Experimental and micromechanical modelling of randomly oriented zalacca fibre/low-density polyethylene composites fabricated by hot-pressing method

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Wahyu Purwo Raharjo¹,²,*, Rudy Soenoko², Anindito Purnowidodo² and Moch Agus Choiron²

Abstract: This study aims to investigate the effect of the volume fraction of zalacca fibres on the tensile strength and modulus of the LDPE-ZF composites both experimentally and analytically. The tensile tests were carried out on the randomly oriented LDPE-ZF composites manufactured by hot-pressing method. Hirsch and Bowyer–Bader’s model was used to predict the tensile strength of the composites where the elastic modulus was determined by using Tsai–Pagano, Manera and Cox–Krenchel’s model.

The experimental results of tensile strength and elastic modulus of the composites was close by the Bowyer–Bader and Tsai–Pagano’s model, respectively. It is due to the consideration of fibre length and orientation factor in Bowyer–Bader’s model and fibre elastic anisotropy in the Tsai–Pagano’s model. This study establishes that the hot press method is applicable for processing LDPE-ZF composites.
Subjects: Structural Mechanical Engineering; Mechanics of Solids; Packaging  
Keywords: micromechanical model; zalacca fibres; LDPE composites; hot press

1. Introduction

Composite materials have been extensively used in many structural applications because of their high strength and stiffness at low weight and good corrosion resistance and fatigue properties (Beylergil, Tanoğlu, & Aktaş, 2017). Trends in an application of natural fibre composites appear along with the technology development and economic growth due to the scarcity of conventional energy resource such as fossil fuels. In the transport sector, the utilization of material with high specific strength is important because fuel consumption correlates directly with vehicle weight (Koronis, Silva, & Fontul, 2013). Other important factors are lower material and manufacturing cost, emission reduction, safety improvement and recycle-ability (ElayaPerumal & Venkateshwaran, 2008). Environmental concerns encourage the development of biocomposites that at least one component, matrix and fibres, are natural or biodegradable.

In general, composite panels utilize a polymer matrix. The most applicable thermoplastic polymer for household and industry is low-density polyethylene (LDPE) (Ibeh, 2011). The advantages of LDPE are its low density, good mechanical properties, high resistance of acid, base and salt, low cost and recycle ability. LDPE has lower tensile strength and elastic modulus but its fracture strain, hardness and impact strength are higher than high-density polyethylene (HDPE) (Bashford, 1997). There are two main drawbacks of LDPE compared with thermoset polymer, namely low tensile strength and elastic modulus. To strengthen and stiffen it, one of the simple methods is by reinforcing fibres to become composite.

LDPE and its composite can be made with several processes such as extrusion, injection moulding, compression moulding and hot press. Compared to extrusion and injection moulding, in the hot pressing process, the mixture of powdered LDPE matrix and the reinforcing fibre are put into the dies then pressed in high temperature. The advantage of hot press is its lower temperature process compared with the extrusion and injection moulding which implies lower manufacturing costs and the easily arranged fibre orientation. In injection molding and extrusion (using screw extruder), a mixture of molten thermoplastic and short fibres is injected from the hot reservoir into a colder metal mould at very high pressure. The composite is allowed to solidify and then ejected. During the processing, the changes in fibre orientation often occur due to the elongational flow and shear flow of the melt (Hull & Clyne, 1996).

Zalacca (Zalacca edulis) is a tropical plant from Southeast Asia, especially Indonesia. It is cultivated for its fruit. Generally, the zalacca is harvested twice a year. After harvest, four to six of its old midribs are cut to maintain the quantity and quality of its crops. The cut midribs are discarded as wastes or compost. Based on the study of Raharjo and co-workers (2016), the zalacca fibre (ZF) has cellulose content of 42.54% and tensile strength and elastic modulus attain 182.12 MPa and 3.36 GPa, respectively. Hence, the fibres of the zalacca midribs have potential as a composite reinforcement.

To avoid costly and time-consuming experiments, the prediction of mechanical properties of the composites such as tensile strength and elastic modulus must be made through established mathematical models. To do this, the intrinsic properties of fibres and matrix are important factors in generating accurate prediction of the mechanical properties of the composites. However, there are no experimental tests of mechanical properties, and micromechanical modelling applied in prediction of tensile strength and elastic modulus of ZF-LDPE composites.

This study compared the experimentally mechanical properties of randomly oriented LDPE-ZF composites fabricated by the hot pressing process with the micromechanical model proposed by...
Hirsch and Bowyer-Bader for predicting the tensile strength and Tsai-Pagano, Manera and Cox-Krenchel’s models for elastic modulus of the randomly oriented ZF reinforced LDPE composites.

Due to the complex structure of the most natural fibre, there is elastic anisotropy in which the longitudinal properties of the fibres are greater than their transverse properties (Sun, Zhao, Wang, & Ma, 2014). The assumptions that the fibre was isotropic are failed to predict the stiffness of randomly oriented natural fibre composites (Epaarachchi, Ku, & Gohel, 2010). Hence, the elastic modulus of ZF is separated as longitudinal and transverse elastic modulus, \( E_{f1} \) and \( E_{f2} \), respectively. These two modulus are used to predict the elastic modulus of the composite by Tsai-Pagano’s model.

2. Experimental

2.1. Material

The matrix used was LDPE Petlin LD C150Y purchased from Petlin Sdn. Bhd., Malaysia. The LDPE was in the form of pellets then powdered before processes by hot press method.

The fibres were extracted from the midribs of three-years-old zalacca plants from the plantation in Yogyakarta, Indonesia. The midribs were cut and retted by soaking them in distilled water for 2 weeks. After separating the husks, the fibres were then washed with distilled water, dried in the open air at room temperature for 48 h. They were dried with a ventilated oven at 105°C for 45 min then cut into of ± 40 mm length.

2.2. Fibre tensile properties

The essential mechanical properties of ZF are tensile strength (\( \sigma_f \)) and elastic modulus (\( E_{f1} \) and \( E_{f2} \)). The tensile strength and longitudinal elastic modulus (\( E_{f1} \)) were obtained by the tensile test of single fibre based on the ASTM C1557-03 standard. The test was performed by universal testing machine JTM-UTS 210 with a load-cell capacity of 0.5 kN and gauge-length of 20 mm. Cross-head speed was maintained at 5 mm/min. Ten specimens were tested. Broken samples near the clamps were not analysed. The tensile strength of each fibre was determined by dividing the applied load by the average cross-sectional area. The single fibre cross-sectional geometry was assumed as circular. The average diameter of the fibre was measured at four locations along the fibre length using an optical microscope Olympus SZX2-TR30 and CellSens® software. The strain was calculated by dividing the fibre length change by the initial length of the fibres. The elastic modulus was determined by dividing the stress by the strain of the fibre in the linear portion of the stress vs. strain curve.

Because of the difficulty of measuring the transverse modulus of single fibre (\( E_{f2} \)), a method based on the work of Cichocki and Thomason (2002) was used in which composites consist of unidirectional ZF in LDPE matrix with transversal fibre direction, as shown in Figure 1. The composite was processed by arranging ZF transversely in the composite dies with a definite volume fraction. The matrix and ZF reinforcement then were processed with hot press at the pressure, temperature and holding time of 5 MPa, 115°C and 5 min, respectively. The composites were tensile tested in accordance with ASTM D638-02 standard using the same testing machine with load-cell capacity, gauge-length and cross-head speed of 20 kN, 50 mm and 5 mm/min, respectively. The average result was then named as the transverse modulus of composite (\( E_{22} \)) and filled with the inverse rule-of-mixture (Gibson, 2012), Equation 1, to get \( E_{f2} \).

\[
\frac{1}{E_{22}} = \frac{V_f}{E_{f2}} + \frac{V_m}{E_m}
\]

in which \( E_{22} \), \( E_{f2} \), \( E_m \), \( V_f \) and \( V_m \) represent the transverse modulus of the composite, transverse modulus of the single fibre, modulus of the matrix, volume fraction of fibre and volume fraction of matrix, respectively.
2.3. Composite preparation

The randomly oriented short fibre composites were processed by powdering the LDPE using a high-power blender and sieving through an 80-mesh strainer then storing it in plastic containers. The fibres were cut into 10 mm length. The weight of LDPE powder and ZF was measured with Ohaus AX423 digital scale to ensure the desired composition. There were six variation of ZF weight fraction: 0, 10, 20, 30, 40 and 50 wt. %. The weight fraction was able to convert to volume fraction by using Equation (2) (Shackelford, Han, Kim, & Kwon, 2016).

\[
V_f = \frac{W_f/\rho_f}{W_f/\rho_f + W_m/\rho_m}
\]  

(2)

in which \(W_f\), \(W_m\), \(\rho_f\), and \(\rho_m\) are the weight fraction of the fibre and the matrix and the density of fibre and matrix, respectively. The LDPE powder and ZF were mixed using an electric mixer for 1 h and then fed into the dies manually. The hot-pressing process was carried out with a hydraulic press machine at a pressure of 5 MPa, a temperature of 115°C and 5 min of holding time. Once cooled with air, the composites were then milled to obtain the desired dimension for testing. The tensile testing specimens accomplished from the process were shown in Figure 6.

2.4. Density and fracture morphology of the composites

The LDPE-ZF composite density was measured and calculated based on the Archimedes principle, in accordance with ASTM D1037 standard. The theoretical density of the composites was determined using the Equation (3) (Gibson, 2012).

\[
\rho_c = \frac{1}{(W_f/\rho_f) + (W_m/\rho_m)}
\]

(3)

2.5. Tensile testing of LDPE-ZF composites

The tensile strength and elastic modulus of random oriented LDPE-ZF composites were determined by tensile testing based on ASTM D638-02 standard using universal testing machine JTM-UTS 210 with load cell capacity of 20 kN, gauge-length of 50 mm, and cross-head speed was maintained 5 mm/min. There were five specimens for each variation of the weight fraction.

Instead of the weight fraction, the volume fraction of the fibre was used in the comparison of the experiment with the micromechanical models.

2.6. Scanning electron microscopy analysis

The fracture surface of LDPE-ZF composites was examined by scanning electron microscopy (SEM) to investigate the cause and mechanism of fracture. The SEM equipment was FEI Type Inspect S50.
The samples were coated with gold using a low-vacuum sputtering machine and were put on the silver-paint holder before they were inserted into the SEM chamber.

3. Result and discussion

3.1. Properties of LDPE matrix and single zalacca fibre

The physical and mechanical properties of LDPE matrix are shown in Table 1, based on the hot-pressing process.

The zalacca fibres were measured and tensile tested. Their physical and mechanical properties are shown in Table 2. By using Equation (1), the transverse modulus of single ZF (\(E_f^2\)) was defined as 565.29 MPa.

3.2. The densities of composites

The composite density was measured and calculated. The measured and calculated density was named experimental and theoretical density, as shown in Figure 2.

In general, the theoretical densities of the composites were higher than the average experimental ones, except for 0 and 50 wt.% of ZF. This is due to the voids in the measured composites whereas the theoretical densities do not calculate the voids. For a neat LDPE, the theoretical and average experimental densities were the same value. It is due to the composites consist only the LDPE matrix. There is no weight fraction of fibres added in Equation (3). For the addition of 50 wt.% ZF, the theoretical density was approximately the same as the average experimental one. This is

| ZF weight fraction (%) | ZF volume fraction (V<sub>f</sub>) | Tensile strength (MPa) | Elastic modulus (MPa) |
|------------------------|-------------------------------|-----------------------|---------------------|
| 0                      | 0                             | 8.85 ± 0.19           | 35.55 ± 1.15       |
| 10                     | 0.14                          | 8.37 ± 0.31           | 152.00 ± 17.97     |
| 20                     | 0.26                          | 10.60 ± 0.29          | 270.46 ± 14.23     |
| 30                     | 0.38                          | 14.45 ± 2.18          | 400.85 ± 56.87     |
| 40                     | 0.50                          | 15.75 ± 1.77          | 483.50 ± 68.07     |
| 50                     | 0.60                          | 11.14 ± 1.20          | 435.54 ± 37.63     |

Table 1. Physical and mechanical properties of LDPE matrix

| Property                  | Value            |
|---------------------------|------------------|
| Density (g/cm³)           | 0.921            |
| Melting temperature (°C)  | 111              |
| Tensile strength (MPa)    | 8.85 ± 0.187     |
| Elastic (Young) modulus (MPa) | 35.55 ± 1.152  |
| Elongation at break (%)   | 746 ± 190        |

Table 2. Physical and mechanical properties of zalacca fibre (ZF)

| Property                  | Value            |
|---------------------------|------------------|
| Diameter (μm)             | 144.85 ± 22.41   |
| Density (g/cm³)           | 0.601 ± 0.0045   |
| Tensile strength (MPa)    | 182.12 ± 79.38   |
| Longitudinal elastic modulus, \(E_f^1\) (GPa) | 3.36 ± 1.24      |
| Elongation at break (%)   | 5.6 ± 0.96       |

Table 3. Experimental tensile strength and elastic modulus of LDPE-ZF composites

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caused by the compactness of the fibre packing in the matrix which reduces the change of the composite volume during the hot-pressing process.

### 3.3. Effect of fibre addition on tensile strength and elastic modulus

The fibre loading of LDPE-ZF composites varied from 0, 10, 20, 30, 40 and 50 wt.%. According to Equation (2), the addition of ZF was expressed by volume fraction of the fibre. The effect of fibre loading on the tensile strength and elastic modulus of LDPE-ZF composites is shown in Table 3.

![Figure 2. Experimental and theoretical density of LDPE-ZF composites.](image_url)

The addition of 10 wt. % ZF (0.14 V_f) slightly decreases the tensile strength, then increases it up to 40 wt. % (0.50 V_f) and then decreases with the higher fibre loading. The fibre loading of 10 wt. % reduces the strength because the volume fraction is below the critical value (Gibson, 2012). For composite in which the matrix failure strain is assumed to be greater than the fibre failure strain, the fracture of the composite is due to the failure of the fibres. When the longitudinal stress of the fibre reaches the fibre tensile stress (s_f1), the longitudinal stress of the matrix achieves the value of s_mf1.

\[
s_{mf1} = E_m \varepsilon_f
\]

Hence, the longitudinal strength of the composite is calculated as:

\[
s_l = s_{f1} V_f + s_{mf1} V_m = s_{f1} V_f + s_{mf1} (1 - V_f)
\]

in which V_f and V_m represent the volume fraction of the fibre and matrix, respectively. If the value of V_f is too low, the longitudinal stress of the composite (s_l) is lower than the longitudinal stress of the matrix (s_m). This is due to s_{mf1} < s_m because \(\varepsilon_f < \varepsilon_m\). This case was nearly similar to the study of Andre and co-workers (2016) for nonwoven kenaf fibre-epoxy composites.

More than 10 wt. %, the addition of ZF increases the tensile strength by up to 40 wt.%. The increase of fibre content enhances the load hold up by the composite. It is due to the fibres as the largest part of the load support. The higher content of ZF, 50 wt. % (0.61 V_f), decreases the tensile strength. It is because the theoretical volume-fraction of the fibre approaches the maximum value of 78.5 and 90.7 % for the square and triangular fibre-packing geometries, respectively (Gibson, 2012). The fibre loading of 50 wt. % causes the imperfect bond between the fibre and matrix during the hot-pressing process.

The fibre loading of ZF consistently raises the elastic modulus of the composites up to 40 wt. % and then decreases. This is related with the stiffness after the fibre addition in the LDPE matrix.
where $\varepsilon_f < \varepsilon_m$. The ZF addition higher than 40 wt.% coming close to the maximum value of theoretical volume-fraction causes the inferior binding between the fibre and matrix.

### 3.4. SEM analysis

The examination of composite fractures using SEM is shown in Figure 3. The composite fracture with 10 wt.% (0.14 Vf), as shown in Figure 3(a), reveals that there are several fibre pullouts and holes in the matrix. This indicates that the external load is borne solely by the matrix. The fibres do not support the load due to the poor adhesion between the fibres and the matrix. The role of the fibres is only to restrict the strain of the LDPE matrix. The decrease of the strength of this composite compared with the neat LDPE is due to the incapability of the matrix in transferring the load to an inadequate amount of fibres so that the fibres seem to be the defects in the matrix. This fact is due to the inadequacy of fibre content under the critical volume fraction of the fibres (Gibson, 2012). The case was met in the study of Mahdavi and co-workers (2010) in the mechanical properties of date palm fibre-PE composites, Lu and Oza (2013) in mechanical properties of hemp fibre composites with virgin and recycled HDPE, Singh and co-workers (2014) in tensile and flexural behaviour of hemp fibre reinforced virgin and recycled HDPE matriced composites.

Figure 3(b) shows that there are more fibres in the LDPE—20 wt.% ZF (0.26 Vf). The higher quantity of the fibres allows more effective load transfer from the matrix to the fibres. This induces the partial load support by the fibre having higher strength and stiffness than the matrix. It leads to higher strength and elastic modulus of the composite.

Further addition of fibres up to 40 wt.% (0.50 Vf) as shown in Figure 3(c) leads to higher load transfer by the matrix that the larger part of the external load is supported by the fibres. It is proved the better bond between the fibres and the matrix causes fewer fibre pullouts and holes. The higher fibre content causes the larger interfacial area between the matrix and the fibres. Hence, it causes the higher load transferred between the matrix and the fibres.

Figure 3. SEM examination of fracture surface of: (a) LDPE—10 wt.% ZF, (b) LDPE—20 wt.% ZF, (c) LDPE—40 wt.% ZF and (d) LDPE—50 wt.% ZF.
Higher fibre loading as shown in Figure 3(d) for LDPE-50 wt.% ZF (0.60 Vf) reveals that there are imperfect bonds between the fibres and the matrix causing a decrease of load transfer to the fibre. This results in the decline of the strength and stiffness of the composite.

3.5. Prediction of the tensile strength of the composites

The mathematical models utilized to predict the tensile strength of randomly oriented LDPE-ZF composites at various Vf were Hirsch’s model and Bowyer–Bader’s model. Both models were considered appropriate and had been used widely in previous studies and had accurate estimation (Andre et al., 2016).

The first model presented by Hirsch is based on a combination (parallel and series) of rule-of-mixture. The tensile strength of the composite is given as

$$\sigma_C = x(\sigma_m V_m + \sigma_f V_f) + (1 - x) \frac{\sigma_m \sigma_f}{\sigma_m V_f + \sigma_f V_m}$$

(6)

In which $\sigma_C$, $\sigma_m$, and $\sigma_f$ represent the tensile strength of the composite, matrix and fibre, respectively. x is the parameter determining the stress transfer between matrix and fibres. For composite with randomly oriented fibre, the value of x is 0.1 based on the compatibility of experimental and theoretical calculation results (Kalaprasad et al., 2004).

The second model by Bowyer and Bader based on the assumption that the tensile strength of thermoplastic matrixed composite with short fibre reinforcement is the sum of its subcritical and supercritical fibre and matrix (Bowyer & Bader, 1972). The tensile strength of the composite is given as:

$$\sigma_C = \sigma_f K_1 K_2 V_f + \sigma_m V_m$$

(7)

in which $K_1$ and $K_2$ are the orientation and length factor of the fibre, respectively. For fibre with $l > l_c$,

$$K_2 = \frac{l}{2l_c}$$

(8)

and for fibre with $l < l_c$,

$$K_2 = \frac{l^2}{2l_c^2}$$

(9)

For random-oriented fibre composites, the value of $K_1$ is 0.2, based on the previous research (Curtis, Bader, & Bailey, 1978).

The theoretical tensile strength was calculated only for fibre loading of LDPE-ZF composite limited to 40 wt.% (50 Vf) due to the decrease of the experimental results caused by higher Vf. The tensile strength of LDPE-ZF composites obtained by Hirch and Bowyer–Bader’s model was shown in Figure 4.

The Hirsch’s model predicted the higher tensile strength compared to Bowyer–Bader’s and the experiment for all fibre loading. From Figure 4, the tensile strength of Hirsch’s model deviated by 50%, 52%, 39% and 56% at 0.14, 0.26, 0.38 and 0.50 Vf from the experimental results, respectively. It is due to the presence of the inverse rule-of-mixture and the non-existent of fibre length factor in Hirsch’s model. The model proposed by Hirch has inverse rule-of-mixture having the values of 8.85–20.92. The Hirsch’s model has only the parameter of the stress transfer between the matrix and fibre, x is 0.1 for randomly oriented fibre composites. The deviation of the strength obtained by the Hirsch’s model from the experimental results is caused by the ever-increased value of the tensile strength calculated from Equation (6) being proportional to the volume fraction of the fibre. The maximum value is achieved from the largest theoretical volume fraction. On the other hand,
The experimental results are not proportional to the volume fraction of the fibre. In the low volume fraction, $0.14 V_f$, the strength decrease due to its lower volume fraction than the critical value. The volume fraction higher than $0.50 V_f$ causes the decrease in the tensile strength of the composite due to the imperfect bond between the matrix and fibres.

The Bowyer–Bader’s model results were relatively close to the experimental results. They vary by $18\%$, $2\%$, $−19\%$ and $−20\%$ at $0.14$, $0.26$, $0.38$ and $0.50 V_f$, respectively. The Boyer-Bader’s model has fibre orientation factor $K_1$ and fibre length factor $K_2$ that have values of $0.2$ and $0.5$, respectively. The values of $(\sigma_f V_f)$ are in the range of $26.00$ and $110.44$, meanwhile $(\sigma_m V_m)$ are between $8.85$ and $3.48$. The divergence between the Boyer-Bader and the experimental results is caused by the linear results proportional to the fibre volume fraction obtained from the Equation (7). Meanwhile, there is decrease of the tensile strength for volume fraction lower than $0.14 V_f$ and higher than $0.5 V_f$.

The advantage of the Bowyer–Bader’s model over Hirsch’s model is that besides the orientation factor $K_1$, there is also the length factor $K_2$ of the fibre. Meanwhile, Hirsch’s model has only the stress transfer parameter $x$ being similar to $K_1$ in Bowyer–Bader’s. Simultaneously, the factor $(1−x)$ multiplied with the inverse rule-of-mixture in Hirsch’s model gives the addition of larger result than that obtained by Boyer-Bader’s model.

The result of the tensile strength of LDPE-ZF composites gained by the tensile test had a little difference from Bowyer–Bader and Hirsch’ model except for fibre loading higher than $0.50 V_f$. It indicates that the hot press is an applicable method for fabricating the LDPE-composites.

### 3.6. Prediction of the elastic modulus of the composites

The modelling by Tsai–Pagano (Gibson, 2012) is used to estimate the elastic modulus of composite with the fibre distributed randomly in a plane. The model is expressed in the equation

$$E_C = \frac{3}{8} E_{11} + \frac{5}{8} E_{22}$$

in which $E_C$, $E_{11}$ dan $E_{22}$ are the elastic modulus of randomly oriented fibre composite, longitudinal and transverse elastic modulus for composite with uniform oriented fibre and similar aspect ratio $(l/d)$ and volume fraction $(V_f)$, respectively. Simultaneously, Halpin–Tsai’s equation is able to calculate $E_{11}$ and $E_{22}$:
\[ E_{11} = E_m \left( 1 + \frac{2}{2} \frac{\eta_1 V_f}{1 - \eta_1 V_f} \right) \quad \text{with} \quad \eta_1 = \frac{E_{f1}}{E_{fl}} - \frac{1}{1 - \frac{E_{f1}}{E_{fl}}} \tag{11} \]

\[ E_{22} = E_m \left( 1 + 2 \frac{\eta_2 V_f}{1 - \eta_2 V_f} \right) \quad \text{with} \quad \eta_2 = \frac{E_{f2}}{E_{fl}} - \frac{1}{1 + \frac{E_{f2}}{E_{fl}}} \tag{12} \]

in which \( E_{f1} \) and \( E_{f2} \) are longitudinal and transverse elastic modulus of a single fibre, respectively.

Manera (1977) has arranged an equation for predicting the elastic modulus of randomly oriented short-fibre composite:

\[ E_C = V_f \left( \frac{16}{45} E_{f1} + 2E_m \right) + \frac{8}{9} E_m \tag{13} \]

This model assumes that the mechanical properties of randomly oriented fibre composite are the same as the properties of laminates with an infinite number of layers oriented in all direction.

The Cox–Krenchel’s model uses modified rule-of-mixture:

\[ E_C = \eta_l \eta_o E_f V_f + E_m V_m \tag{14} \]

in which \( \eta_l \) and \( \eta_o \) are the fibre length distribution and orientation factors, respectively. In this model, the existence of void can be ignored (Cox, 1952). According to Krenchel, the fibre orientation factor is expressed as:

\[ \eta_o = \cos^4(\alpha_o) \tag{15} \]

where \( \alpha_o \) is the limiting angle of fibre orientation. The value of \( \eta_o \) according to Thomason and Vlug (1996) for laminate containing randomly oriented fibre in a plane is 0.375. At the same time, \( \eta_l \) calculated based on Cox’s theory is given as:

\[ \eta_l = 1 - \frac{\tanh \left( \frac{\beta l}{2} \right)}{\frac{\beta l}{2}} \tag{16} \]

\[ \beta = \frac{2}{d} \sqrt{\frac{E_m}{E_m(1-v_M)\ln \left( \frac{d}{x_i V_f} \right)}} \tag{17} \]

in which shear parameter \( \beta \) expresses the stress concentration coefficient in the fibre ends, \( v_M \) is the Poisson’s ratio of the matrix and \( l \) is fibre length. In this case, the fibre arrangement was considered as rectangular so that \( x_i = 4 \).

Figure 5 showed the theoretical elastic modulus predicted by Tsai–Pagano, Manera and Cox–Krenchel. As for the tensile strength, the elastic modulus was only considered up to 40 wt.% (50 \( V_f \)) due to the descend of the experimental result after higher \( V_f \). For higher volume fraction than 0.4 there is a decline in tensile strength and elastic modulus from the experiment. It is due to the imperfect bonds between the fibres and matrix because of the approached maximum volume fraction of the fibre and stress-concentration points in fibre-rich area at higher \( V_f \) (Andre et al., 2016). The maximum theoretical volume fraction is 0.907 and 0.785 for hexagonal and square array fibre-packing geometries, respectively. That maximum value is achieved when the fibres diameter is uniform and the fibres are touching. When the fibres packing is non-regular and the fibres diameter is not uniform, the fibre volume fraction is difficult to achieve the value higher than 0.7 (Hull & Clyne, 1996). Furthermore, it is more problem for randomly oriented fibre composites. Hence, the validity of the micromodelling prediction for randomly oriented fibre composites is limited for the fibre content up to 0.4 \( V_f \).
Compared to the other theoretical results, the tensile modulus obtained by Manera was the highest in all volume fraction. This model deviated 40%, 35%, 29% and 36% at 0.14, 0.26, 0.38 and 0.5 V\textsubscript{f} from the experimental results, respectively. It is due to the fact that the LDPE matrix and zalacca fibre are not eligible by Manera, those are the E\textsubscript{m} ranges from 2 to 4 GPa and high aspect ratio (l/d > 300) of the fibres (Manera, 1977). The Manera’s model also does not consider the fibre length and orientation distribution.

The results of Cox–Krenchel’s model for tensile modulus deviated 20%, 18%, 14% and 23% at 0.14, 0.26, 0.38 and 0.5 V\textsubscript{f} from the experimental results, respectively. It is due to the neglect of voids. However, Cox–Krenchel’s model still recognizes the fibre length and orientation distribution factors (\(\eta_0\)), shear parameter, Poisson’s ratio of matrix and length of the fibre. In addition, there is the fibre orientation limit angle, \(\alpha_0\) in \(\eta_0\).

The Tsai–Pagano’s model resulted in the tensile modulus deviated by 3%, 1%, 0.9% and 12% at 0.14, 0.26, 0.38 and 0.5 V\textsubscript{f} from the experimental results, respectively. It indicates that the Tsai–Pagano’s model is the most accurate in predicting the elastic modulus of LDPE-ZF composites. It is due to the anisotropic characteristic in nature and aspect ratio of ZF considered in that model, meanwhile the Manera’s model only takes into account longitudinal elastic modulus.

The advantages of Tsai–Pagano’s over the Cox–Krenchel’s and Manera’s model are the existence of aspect ratio and the transverse elastic modulus of the fibres in the Halpin–Tsai’s equation for obtaining the longitudinal and transverse elastic modulus of composites. The Manera’s model calculates only the longitudinal modulus of fibre, meanwhile the Cox–Krenchel’s model takes only the length distribution and orientation of the fibres.

The proximity of the results of elastic modulus obtained by the experiment and model reveals that the hot press method for processing the LDPE-ZF composites is acceptable.

4. Conclusion

The addition of random oriented ZF to the LDPE matrix resulted in increased tensile strength and elastic modulus. However, the tensile strength of LDPE-ZF composites slightly decreased after the fibre loading of 10 wt.% (0.14 V\textsubscript{f}) but showed an increase in the higher V\textsubscript{f}. The highest tensile strength and elastic modulus were achieved at 40 wt.% (0.5 V\textsubscript{f}). The transverse modulus E\textsubscript{f2} of single ZF was obtained by entering the transverse modulus of the unidirectional LDPE-ZF composite with definite V\textsubscript{f} into the inverse rule-of-mixture. Hence, ZF is anisotropic with E\textsubscript{f1} and E\textsubscript{f2} values of 3.36 and 0.57 GPa, respectively. The comparative study between the tensile strength obtained
by experiment and selected model showed that the Bowyer–Bader’s model produced more accurate prediction due to its fibre length and orientation factors. Based on this anisotropic characteristic in nature and aspect ratio of ZF, the Tsai–Pagano’s model in relation to the Halpin–Tsai’s model has resulted in the excellent prediction of $E_z$. The concordance between the experimental and the micromechanical modelling of tensile strength and elastic modulus reveals that the hot press is suitable method for fabricating LDPE-ZF composites.

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Author details
Wahyu Purwo Raharjo1,2, E-mail: wahyupurwodaharjo@ft.uns.ac.id
Rudy Soenoko1, E-mail: rudysoen@ub.ac.id
Anindito Purnowidodo2, E-mail: anindito@ub.ac.id
Moch Agus Choiron2, E-mail: agus_choiron@ub.ac.id

Universitas Sebelas Maret, Surakarta, Indonesia.
1
Universitas Brawijaya, Malang, Indonesia.
2

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