1 Introduction

Continuous fiber reinforced thermoplastic composites (c-FRTP) have been attractive due to the recycle ability and possibility of secondary processing [1-2]. However, the impregnation of thermoplastic resin into fiber bundle is difficult because of the continuous fiber and the high melt viscosity of the matrix. In this study, in order to improve difficulty of impregnation, fibrous intermediate materials of carbon fibers and PA fibers were developed by using commingle technique. This intermediate material enables the continuous fiber reinforced thermoplastic composite to be fabricated with low cost and short molding time; therefore the application field of fibrous intermediate material for c-FRTP becomes wide [3].

Commingled ratio affects impregnation state and mechanical property of composites. In this paper, the dispersion ratio of fibers in intermediate material is represented as commingled ratio. Dispersion ratio was measured by changing the sizing contents in carbon fiber and the production process. The effects of dispersion ratio were discussed in terms of the strength of intermediate material and impregnation states of the composite. The mechanical properties of unidirectional CF/PA composite were evaluated by tensile test.

2 Experiments

2.1 Intermediate materials

Fig. 1 shows the commingled machine used in the production of commingled yarn. Reinforcing fibers and resin fibers were sent out with each servomotor creel. Sent fibers were mixed with commingled unit. Then, the commingled yarn was completed by winding device. Commingled yarn was produced, in which the volume fraction (Vf) of reinforcing fiber and resin fiber was about 50%, respectively. Fig. 2 shows the crank structure in the commingled machine. Reinforcing fiber bundle was spread by physical oscillation of the crank.
Two types of carbon fibers (T700-12000-60E, T700-12000-50C, 800tex, TORAY) were used as the reinforcing fiber. The carbon fibers had a different amount of sizing content; 60E was 0.2wt% and 50C was 1.1wt%. The PA resin fiber (Leonar-235T36B, PA66, 235dtex, melting temperature; 265°C, Asahi Kasei Fibers) was used as the matrix resin. Table 1 and Table 2 show the properties of reinforcing fiber and resin fiber. Carbon fibers and PA fibers were commingled in commingled machine. Specification of the specimens was shown in Table 3. Commingled yarns were prepared by changing sizing contents of carbon fiber and a crank stroke for spreading carbon fiber bundle. The crank stroke was changed in 3 types (0, 6, 30mm).

Table 1. Properties of carbon fiber.

<table>
<thead>
<tr>
<th>Carbon fiber</th>
<th>Sizing content (wt%)</th>
<th>Filament number</th>
<th>Filament diameter (µm)</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60E</td>
<td>0.2</td>
<td>12000</td>
<td>6.9</td>
<td>1.8</td>
</tr>
<tr>
<td>50C</td>
<td>1.1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Property of resin fiber.

<table>
<thead>
<tr>
<th>Resin fiber</th>
<th>Melting temperature (°C)</th>
<th>Filament number</th>
<th>Filament diameter (µm)</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PA66</td>
<td>265</td>
<td>3760</td>
<td>12.3</td>
<td>1.14</td>
</tr>
</tbody>
</table>

Table 3. Type of specimens.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>No.1</th>
<th>No.2</th>
<th>No.3</th>
<th>No.4</th>
<th>No.5</th>
<th>No.6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sizing content</td>
<td>0.2wt%</td>
<td>1.1wt%</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stroke (mm)</td>
<td>0</td>
<td>6</td>
<td>30</td>
<td>0</td>
<td>6</td>
<td>30</td>
</tr>
</tbody>
</table>

2.2 Dispersion ratio

Dispersion ratio is very important factor for impregnation property. In order to enable the high cycle molding, it is necessary to reduce the impregnation distance as much as possible. In this study, the impregnation distance was examined by dispersion ratio. If dispersion ratio is high, impregnation distance becomes short.

In order to quantitatively evaluate the dispersion state of intermediate material, dispersion ratio was measured as shown in Fig. 3. The intermediate material was inserted into heat-shrinkable tube. Deflating the tube by heating, fibers were densely packed and formed cylindrical shape. The tube was embedded into a resin, and then cross-sectional observation with optical microscope was performed. Radial lines were drawn by every 30 degree on the cross-sectional photograph of fiber bundle with circle shape. There was agglomeration of reinforcing fiber or resin fiber on each radial line. The overlapping length of $a_i$ and $b_i$ which reinforcing fiber and resin fiber continuously exist on each radial line was measured. Dispersion ratio was expressed by equation (1).

$$\text{Dispersion ratio (\%)} = 100 \left(1 - \frac{1}{n} \left(\frac{\sum a_i + \sum b_i}{\sum a_i + \sum b_i} \right)^2\right)$$

where $n$ is the total number of measurement. The value that average length of $a_i$ and $b_i$ divided by total length of both fibers was calculated. Each dispersion ratio was defined by subtracting the value from 1. When the reinforcing fibers and resin fiber are completely separate like parallel yarn, dispersion ratio is 50% of the minimum value. When the filament of reinforcing fibers and resin fibers are arranged alternately, for the filament diameter of the carbon fiber and resin fiber with 6.9, 12.3 (µm), respectively, dispersion ratio is 99.1% of the maximum value. In this way, as average length of fiber agglomeration become small, dispersion ratio comes close to 100%, and the minimum value is 50%.
2.3 Strength of intermediate material

The strength of intermediate material that was damaged by commingle-process was investigated by tensile test of intermediate materials. Intermediate material with 300mm length was clipped by paper tab at the both ends and span length between both paper tabs was 200mm. In this study, fiber strength was calculated by dividing max load by total cross sectional area of carbon fibers.

2.4 Impregnation state and mechanical property of composites

In order to investigate the impregnation state of molding, unidirectional composite was fabricated. Fig. 4 shows the fabrication method of unidirectional composite. Fiber bundles were wound 12 times unidirectional-aligned on metal flame. It was molded by heating and compression molding method. Molding pressure was 3MPa, molding time was 5min, and molding temperature was 290 °C. Finally, unidirectional plate (20mm in width, 200mm in length) was molded and the cross-sectional observation and the tensile test were carried out. Un-impregnation ratio of composites was measured by using image analysis. And tensile test was carried out under the conditions of 200mm length, 100mm distance between the grips, the test speed of 1mm/min by using INSTRON(Type4206).

3 Results

3.1 Dispersion ratio and fiber strength

Fig. 5 shows cross-sectional photograph of the fiber bundle. The dark-colored fibers indicate resin fiber and the light-colored fibers indicate carbon fiber. Degree of dispersion of carbon fiber with 0.2wt% was higher than that with 1.1wt%. Results of dispersion ratio and fiber strength are shown in Table 4. No. 3 could not be prepared because carbon fiber was tangle on the commingled machine. Dispersion ratio of 0.2wt% was increased with increasing the crank stroke, whereas dispersion ratios of 1.1wt% with 0mm and 6mm were almost constant and that with 30mm was slightly increased. Dispersion ratio of 0.2wt% was larger than that of 1.1wt% because spreading carbon fiber bundle was inhibited due to sizing treatment. Fiber strengths of 0.2wt% and 1.1wt% with 0mm and 6mm were almost same. But that of 30mm with 1.1wt% was the lowest. While increasing a crank stroke increased dispersion ratio, the carbon fiber was damaged by long stroke. As a result, commingled yarn of 0.2wt% with 6mm had both high dispersion ratio and high fiber strength.
3.2 Impregnation state and mechanical property of composites

Fig. 6 shows cross-sectional photograph of composites. The light-colored area indicates carbon fiber, and the dark-colored area indicates resin rich region. And the black area within the dotted line indicates un-impregnation area. Un-impregnation area was confirmed in both 0.2wt% and 1.1wt%. A tendency of decrease in un-impregnation area with increasing dispersion ratio was confirmed.

Fig. 7 shows relationship between un-impregnation ratio and dispersion ratio. For both 0.2wt% and 1.1wt%, increasing dispersion ratio decreased un-impregnation ratio because the impregnation distance was decreased by increasing dispersion ratio.

Fig. 8 shows relationship between elastic modulus and un-impregnation ratio. Elastic modulus was increased with decreasing un-impregnation ratio. Fig. 9 shows relationship between tensile strength and un-impregnation ratio. As with elastic modulus, tensile strength was increased with decreasing un-impregnation ratio. In order to increase the mechanical properties of composites, un-impregnation ratio should be decreased. However, tensile strength of 1.1wt% with 30mm (No.6) (the plot indicated by arrow at Fig.8) was decreased from the trend line because fiber strength of No.6 was greatly decreased. Therefore, if fiber damage was inhibited, impregnation state and mechanical property were improved only by increasing dispersion ratio.

Table 4. Dispersion ratio and fiber strength.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Sizing content</th>
<th>Crank stroke (mm)</th>
<th>Dispersion ratio (%)</th>
<th>Fiber strength (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.1</td>
<td>0.2wt%</td>
<td>0</td>
<td>62.5</td>
<td>1.9</td>
</tr>
<tr>
<td>No.2</td>
<td></td>
<td>6</td>
<td>79.6</td>
<td>2.0</td>
</tr>
<tr>
<td>No.3</td>
<td></td>
<td>30</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>No.4</td>
<td>1.1wt%</td>
<td>0</td>
<td>53.4</td>
<td>1.7</td>
</tr>
<tr>
<td>No.5</td>
<td></td>
<td>6</td>
<td>52.2</td>
<td>1.7</td>
</tr>
<tr>
<td>No.6</td>
<td></td>
<td>30</td>
<td>59.4</td>
<td>1.0</td>
</tr>
</tbody>
</table>
DEVELOPMENT AND PROCESSING OF INTERMEDIATE MATERIAL FOR CONTINUOUS FIBER REINFORCED THERMOPLASTIC COMPOSITES

4 Twisting processing of commingled yarn

4.1 Fabrication method

The commingled yarn of this study has the following problems; when the composites were molded, the orientation of the reinforcing fiber bundle becomes misaligned because of the different thermal shrinkage rate and fineness between the resin fiber and reinforcing fiber. And the commingled yarn has low handling ability because sizing agent was lost by commingled process. In this study, twisting processing was applied to commingled yarn in order to solve these problems. The effects of twisting number on impregnation property and mechanical property of unidirectional composites were discussed.

As a material, commingled yarn with lower damage and the highest dispersion ratio shown in previous chapter was used. This commingled yarn was produced with 6mm crank stroke and 0.2wt% sizing content (No.2). 6 types of (0, 10, 30, 50, 100, 200 T/m) twisting processing was performed by using ring twister.

Fig. 10 shows photograph of twisted commingled yarn. The apparent fineness of commingled yarn was decreased with increasing number of twist. Fig. 11 shows relationship between twist angle and number of twist. Twist angle was increased with increasing number of twist and increasing rate of twist angle decreased gradually.

Fig. 7. Relationship between un-impregnation ratio and dispersion ratio.

Fig. 8. Relationship between elastic modulus and un-impregnation ratio.

Fig. 9. Relationship between tensile strength and un-impregnation ratio.

Fig. 10. photograph of twisted commingled yarn
4.2 Experiments

4.2.1 Impregnation state and mechanical property of composites

Unidirectional composites were fabricated by using twisted commingled yarn. And the impregnation state and the mechanical property of the composites were investigated by same method as previous chapter.

4.2.2 Orientation of fiber bundle

As mentioned in the paragraph 4.1, when the composites with non-twisted commingled yarn were molded, the orientation of the reinforcing fiber bundle becomes misaligned because of the different thermal shrinkage rate and fineness between the resin fiber and reinforcing fiber. The misalignment was possible to decrease the mechanical properties of the composites. Fig. 12 shows surface of specimens with non-twisted commingled yarn. In this study a fiber bundle oriented to the loading axis was referred to as “On-axis fiber bundle”, whereas misaligned fiber bundle was referred to as “Off-axis fiber bundle”. In order to quantitatively evaluate the orientation of the reinforcing fiber bundle on composites with twisted commingled yarn, surface of the composites was observed. “Off-axis fiber bundle ratio (OAR)” was measured by using image analysis. OAR was defined as the ratio of the surface area occupied by Off-axis fiber bundle and was calculated by dividing the area of Off-axis fiber bundle by the surface area of composite. OAR was measured at three times for each composite and averaged.

Off-axis fiber bundle

On-axis fiber bundle

Fig. 12 Surface of specimens with non-twisted commingled yarn

4.3 Results

Fig. 13 shows cross-sectional photograph of specimens by changing number of twist. Reinforcing fiber bundle became circular with increasing number of twist and resin rich regions were also increased up to 50 (T/m). Fig. 14 shows relationship between un-impregnation ratio and number of twist. Un-impregnation ratio was decreased with increasing number of twist up to 50 (T/m), whereas that was increased over 50 (T/m). The reason for reduction in un-impregnation ratio up to 50 (T/m) was the effect of the molding pressure. So, it was considered that the molding pressure could be homogeneously applied to the fiber and the impregnation of thermoplastic resin into fiber bundle was improved. The reason for increase in un-impregnation ratio over 50 (T/m) was the effect of the Vf in fiber bundle. Carbon fiber was unspreaded by twisting and then the Vf in fiber bundle was increased and the flow of resin to reinforcing fiber bundle was inhibited.

Fig. 15 shows surface of specimens. The orientation of the fiber bundle was improved by twisting processing. Fig. 16 shows relationship between OAR and number of twist. While OAR was greatly decreased with increasing number of twist under 50 (T/m), that was gradually decreased with increasing number of twist over 50 (T/m). Therefore, twisting Processing of about 50 (T/m) for commingled yarn was effective to inhibit that the orientation of the carbon fiber bundle was misaligned. From these results, while non-twisted commingled yarn was inhomogeneously arranged on die, twisted commingled yarn was arranged at equal spaced intervals on die.

Fig. 17 shows relationship between elastic modulus and number of twist. While elastic modulus was almost same under 50 (T/m), that was decreased
DEVELOPMENT AND PROCESSING OF INTERMEDIATE MATERIAL FOR CONTINUOUS FIBER REINFORCED THERMOPLASTIC COMPOSITES

with increasing number of twist over 50 (T/m). Fig. 18 shows relationship between tensile strength and number of twist. While tensile strength was linearly increased with increasing number of twist under 50 (T/m), that was greatly decreased with increasing number of twist over 50 (T/m). Twisting processing of commingled yarn was expected to decrease the mechanical property of the composites due to the effect to an increase in the fiber orientation angle for the yarn axis. However, under 50 (T/m), tensile strength was increased with increasing number of twist because of increase in both impregnation state and the orientation of the fiber bundle. But mechanical properties of the composites were decreased with increasing number of twist over 50 (T/m) because the effect of the orientation of the fiber orientation angle was dominant. From these results, if number of twist was under 50 (T/m), it was considered that the effects of the impregnation state and the orientation of the fiber bundle were greater than the effect of the fiber orientation angle. Therefore, it was clarified that the twisting of commingled yarn with optimum twisting number was effected for increasing the mechanical properties of composites. And non-twisted commingled yarn was low handling ability. So commingled yarn was needed surface processing such as sizing agent for improving handling ability. However, This surface processing may make interfacial adhesion between reinforcing fiber and thermoplastic resin worse. Thus, it was considered that twisting processing was also effective in term of handling ability in addition to improving of the impregnation property and the mechanical properties.
Conclusions

In this study, new intermediate materials of carbon fibers and PA fibers were developed by using commingled technique. And the dispersion ratio of fibers in intermediate material is represented as commingled ratio. The damage degree of the reinforcing fiber in commingled process was evaluated as fiber strength. These were affected by changing the sizing contents in carbon fiber and crank stroke for spreading carbon fiber bundle. The effects of dispersion ratio and fiber strength were discussed in terms of the strength of intermediate material and impregnation states of the composite. Moreover, in order to improve the orientation of the reinforcing fiber bundle and the handling ability, the commingled yarn of high fiber strength and the highest dispersion ratio was twisted. And the effects of twisting number on impregnation property and mechanical property of unidirectional composites were discussed.

As results, in the materials used in this study, it was revealed that commingled yarn, used carbon fiber of 0.2wt% sizing content, produced 6mm crank stroke was high dispersion ratio and high fiber strength. And high dispersion ratio and high fiber strength of commingled yarn produced both decrease in un-impregnation ratio and increase in mechanical property of the composites. Moreover, twisting processing under 50 (T/m) increased the mechanical property by improving the impregnation property and the orientation of the fiber bundle of the composites.

Therefore, in order to increase dispersion ratio for improving impregnation state, carbon fibers
with small amount of sizing agent and crank stroke both to inhibit fiber damage and increase dispersion ratio should be selected. Moreover, it was clarified that the twisting of intermediate material with optimum twisting number was effected for increasing the processability and the mechanical properties of composite.

Reference