). This is a big problem to environment and waste management. On the other hand, this is also
potential as a material resource for WPC production because of its slight changes on mechanical properties compared to the virgin HDPE [2].

The use of recycled HDPE from post-consumer goods, including WPB for WPC production has been explored by many studies [2, 5, 6]. However, there is lack information on how material composition is optimized in order to comply with standard. Therefore, further study is needed to formulate the optimum compositions of WPC consisted of mostly wasted material including waste wood and WPB.

A technique that could be applied to formulate WPC is mixture design of experiment. It is a special technique of response surface analysis in which investigated product is made from several ingredients that have 100% in total [7]. It was reported that this technique has been applied to formulate many material compositions [8-13]. The formulation could be done using simplex method (lattice or centroid) or extreme vertex. However, due to the nature of materials and mixture boundaries that need to be done in very small portion, extreme vertex is more reasonable to be performed.

In this work, WPC from WPB was produced and optimized using extreme vertex mixture design. The formulation was performed to obtain minimum flexural modulus and strength that comply to Indonesian Standard (SNI). Other properties of optimized and verified sample as water content, density, swelling and formaldehyde content were then tested and analyzed compared to standard requirement.

2. Experimental method

2.1 Materials

Neat high-density polyethylene (HDPE) Titanvene 6070EA (flexural modulus 1700 MPa) from Lotte Chemical Titan Nusantara was utilized as matrix in WPC along with shredded waste plastic bags WPB as co-matrix. 80-mesh size sawdust from various hard woods was used as filler for reinforcement. Sawdust was pre-dried at 80 °C for 24 hours in advance of usage to reduce water content. Polyethylene-graft-maleic anhydride (PE-g-MA) used as compatibilizer was produced based on the route developed in our laboratory [14] using HDPE Titanvene 6070EA, maleic anhydride (MA) and benzoyl peroxide (BPO). MA and BPO were purchased from Merck.

2.2 Formulations and manufacturing of WPC

WPC were compounded according to formulas listed in Table 1. These formulas were generated in Minitab 16 based on extreme vertices mixture design. Sawdust content was chosen within the range of 50% to 70% due to expectation to prediction optimum amount that existed in this boundary, and processing issue. Neat HDPE contents were ranged from 0% to 50%, WPB from 0% to 30%, PE-g-MA from 0% to 20% to give polymer bi-matrix concentration of 50% in each formula and economical reason.

| Formula | HDPE [wt%] | WPB [wt%] | Sawdust [wt%] | PE-g-MA [wt%] |
|---------|------------|-----------|---------------|---------------|
| 1       | 50.0       | 0.00      | 50.0          | 0.00          |
| 2       | 30.0       | 0.00      | 70.0          | 0.00          |
| 3       | 30.0       | 0.00      | 60.0          | 10.0          |
| 4       | 40.0       | 0.00      | 50.0          | 10.0          |
| 5       | 30.0       | 20.0      | 50.0          | 0.00          |
| 6       | 30.0       | 10.0      | 50.0          | 10.0          |
| 7       | 35.0       | 5.00      | 55.0          | 5.00          |

WPC compounding was done using Collins ZK 25 twin screw extruder. All ingredients were hand-mixed prior to compounding. The resulted mixture was poured into hopper at once and processed according to conditions listed in Table 2. The resulted pellets were air dried and transferred to
compression moulding machine Collins P300P for specimen preparations. Conditions for compression moulding are listed in Table 3.

| Temperature (°C) | Screw Speed (rpm) |
|------------------|-------------------|
| T₁               | 40                |
| T₂               | 170               |
| T₃               | 175               |
| T₄               | 180               |
| T₅               | 185               |
| T₆               | 190               |
| T₇               | 190               |
| T₈               | 190               |
|                  | 60                |

**Table 3. Compression Molding Conditions.**

| Parameter          | Steps | 1 | 2 | 3 | 4 | 5 |
|--------------------|-------|---|---|---|---|---|
| Temperature [°C]   |       | 190|190|190|190|40|
| Pressure [Bar]     |       | 5 | 5 | 5 | 5 | 5 |
| Time [Minute]      |       | 5 | 5 | 5 | 5 | 15|

2.3 Characterizations

2.3.1. Flexural modulus and strength. In advance of testing, the molded samples were cut to 50 mm x 150 mm and pre-conditioned at humidity 50 ± 5 % and temperature 23 ± 2 °C for more than 40 hours. Flexural characteristics of WPC were obtained according to ISO 16978 standard using Universal Testing Machine (UTM) Shimadzu AG-X Plus 50 kN. The load speed was set at 0.3 MPa/sec. Flexural modulus was calculated from the load between 10 % and 40 % of maximum load. The ultimate flexural strength was recorded automatically. The results were compared to required value in SNI 8154 – 2015.

2.3.2. Water content. Water content was measured following the procedure in SNI ISO 16979 standard. 20 miligrams of samples were weighted, followed by drying at 103 °C in an oven until mass equilibrium was reached. The dried sample was weighted. Water content of sample was calculated using equation (1) and averaged for 5 test specimens, where M₀ represents initial mass and M₁ is dried mass.

\[
\text{Water content} (\%) = \frac{M₀ - M₁}{M₁} \times 100\% \tag{1}
\]

2.3.3. Density. Density of WPC was measured according to SNI ISO 9427 standard. The sample was cut to a size of 50 x 50 mm. Thickness was measured at the position of the diagonal intersection of the sample. Length and width were measured in the center of the sample, which passes through the midpoint of the intersection between diagonals. The mass of the sample was measured using a density balance with an accuracy of 0.0001 grams. Sample density was calculated according to equation (2) and averaged for 6 test specimens.

\[
\text{Density} (\rho) \left(\frac{\text{kg}}{\text{m}^3}\right) = \frac{m}{b₁ \times b₂ \times t} \times 10^6 \tag{2}
\]

Where
- \(m\) = initial mass
- \(b₁\) = length
- \(b₂\) = width
- \(t\) = thickness

2.3.4. Swelling. Percentage of swelling of sample WPC was carried out according to SNI ISO 16983 standard. The sample was cut to 50 x 50 mm specimen, and soaked in water of pH 7.0, at 20 ± 1 °C, for 24 hours. The specimen was vertically placed in thermal humidity chamber. The thickness of the specimen before and after soaking was measured using micrometer at the point of diagonal intersection.
of the specimen. Percentage of swelling was calculated based on equation (3) and averaged for 8 test specimens.

\[
\text{Percentage of swelling (\%) } = \frac{t_2 - t_1}{t_1} \times 100\%
\]

(3)

Where \( t_1 \) = thickness before soaked
\( t_2 \) = thickness after soaked

2.3.5. Formaldehyde content. Formaldehyde content was measured according to JIS A 1460 – 2001 standard. Prior to measurement, standard solution containing formaldehyde i.e 0, 0.2, 0.4, 0.8, 2.0 and 4.0 mg/L were made and the absorbance of solution was measured using UV-Vis. Calibration curve was obtained. To measure formaldehyde content of WPC, 10 WPC specimens were placed above a cup containing 300 mL distilled water. The specimens and the cup were isolated in a dessicator for 24 hours. The test environment was controlled at temperature 20 ± 2 °C, and humidity 65 ± 5%. After 24 hours, the DI water is reacted with acetylacetone ammonium acetate (AAAA). The absorbance of formaldehyde contained distilled water was measured. Formaldehyde content was calculated using equation (4).

\[
\text{Formaldehyde content (mg/L) } = \frac{F \times (A_s - A_b) \times 1800}{S} + \text{Intercept}
\]

(4)

Where \( F \) = Slope of calibration curve (mg/L)
\( A_s \) = Absorbance of formaldehyde contained distilled water
\( A_b \) = Absorbance at standard curve
\( S \) = Total area of samples (cm²)

3. Results and discussions

3.1 Flexural modulus and strength

The resulted flexural modulus and strength is listed in Table 4. It shows that flexural modulus of WPC is within the range of 1630 MPa (formula 5) to 2173 MPa (formula 4), while strength of WPC is spread between 11.2 MPa (formula 2) to 29.1 MPa (formula 4). The average modulus of all samples is higher than neat HDPE.

![Table 4. Flexural Strength and Modulus of WPC.](image)

| Formula | HDPE [wt%] | WPB [wt%] | Sawdust [wt%] | PE-g-MA [wt%] | Sawdust : HDPE | Modulus [MPa] | Strength [MPa] |
|---------|------------|------------|---------------|---------------|----------------|---------------|---------------|
| 1       | 50.0       | 0.00       | 50.0          | 0.00          | 1.00           | 1748          | 2000          | 21.5          | 18           |
| 2       | 30.0       | 0.00       | 70.0          | 0.00          | 2.33           | 1799          | 2000          | 11.2          | 18           |
| 3       | 30.0       | 0.00       | 60.0          | 10.0          | 2.00           | 2093          | 2000          | 21.1          | 18           |
| 4       | 40.0       | 0.00       | 50.0          | 10.0          | 1.25           | 2173          | 2000          | 29.1          | 18           |
| 5       | 30.0       | 20.0       | 50.0          | 0.00          | 1.66           | 1630          | 2000          | 17.2          | 18           |
| 6       | 30.0       | 10.0       | 50.0          | 10.0          | 1.66           | 1924          | 2000          | 23.0          | 18           |
| 7       | 35.0       | 5.00       | 55.0          | 5.00          | 1.57           | 1922          | 2000          | 20.5          | 18           |

Average 1898
Standard Deviation 191

However, comparison on the obtained flexural properties and SNI 8154 – 2015 show that there are only two samples (formula 3 and 4) comply to standard, namely those with limited amount of WPB and high PE-g-MA level success. In contrast, most of samples (except formula 2 and 5) have strength that higher than that of stated in SNI. The highest strength is produced by sample with low content of sawdust.
and high level of PE-g-MA, while the lowest is obtained by that of with high percentage of sawdust and limited amount of PE-g-MA. Unexpected low strength for sample 2 and 5 might be influenced by weak interfacial bonding by the absent of compatibilizer (PE-g-MA).

Meanwhile, Table 4 also presents the effect of sawdust, WPB, and PE-g-MA to neat HDPE. The Table shows that mixing sawdust with neat HDPE (formula 1) successfully increase the modulus of the latter. The modulus increases from 1700 MPa to 1748 MPa. This means that sawdust has positive impact on modulus of WPC. However, further addition of sawdust, increase modulus only slightly (51 MPa) by increasing amount of sawdust to 70% as in formula 2 (ratio of sawdust to HDPE is 2.33). In contrary, unexpected decrease of strength is observed by the increase of sawdust. The strength is down from 21.5 MPa in formula 1 to 11.2 MPa in formula 2. This is probably due to insufficient amount of HDPE to wrap the surface of sawdust that tend to be agglomerated, as well as the absent of PE-g-MA, leaving poor interconnection of WPC constituent in plastic region.

In formula 3, the incorporation of PE-g-MA while keeping the neat HDPE constant, reduces the amount of sawdust, hence the ratio is down to 2.00. The strength of WPC then arises to 21.1 MPa. Further lowering the ratio to 1.25 by raising the amount of neat HDPE in formula 4, results in the highest strength of WPC (29.1 MPa). These results indicate that the lower the ratio of sawdust to HDPE, the more HDPE is available to wrap the surface of sawdust, the higher the strength. Other than that, the incorporation of PE-g-MA into WPC promotes good bonding between sawdust and neat HDPE that produce better modulus and strength simultaneously.

Moreover, introducing WPB into HDPE and sawdust as in formula 5, reduced modulus of WPC compared to that of neat HDPE. However, modulus and strength of formula 5 (1630 MPa and 17.2 MPa) are lower than those of formula 1 (1748 MPa/21.5 MPa). This result indicates that WPB has inferior effect compare with neat HDPE probably due to truncated polymer chain of the former as result of reprocessing as well as the existence of other ingredients in WPB such as colorant, extender and etc. Improved results are achieved by adding PE-g-MA to the formulation (formula 6). The resulted WPC has higher properties than that of without PE-g-MA due to good bonding among materials.

The result in Table 4 was then analyzed using Minitab 16 and the models are generated. Equation (5) and equation (6) predict the flexural modulus and strength of WPC according to its composition. Equation (5) shows that neat HDPE, WPB and sawdust independently have positive effect on flexural modulus of WPC. In contrary, sole PE-g-MA has the opposite influence. This might be caused by can be explained by the fact of its low intrinsic mechanical properties. The entanglement and interconnection among HDPE chains are loosen by grafting maleic anhydrate pendant groups on polymer backbones. This results in separation of polymer molecules that deprive the mechanical properties thereof. However, interaction between neat HDPE and PE-g-MA has the highest positive influence on this property. The interaction of both materials effects flexural modulus 100 times higher than that of PE-g-MA alone. This is because PE-g-MA and neat HDPE have identical polymer backbone which results in good interaction between both polymers as well as between sawdust. This is also true on flexural strength of WPC as describe on equation (6). Interaction between neat HDPE and PE-g-MA as well as neat HDPE and WPB alone are beneficial to flexural strength of WPC. In contrary, sawdust and PE-g-MA independently have detrimental effect on the property.

Modulus elasticity = 1577*HDPE + 907*WPB + 1921*Sawdust – 146.5*PE-g-MA +14976*HDPE*PE-g-MA

\[ E = 1577\times \text{HDPE} + 907\times \text{WPB} + 1921\times \text{Sawdust} - 146.5\times \text{PE-g-MA} + 14976\times \text{HDPE}\times \text{PE-g-MA} \] (5)

Strength = 46.7*HDPE + 24.0*WPB - 3.8*Sawdust – 12.0*PE-g-MA + 336.0*HDPE*PE-g-MA

\[ q = 46.7\times \text{HDPE} + 24.0\times \text{WPB} - 3.8\times \text{Sawdust} - 12.0\times \text{PE-g-MA} + 336.0\times \text{HDPE}\times \text{PE-g-MA} \] (6)

Predictions of optimized compositions are shown in Figure 1 to 3. Figure 1 depicts WPC formulations with maximum flexural properties. It is shown that the maximum modulus would be obtained at composite with no additional WPB. However, this was not the desired output of this research, as the WPB should be maximized. For this reason, the composition was predicted by manipulating the vertical red line on the generated response optimizer to left or right side. By doing this at particular line,
it would change another composition. The result shows that the maximum WPB that can be added to mixture and satisfy the SNI 8154 – 2015 requirement was 8%, resulted 2002 MPa for modulus and 24.5 MPa for strength (Figure 2). However, due to safety factor and standard deviation, the formula was predicted to produce modulus higher than 2100 MPa (Figure 3).

**Figure 1.** WPC formulation for maximum flexural properties.

**Figure 2.** WPC formulation at maximum WPB level that satisfy SNI 8154 – 2015 for flexural.

**Figure 3.** WPC formulation that satisfy SNI 8154 – 2015 for flexural.

| WPB [wt%] | Sawdust [wt%] | HDPE [wt%] | PE-g-MA [wt%] | Modulus [MPa] | Error [%] | Strength [MPa] | Error [%] |
|-----------|---------------|------------|---------------|---------------|-----------|---------------|-----------|
| 3.00      | 50.0          | 37.0       | 10.0          | 2111          | 3%        | 27.3          | 6%        |

Table 5. Flexural Strength and Modulus of Verified WPC formula.

To verify the desired composition, a verification sample was prepared and tested. Results of verification experiment were summarized in Table 5. It is shown that there is good agreement between models and verification data. 3% and 6% of errors are achieved for modulus and strength, respectively. Slightly higher error for strength can be explained by the fact that the composition was calculated based on the model for modulus. However, these relatively low errors indicate that good manufacturing of WPC from HDPE, WPB, sawdust and PE-g-MA is possible.
3.2 Characterization of optimized sample

The verified WPC sample is tested according to SNI 8154 – 2015 and JIS A 5741. All the testing results is compiled in Table 6. In general, the verified WPC sample is able to satisfy all the criteria specified by the standard.

| No | Test Item               | Result   | Specification |
|----|-------------------------|----------|---------------|
| 1  | Water content [%]       | 0.6 ± 0.1| ≤ 12%         |
| 2  | Density [g/cm\(^3\)]   | 1.13 ± 0.01| ≥ 0.60      |
| 3  | Swelling [%]            | 1.7 ± 1.4| ≤ 4%          |
| 4  | Formaldehyde content [mg/L] | 0.015 ± 0.000| ≤ 0.4     |

The water content of the sample is 0.5687 which is much lower than specified by the standard. This value illustrates the good bonding between polymer matrix and sawdust, which manage to prevent the return of moisture into sawdust after manufacturing.

The sample density of verified WPC is 1.13 g/cm\(^3\). This value is almost double the specifications determined by SNI (> 0.6 g / cm\(^3\)). This indicates the good interaction between matrix and filler as well as less void that occurs in the sample, hence increases the flexural modulus of elasticity and strength.

The swelling of verified WPC sample that occurred after 24 hours is 1.71% is listed in Table 6. This value is below the maximum limit allowed by SNI. As explained in density test section, the compactness and the reduce void in the sample as well as good bonding between matrix and filler prevent the re-entry of water into the sample, hence hindering the swelling to be occur.

![Figure 4](image)

**Figure 4.** (a) Specimens set up before test, (b) specimens position in dessicator, (c) distilled water containing formaldehyde in desiccator reacted with AAAA solution

Test of formaldehyde content was depicted in Figure 4. The test results show that formaldehyde content in the sample is 0.015 mg/L. This formaldehyde content can be originated from recycle material used in making verified WPC. However, the amount of formaldehyde released can be minimized if there is good interaction and strong bonds between matrix and filler. Polymer matrices that wrap sawdust well, not only prevent the return of moisture to sawdust, but also prevent the release of formaldehyde from sawdust.

4. Conclusion

Formula optimization of wood plastic composites (WPC) from waste plastic bags (WPB) high density polyethylene (HDPE) has been done using extreme vertice mixture design in order to comply to Indonesian Standard (SNI) 8154 – 2015. Formulation was conducted using twin screw extruder with material boundaries were set in the range 0 to 20 wt% of WPB, 50 to 70 wt% of sawdust, 30 to 50 wt% of neat HDPE, and 0 to 10 wt% of HDPE grafting maleic anhydride (PE-g-MA). The optimized formula was then verified and observed its water content, density, swelling, and formaldehyde content.
The results showed that 3 wt% of WPB, 50 wt% of sawdust, 37 wt% of neat HDPE, and 10 wt% of PE-g-MA would give optimum values of flexural modulus and strength (2111 MPa and 27.3 MPa), that higher than minimum specification stated in SNI 8154 – 2015 (2000 MPa for modulus, 20 MPa for strength). Further analysis on verification samples resulted 2173 MPa and 25.7 MPa in modulus and strength, respectively. It also had 0.6 ± 0.1% of water content, 1.7 ± 1.4% of swelling, and 0.015 ± 0.000 mg/L of formaldehyde content that lower than maximum specifications, and 1.13 ± 0.01 g/cm³ of density that above the minimum requirement. Good results of verification sample show that WPC from WPB is possible to be processed and complied to Indonesian Standard (SNI).

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