Fabrication of Composite Material Using Gettou Fiber by Injection Molding*

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Abstract
This study investigated the mechanical properties of composite using gettou (shell ginger) fiber as reinforcement fabricated from injection molding. Gettou fiber is a natural fiber made from gettou, a subtropical plant that is largely abundant in Okinawa, Japan. We used the stem part of gettou plant and made the gettou fiber by crushing the stem. The composite using gettou fiber contributed to low shrinkage ratio, high bending strength and high flexural modulus. The mechanical strength of composite using long gettou fiber showed higher value than composite using short gettou fiber. Next, because gettou is particularly known for its anti-mold characteristic, we investigated the characteristic in gettou plastic composite. The composite was tested against two molds: aspergillus niger and penicillium funiculorum. The 60% gettou fiber plastic composite was found to satisfy the JISZ2801 criterion. Finally, in order to predict the flexural modulus of composite using gettou fiber by Halpin-Tsai equation, the tensile elastic modulus of single gettou fiber was measured. The tendency of the experimental results of composite using gettou fiber was in good agreement with Halpin-Tsai equation.

Key words: Gettou, Shell Ginger, Natural Fiber, Injection Molding, Composite, Anti-Mold, FRP, Halpin-Tsai Equation

1. Introduction
Traditionally, FRP (Fiber Reinforced Plastic) has widely used glass fiber in order to improve the mechanical properties. Composites using glass fiber are, however, difficult to dispose of because of their high strength, durability and flame retardancy. From an environmental standpoint, several studies have developed eco-friendly and green composites using natural plant fibers such as, bamboo, hemp, kenaf and ramie. We have reported a composite using bagasse fiber as filler and reinforcement. Baggage is a natural fiber obtained from sugarcane, one of the most important agriculture plants in Okinawa.

Similar in nature to sugarcane, gettou (Scientific name: alpinia speciosa, English name: shell ginger) is widely vegetated in subtropical regions such as Okinawa. Gettou is a perennial plant of the ginger family. Also, gettou is used in cosmetic products, health goods, and even as a wrapper when cooking traditional Okinawan rice cakes. A more significant use is its application in Japanese wall paper. Gettou is specifically applied, because its anti-mold characteristic keeps the paper fresh for a long period of time.

Focusing on the anti-mold characteristic of gettou and the possibility that it can be used as a reinforcing fiber, we decided to apply injection molding to find a highly effective use for gettou. Injection molding is the most used plastic forming process in industry, because it
allows for the mass production of complicated 3-D products. And, the use of injection molding to fabricate natural FRP is a growing market in the automotive industry. Also, we would like to point out that the possible anti-mold characteristic of gettou fiber plastics would be attractive to the plastic industry. This is because many plastic products are often exposed to wet and moist conditions.

The objective of this study was to report the mechanical properties of composite using gettou fiber by injection molding. Liner low density polyethylene (LLDPE) was chosen for the matrix. Also, an investigation of the anti-mold characteristic of composite composed of LLDPE and gettou fiber was evaluated. In addition, the tensile elastic modulus of single gettou fiber was measured in order to predict the flexural modulus of composite from the Halpin-Tsai equation. This was done to confirm the validity of the experimental results.

2. Experimental Materials and Methods

2.1 Material and injection molding

LLDPE was chosen as the matrix resin. The gettou stems were received from the Nihon Gettou Co., Ltd., Japan. Using a crushing machine, the stems were crushed and separated by screens into short and long fibers. Figure 1 shows the photograph of gettou fibers, and Table 1 shows the dimensions of gettou fibers. A twin type screw kneading machine was used to compound and pelletize the gettou fibers and LLDPE into different weight ratios of 10, 20, and 40%. Compounding temperature was set to 463K and held for 80 minutes. The pellets were used for an injection molding machine (Nissei Co. Ltd., PS10E1ASE). Injection molding conditions were at a pressure of 3.9 ~ 6.9MPa, a cylinder temperature of 403 ~ 413K, and a mold temperature of 313 ~ 333K. An injection molded product is shown in Fig. 2, the section within the dotted lines was cut and used for the test specimen.

2.2 Shrinkage ratio and bending test

The shrinkage ratio of the injection molded product was determined by comparing the measurements of the diameter taken from 3 points along the test specimen with the original mold dimensions. The average of 5 test specimens was measured.

A 3 point bending test was conducted in order to measure the bending strength and flexural modulus. The average of 3 test specimens was measured. The span was set to 50mm. The specimen length was 58mm, and the diameter of the specimen was 6mm. The crosshead speed was set to 0.5mm/min. The bending strength $\sigma_B$ was calculated from the maximum load $P$ using the following formula:

$$\sigma_B = \frac{8PL}{\pi D^3}$$

Fig. 1 Gettou fiber

(a) Short fiber (b) Long fiber
Here, $L_B$ is the span length, and $D$ is the diameter of the test specimen. The flexural modulus $E_B$ was calculated from the load–displacement curve obtained from the 3 point bending test. A tangential line was drawn on the linear section of the curve in the elastic zone. The flexural modulus $E_B$ was calculated using the following equation:

$$E_B = \frac{P}{48I} \times \frac{\Delta P}{\Delta \delta}$$

(2)

Here $I$ is the second moment of inertia, $\Delta P$ is the difference of the load on the tangential line, $\Delta \delta$ is the difference of the displacement on the tangential line, and $\Delta P/\Delta \delta$ is the tangential slope.

### 2.3 Anti-mold test

The anti-mold test for aspergillus niger and penicillium funiculosum was carried out according to the JIS Z2801 criterion\(^{(22),(23)}\). This criterion is a standard for determining the anti-mold properties of plastic surfaces. A value, known as the antimicrobial activity value, is given in this criterion. If the antimicrobial activity value is above 2.0, then the plastic is recognized to have anti-mold properties. The JISZ2801 test procedures are as follows: First, the specimen is prepared to the square size of 50mm x 50mm. Second, the mold liquid is adjusted with 1/500 normal bouillon, and dropped on the specimen surface. Third, the specimens are wrapped in a film, and stored at 308K. Finally, after 24 hours, the amount of mold on specimen surface is measured. The antimicrobial activity value is calculated using the following equation:

$$R = (U_t - U_0) - (A_t - A_0) = U_t - A_t$$

(3)

where $R$ is the antimicrobial activity value, $U_0$ is the average logarithm of the initial mold amount on the LLDPE only specimen, $U_t$ is the average logarithm of the mold amount after 24 hours on the LLDPE only specimen, and $A_t$ is the mold amount on the composite specimen after 24 hours. The anti-mold characteristic test and calculations for the antimicrobial activity value were carried out by the Kyoto Biseibutsu Kenkyusho.

The anti-mold specimen dimension is required to be 50mm x 50mm. For this reason, the anti-mold specimen was fabricated by hot press forming. The pellets used during hot
press forming were fabricated under the same conditions as the short gettou fiber pellets used during injecting molding. The hot press forming specimens contained a content weight ratio of 20% and 60% short gettou fiber. Also, the LLDPE only test specimen was made by hot press forming. The dimensions of the plate mold were 70mm x 130mm x 2mm. The hot press temperature was 413K. The holding time was 10 minutes, and the press pressure was 4.3MPa. After hot press forming, the specimens were cut to 50mm x 50mm x 2mm dimensions. The samples are shown in Fig. 3.

2.4 Tensile test for single fiber

In order to predict the flexural modulus from the Halpin-Tsai equation, the flexural modulus of gettou fiber needed to be measured. For this reason, a single fiber tensile test for gettou fiber was conducted referencing the Takagi et al. method\(^{(14,24)}\). The tensile test specimen for single gettou fiber is shown in Fig. 4. Before the experiment, the paper specimen after being fixed to the tensile test machine was cut along the line AB in Fig. 4. The tensile test was carried out at a crosshead speed of 0.5mm/min. The cross section of the gettou fibers was assumed to be circular, and the diameter was measured using optical microscope.

The tensile strength of gettou fiber was obtained by dividing the maximum load \(P\) by the cross sectional area \(A\)\(^{(14)}\). The cross sectional area \(A\) was calculated before the experiment from the diameter of measured fiber. The tensile elastic modulus \(E_T\) was determined from the load–displacement curve obtained from the tensile test. A tangential line was drawn on the linear section of the curve in the elastic zone. The tensile elastic modulus was calculated using the following

\[
E_T = \frac{\sigma_T}{\varepsilon_T} = \frac{L_T}{A} \times \frac{\Delta P}{\Delta \delta}
\]  

(4)

Here, \(L_T\) is the length of the gettou fiber specimen (\(L_T=10\)mm), \(A\) is the cross sectional area of the gettou fiber, \(\sigma_T\) is the stress of gettou fiber, and \(\varepsilon_T\) is the strain gettou fiber. Also, in Equation (4), \(\sigma_T\) is \(\Delta P / A\), and \(\varepsilon_T\) is \(\Delta \delta / L_T\).

![Fig.3 Specimen of anti-mold characteristic test](image)

(a) LLDPE  
(b) Composite

![Fig.4 Tensile test specimen for single gettou fiber](image)
3. Experimental Results and Discussion

3.1 Effect of gettou fiber on mechanical properties of composite

The properties of injection molded composite containing short gettou fiber were first examined. Injection molded products often use fillers to prevent shrinkage. Therefore, we investigated the shrinkage ratio. By varying the short gettou fiber content from 0 to 40%, shrinkage of composite was examined. Figure 5 shows the relationship between the shrinkage ratio and content of gettou fiber. The shrinkage ratio decreased by increasing the content of gettou fiber. Generally, the linear coefficient of expansion of polyethylene is $100 \sim 200 \times 10^{-6}/K^{(25)}$, and the linear coefficient of expansion of wood materials is $3 \sim 6 \times 10^{-6}/K^{(25)}$. For this reason, the coefficient of volumetric expansion of gettou fiber is considered to be lower than polyethylene. Therefore, gettou fiber was considered to act as a stopper to prevent shrinkage in the mold when the resin solidifies. Figure 6 shows the bending stress–displacement curve obtained from 3 point bending test. The flexural modulus and bending strength increased with increasing content of gettou fiber. Figure 7 shows the relationship between the flexural modulus and content of gettou fiber. The flexural modulus increased linearly as the content of gettou fiber was increased. Figure 8 shows relationship between the bending strength and content of gettou fiber. The results showed that the bending strength steadily increased until fiber content was 20%. Over 20%, the bending strength slightly increased. Figure 9 shows the cross section after 3 point bending test. We recognized that the crack bypassed the gettou fiber, because unbroken gettou fiber was observed in Fig. 9. Also, the crack propagation was stopped by gettou fiber at crack tip. From these results, gettou fiber plays a role preventing the crack propagation.

![Fig.5 Relationship between shrinkage ratio and content of short gettou fiber](image)

![Fig.6 Bending stress–displacement curve of short gettou fiber composite](image)
Next, in order to investigate the internal distribution of gettou fiber of injection molded product, the longitudinal section of each test specimen was cut and observed using an optical microscope. Figure 10 shows the longitudinal section ((a) 10%, (b) 20%, (c) 40%) of composite using short gettou fiber. The density of the distributed fibers increased with additional content of gettou fiber. Near the surface of injection molded product, direction of gettou fibers seems to be parallel with the injection direction. In the center, gettou fibers look to be inclined with injection direction. Figure 11 shows the orientation angle of short gettou fiber for injection direction. The orientation angle of short gettou fiber for injection direction was determined from Fig. 10. By increasing the content of gettou fiber, inclination gettou fiber for injection direction was increased. As shown in Fig. 9, gettou fiber acts in preventing the crack propagation. In each case (10%, 20%, and 40%) in Fig. 9, the crack propagated vertically through the longitudinal specimen. Therefore, the fibers directed parallel with the injection direction had a larger effect than the fibers inclined at an angle on the prevention of crack propagation. Thus, the bending strength increased slightly over gettou fiber 20%.

Finally, the effect of the gettou fiber length on bending strength and flexural modulus was examined. Two types of short and long gettou fiber composites at weight ratio of 40% were compared. Figure 12 and Fig. 13 show the comparison of the bending strength and flexural modulus of composite. These results show the long gettou fiber composite has a higher bending strength and flexural modulus compared with composite using short gettou.
fiber. In order to examine reason of the high bending strength of composite using long gettou fiber, the longitudinal section of the composite test specimen using long gettou fiber was cut and observed by optical microscope. The composite using 40% long gettou fiber is shown in Fig. 14. In general, reinforcement fibers in injection molded composites become orientated from the effects of resin flow within the mold. The shear flow velocity of resin in the mold is fast in the center, and slow near the surfaces. This difference in flow velocity within the mold produces rotational moments that act on the fibers. A longer span creates a larger rotational moment; thus, long fibers have larger rotational moments than short fibers. Because long fibers have larger rotational moments, they are considered to align with the injection direction. By comparing Fig.10(c) and Fig.14, it can be observed that the orientation of gettou fibers is different in the center sections. In the case of the composite containing short gettou fibers, a large number of gettou fibers can be seen to be inclined to the injection direction. As for the case of the composite containing long gettou fibers, the gettou fibers can be recognized to be aligned with the injection direction. Figure 15 shows the comparison of orientation angle for injection direction between short and long gettou fiber. The orientation angle of long gettou fiber for injection direction was determined from Fig. 14. In the case of the composite containing long gettou fiber, the frequency of the orientation angle from 0 to 15° in relation to the injection direction showed a higher value in comparison to the short gettou fiber. From this result, the bending strength of composite using long gettou fiber showed higher value than composite using short gettou fiber.
Fig. 10 Distribution of short gettou fiber of longitudinal section

Fig. 11 Orientation angle of short gettou fiber for injection direction
Fig. 12 Comparison of bending strength between short and long gettou fiber

Fig. 13 Comparison of flexural modulus between short and long gettou fiber

Fig. 14 Distribution of long gettou fiber of longitudinal section
3.2 Anti-mold characteristic of composite material

Table 2 shows the result of the anti-mold test and Fig.16 shows the relationship between the antimicrobial activity value and content of gettou fiber. The antimicrobial activity value of LLDPE only showed a value of about 0 for aspergillus niger and penicillium funiculosum. Also, the antimicrobial activity value of composite using gettou fiber 20% showed a value of about 1.0 for aspergillus niger and penicillium funiculosum. On the other hand, the composite using 60% gettou fiber had antimicrobial activity value over 2.0 for aspergillus niger and penicillium funiculosum. This value indicates the 60% gettou-plastic composite has anti-mold properties.

Figure 17 shows the observation of surface structure of test piece by optical microscope. Composite surface containing 20% gettou fiber is covered completely with resin. On the other hand, lots of gettou fibers are exposed on the surface of the composite using gettou fiber 60%. The similar exposing phenomenon using 66.6mass% bamboo powder was reported by Takagi et al. (5). Also, Kimura et al. (22) reported the antibacterial properties for staphylococcus aureus and escherichia coli of polypropylene (PP) containing silver-ion supported by zeolite. In Kimura et al. paper, the silver-ion supported by zeolite is antibacterial additive. Kimura et al. reported that silver-ion supported by zeolite is required.

<table>
<thead>
<tr>
<th>Content of Gettou fiber</th>
<th>Temporal change in amount of mold</th>
<th>Antimicrobial activity value</th>
</tr>
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<tr>
<td></td>
<td>Initial</td>
<td>24hrs-1</td>
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<tr>
<td>0</td>
<td>1.5×10^5</td>
<td>6.3×10^4</td>
</tr>
<tr>
<td>20</td>
<td>1.3×10^5</td>
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<tr>
<td>60</td>
<td>1.5×10^7</td>
<td>2.0×10^3</td>
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</table>

(b) Penicillium funiculosum

<table>
<thead>
<tr>
<th>Content of Gettou fiber</th>
<th>Temporal change in amount of mold</th>
<th>Antimicrobial activity value</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Initial</td>
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</tr>
<tr>
<td>0</td>
<td>1.5×10^5</td>
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<tr>
<td>20</td>
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<tr>
<td>60</td>
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</tbody>
</table>

*<10 Undetected
to be exposed on the specimen surface in order to have the antibacterial characteristic for PP. Thus, silver-ion supported by zeolite need direct contact with bacteria. The results of Fig. 17 suggest that the gettou fibers come in direct contact with the mold. Therefore, we could confirm that composite using gettou fiber 60% has anti-mold characteristic for aspergillus niger and penicillium funiculosum.

3.3 Verification of flexural modulus using Halpin-Tsai equation

In order to verify the flexural modulus, the experimental results were compared with the Halpin-Tsai equation. The Halpin-Tsai equation is as follows

\[
E_c = E_m \left(1 + \frac{\eta V_f}{E_f} \right), \quad \eta = \frac{(E_f/E_m) - 1}{(E_f/E_m) + \xi}, \quad \xi = \left(\frac{L_f}{D_f}\right)
\]  

(5)

where \(E_c\), \(E_m\) and \(E_f\) are the elastic modulus of composite, matrix, and fiber. \(L_f\) is the fiber length, and \(D_f\) is the fiber diameter. \(V_m\) and \(V_f\) are the volume fraction of matrix, and volume fraction of fiber, respectively. \(V_f\) was calculated using the following equation\(^{(15,16)}\)

\[
V_f = \left[ V - \left(\frac{W - W_f}{\rho_m}\right) \right] / V
\]  

(6)

where \(V\) is the volume of the specimen, \(W\) is the weight of the specimen, \(W_f\) is the weight of fiber, and \(\rho_m\) is the density of LLDPE. \(W_f\) was calculated from the weight ratio of gettou
fiber multiplied by the weight of specimen.

The results of the tensile elastic modulus and tensile strength in comparison to the fiber diameter are shown in Fig.18 and Fig. 19. Coefficient of correlation is -0.497 in Fig. 18, and -0.498 in Fig. 19, respectively. These results suggest that there is little relation during these factors. Figure 20 shows the relationship between tensile strength and tensile elastic modulus. The relationship between tensile strength and tensile elastic modulus is linear because coefficient of correlation is 0.797.

In order to predict the flexural modulus from the Halpin-Tsai equation, Eq. (5), the measurements of the $E_m$, $E_f$, $D_f$, and $L_f$ are required. Table 3 show the material constants used to calculated the Halpin-Tsai equation. The elastic modulus of LLDPE ($E_m$) was determined from the result of the bending test. The elastic modulus of gettou fiber ($E_f$) was determined from the result of the single fiber tensile test. The diameter ($D_f$) and length ($L_f$) were measured from gettou fibers contained within the composite specimens. The measurements of the fibers were taken from the longitudinal section.

In Fig.21, the flexural modulus predicted by the Halpin-Tsai equation is compared with the experimental results. From the results, the experimental flexural modulus is slightly higher than that predicted by the Halpin-Tsai equation. In previous studies, Shibata et al.\cite{15,16} has investigated the flexural modulus in kenaf and bagasse fiber injection molded composites. These studies have shown that cross section of natural fibers within injection molded composites underwent compression, and the tensile elastic modulus of the fiber within the composites increased. Considering Shibata et al. studies, the results in Fig.21 suggest that the gettou fiber within the composite is reinforced by compression during injection molding process. Therefore, the experimental results showed a higher value than that predicted by the Halpin-Tsai equation. However, the overall tendency of the experimental result and Halpin-Tsai equation was in good agreement, and the validity of the experimental results was confirmed. In addition, as a result of the comparison of the experimental results and Halpin-Tsai equation, it can be considered that the tensile elastic modulus of gettou fiber increased by compression of the cross section of gettou fibers that occurred during injection molding. This detail consideration will be discussed in our future aspect.

![Fig.18 Relationship between tensile elastic modulus and diameter of single gettou fiber](image-url)
Fig. 19 Relationship between tensile strength and diameter of single gettou fiber

Table 3 Material constants used for Halpin-Tsai equation

<table>
<thead>
<tr>
<th>Material</th>
<th>Elastic modulus (MPa)</th>
<th>Diameter (mm)</th>
<th>Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLDPE</td>
<td>253</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Gettou fiber</td>
<td>3175</td>
<td>0.063</td>
<td>1.13</td>
</tr>
</tbody>
</table>

Fig. 21 Comparison between experimental result and Halpin-Tsai equation
4. Conclusion

Composite material composed of gettou fiber reinforced LLDPE was fabricated by injection molding. By changing the fiber content and fiber length of composite, mechanical properties and anti-mold characteristic were investigated. The obtained results are shown as follows:

1. The additional content of gettou fiber decreased the shrinkage ratio. Also, the bending strength and flexural modulus increased. The 40% composite showed a bending strength of 19.8MPa, and flexural modulus of 1.1GPa. Therefore, gettou fibers played a role as reinforcement in composite material. The composite using long gettou fiber showed a bending strength of 21.3MPa, and flexural modulus of 1.3GPa. The long gettou fiber results of bending strength and flexural modulus were higher in comparison to composite using short gettou fiber.

2. The anti-mold test showed that the antimicrobial activity value for the LLDPE was about 0 for both aspergillus niger and penicillium funiculosum. By increasing the content of gettou fiber, the antimicrobial activity value increased. Particularly, the composite containing 60% gettou fiber showed an antimicrobial activity value over 2.0 (JISZ2801 criterion). Therefore, it was found that the composite using gettou fiber of 60% has an anti-mold characteristic.

3. The experimental values of flexural modulus were slightly higher than that predicted by Halpin-Tsai equation. However, as a whole the tendency of experimental result and calculation predict from Halpin-Tsai equation were in good agreement. From the experimental results and calculation of Halpin-Tsai equation, it was obvious that gettou fiber played a role as reinforcement.

Acknowledgment

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References

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