

Effect of Cooling Rate on the Mechanical Strength of Carbon Fiber-Reinforced Thermoplastic Sheets in Press Forming

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Effect of cooling rate on the mechanical strength of carbon fibre reinforced thermoplastic sheets in press forming

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Abstract

The purpose of this study is to elucidate the effect of the cooling rate of the carbon fibre reinforced thermoplastic (CFRTP) sheets on the mechanical property in the press forming within 1 min cycle time. In order to pay attention only to the compression stage after the deformation stage in press forming, a flat sheet of dimensions 200 mm \times 100 mm \times 3 mm was produced. It was fabricated by stacking 15 CFRTP sheets of 0.2 mm-thick plain woven fabric impregnated with PA6, preheating them to 280 °C and pressing them at 5 MPa using a die cooled from near the melting temperature of PA6 with various cooling rates. Cooling rate of -26 °C/s with pressure holding time (defined in this study as the period that the pressure sensor detects high pressure) of 7 s and that of -4.4 °C/s with pressure holding time of 18 s gave a flexural strength of 536 MPa and 733 MPa, respectively. It was found that the cooling rate during pressure holding is related to the mechanical property of press-formed CFRTP part.

1. Introduction

Carbon fibre reinforced thermoplastics (CFRTPs) are expected to be used as structural components for applications such as automobiles because of their potential for achieving a short processing time as the resin can melt, and then, solidify, upon heating and cooling, respectively [1]. CFRTPs reinforced by continuous fibres such as woven fabrics are difficult to deform owing to their anisotropic deformation behaviour. However, they show very high mechanical properties compared to CFRTPs fabricated using discontinuous fibres; therefore, forming technologies using CFRTP with continuous fibre have been studied.

Unlike the resin transfer moulding method utilized for thermoset CFRPs, the high viscosity of the molten thermoplastic resin makes it difficult to impregnate the tiny spaces between the fibres [2]. Some forming methods have been developed and studied to overcome this, such as; forming method using commingled yarn [3], in-situ polymerization method [4], and the combination of film stacking and diaphragm forming [5]. However, the material stays in the die long period owing to the impregnation time.

In this study, we deal with press forming method which enables short processing time. Press forming is a method where the fabrication of the CFRTP sheets by impregnation and the forming are separated. CFRTP sheets to be press formed are fabricated by methods such as film stacking or double belt press to impregnate the resin into the reinforcing fibres [6-8]. The CFRTP sheets fabricated are preheated above the melting temperature, and then they are press formed and cooled using the die.

Press forming consists of two stages. (1) Deformation stage to deform the molten CFRTP sheets into the desired shape. (2) Compression stage to apply pressure to the deformed CFRTP sheets. The CFRTP sheets are cooled in compression stage.

Two forms of materials can be used in press forming. (a) A multi-layered consolidated CFRTP sheet whose thickness is the same as that of the press-formed part. (b) Single layer CFRTP sheets that are stacked to obtain the desired thickness of the press-formed part and are consolidated when they are press-formed in the die.

Both consolidated CFRTP sheets and single CFRTP sheets are available in the market; however, the quality of impregnation or the mechanical properties are not specifically guaranteed for the provided materials. In addition, preheating before press forming causes de-bonding of the interface between the fibre and the resin or interlayers [9,10]. Therefore, in compression stage of press forming, the pressure and temperature condition are important for the void content and the mechanical properties of the press-formed part.

The effect of the die temperature on the mechanical strength after forming was studied [11-13]. The relationship between the forming pressure and the mechanical strength has been studied [14, 15]. The effect of the temperature of material on the mechanical strength has been studied [16]. The effect of the pressure on the internal structure after pressing has been investigated [17, 18]. Most of these studies were performed with slow cooling rate of the die or long pressing time. Therefore, the effect of pressure and temperature in short cycle time has not been investigated sufficiently.

The purpose of this study is to elucidate the relationship between the cooling rate of the CFRTP sheet in compression stage and the mechanical property of the formed part in press forming within 1 min cycle time. The cycle time in this study is the period from where the heated CFRTP sheets are placed in the die to where the removal of the part. The attention is paid only to the compression stage. In addition, the pressure in the die and the temperature in the CFRTP sheets in compression stage under various cooling rates are analysed and the relationship between those factors and the mechanical strength are discussed.

2. Experiment

2.1 Experimental procedure

Fig. 1 shows the experimental process and the items to be investigated in this study. The CFRTP sheets were stacked for 15 layers and they were preheated by a halogen heater. The sheets were then transferred to the lower die as quickly as possible and they were pressed under various cooling rates of the CFRTP sheets. The cooling rate was determined by the combination of the die temperature before press forming (heated below the melting temperature of the matrix) and the temperature of the cooling plate. The power of the cartridge heaters in the die was kept off during pressing so that the die and the CFRTP sheets cools rapidly. The flat sheet with 200 mm × 100 mm × t3 mm was obtained.

During pressing, the temperature in the sheets are recorded by the thermocouples, and the pressure sensor in the lower die records the pressure in the die. The relationship between the cooling rate and the flexural strength of the consolidated flat sheet is investigated. The sheet temperature and the pressure during pressing are investigated under various cooling rates.

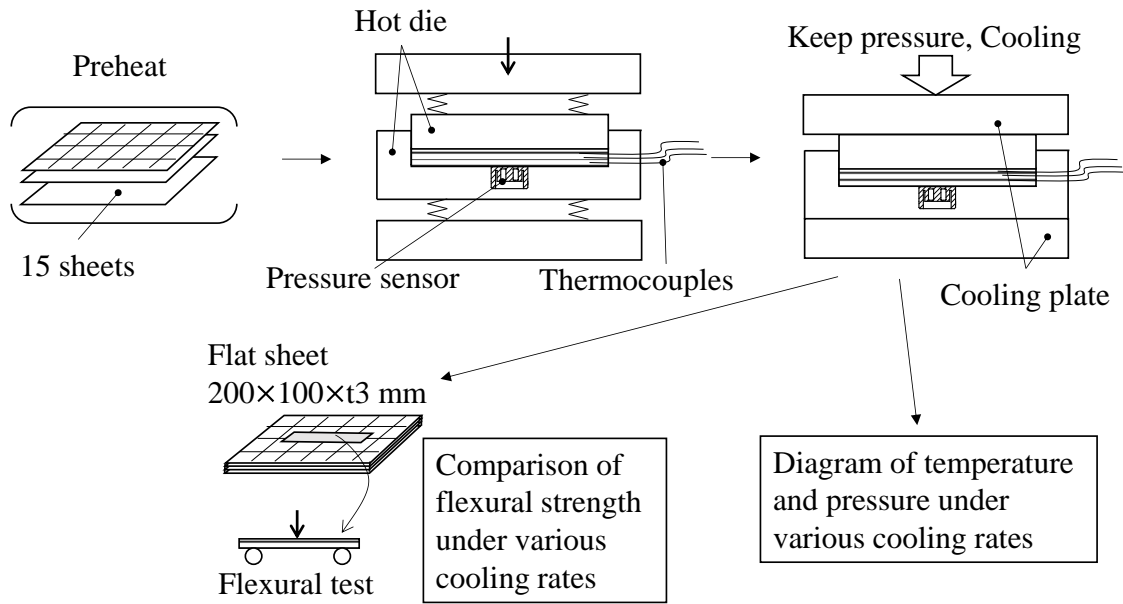


Fig.1 Experimental process and investigation items

2.2 Experimental condition

The experimental conditions are shown in Table 1. The cooling rate of the CFRTP sheets in the die were $-26\text{ }^{\circ}\text{C}$, $-6.6\text{ }^{\circ}\text{C}$ and $-4.4\text{ }^{\circ}\text{C}$. The cooling rate was determined by the combination of the initial die temperature (die temperature before pressing) and the cooling water temperature in the Table 1.

The pressure and the pressing time were; 5 MPa (press load of 100 kN divided by the area of the flat laminate $200\text{ mm} \times 100\text{ mm}$) and 25 s (the period that the press machine applies the press load to the die in compression stage), respectively.

The reasons why 5 MPa was used as the press forming pressure are: (a) previous experiments showed that a pressure of 5 MPa resulted in a part with higher mechanical strength compared to parts fabricated at lower pressures. (b) To prevent a much greater size and power of the press machine compared to the size of the part, so that the part can be produced with low energy consumption.

The pressing time of 25 s was determined because it should be less than 30 s in order to achieve the 1 min cycle time of the press forming process, including transfer, die closing, die opening and removal of the part.

The thickness of the composite component is generally in the range of 1 to 3 mm. The reason we chose a 3-mm-thick laminate in this experiment is because we have estimated that the effect of cooling rate of the 3-mm-thick laminate can also be applied to the press forming of thinner laminates.

Table 1 Experimental condition

Cooling rate of sheet	Initial die temperature	Cooling water temperature
-26 °C/s	Room temperature (about 25 °C)	30 °C
-6.2 °C/s	170 °C	30 °C
-4.4 °C/s	220 °C	80 °C

2.3 CFRTP sheet

The CFRTP sheet used in the experiments was a plain woven carbon fibre impregnated with PA6 (Ichimura Sangyo CO., LTD.). The volume fraction of the fibre is 50%, the thickness is 0.2 mm and the dimensions are 200 mm × 100 mm. The inner structure and the surface of the CFRTP sheet are shown in Fig. 2. The resin is impregnated in the fibre bundles. The space where the fibre bundles cross is filled with the resin. The resin does not fully cover the surface of the CFRTP sheet and some fibres are exposed to air. The fibres are oriented in 0/90 direction. The sheets are stacked in the same direction to investigate the basic laminate sequence.

The reason for consolidating CFRTP sheets in press forming is that it is easier to prepare the laminates with variety of fibre directions than using consolidated multi-layered CFRTP sheets. The flat sheet after press forming is shown in Fig. 3

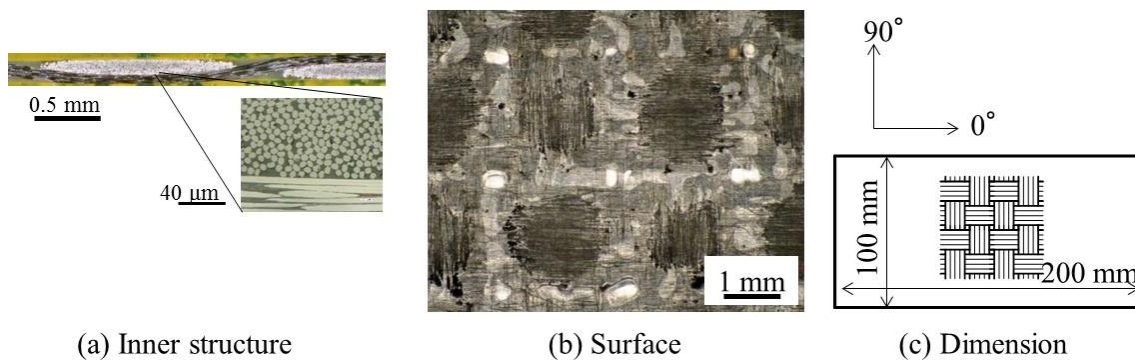


Fig. 2 CFRTP sheet

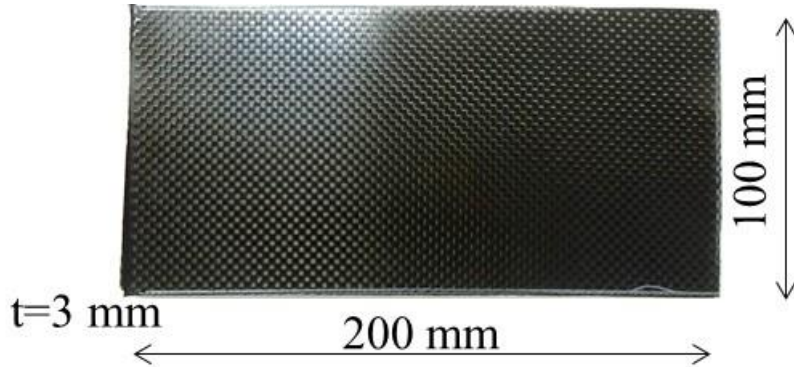


Fig. 3 Flat sheet after press forming

2.4 Die

The appearance of the die is shown in Fig. 4. The lower die is surrounded by a vertical wall to create a space of dimensions $200\text{ mm} \times 100\text{ mm}$, where the preheated CF RTP sheets are placed. The cartridge heaters are inserted in the die. The channels for the cooling water are drilled in the cooling plate.

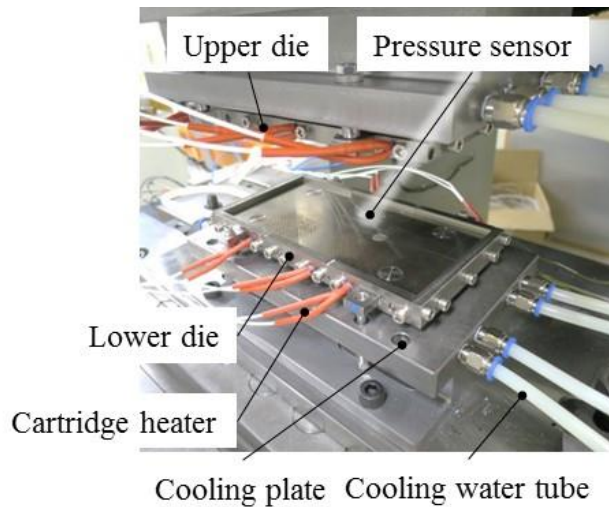


Fig. 4 Appearance of the die

The pressure sensor originally developed, shown in Fig. 5, was inserted in the lower die. The pressure detection area of the sensor is $\varnothing 8\text{ mm}$. Four strain gauges, which are fitted at the bottom of the lower thin plate, are connected with the bridge circuit. The elastic deformation of the lower thin plate caused by the pressure on the pressure detection area is detected by the strain gauges and translated into the pressure.

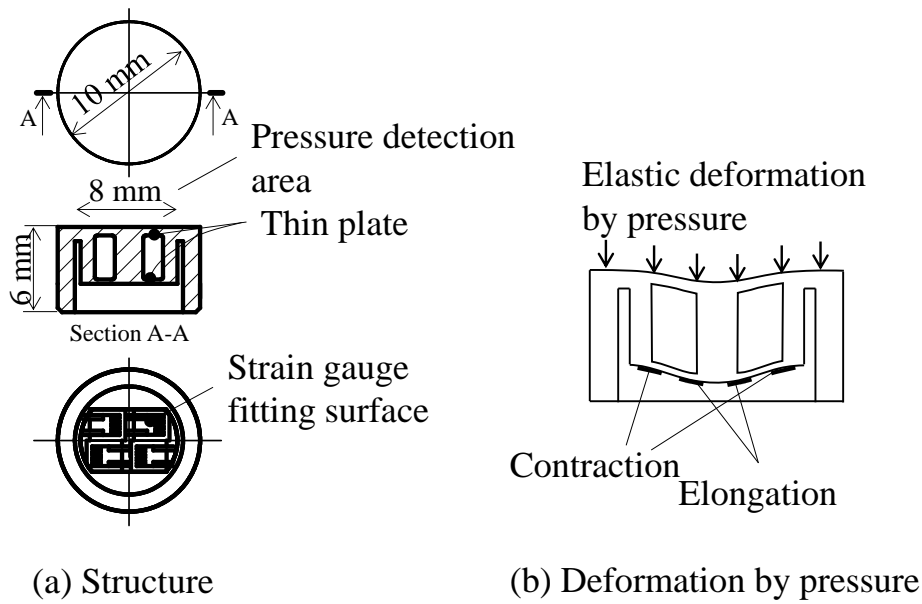


Fig. 5 Pressure sensor structure

A mechanical servo press H1F45 (Komatsu Industries Corp.) was used for press forming. The slide stroke is 100 mm and the maximum press load is 450 kN. Generally, unlike hydraulic presses, the mechanical servo presses do not maintain a constant press load during press forming. Therefore, press load decreases owing to the thermal shrinkage of the resin during cooling. To avoid this, a load control program that maintains the target press load was installed in the press machine. The press load was recorded in the data collection system of the press machine.

3. Results

3.1 Temperature change during preheating

The temperature curve of the CFRTP sheets during the preheating is shown in Fig. 6. The temperature in the diagram is shown from 100 °C because the recording was started 10 s after the onset of preheating. The temperature of the 2nd and the 14th layers increased rapidly for 20 s, and then, increases gradually. The temperature of the 8th layer increases relatively slowly and linearly. The 8th layer become 280 °C after 100 s. On the other hand, the 2nd and the 14th layers are at 360 °C and 340 °C, respectively after 100 s. Within the 10 s of transferring, the 2nd and the 14th layers become 240 °C and the 8th layer become 260 °C. The temperature curves in several experiments showed almost the same trend.

The temperature difference in the thickness direction of the CFRTP sheets during

the preheating is caused by the heat transfer where the temperature of the surface layers, which increase first, is transferred to the middle layer. During the transfer to the die, while the temperature of the surface layers drops sharply, the cooling of the middle layer is slow because of the thermal insulation caused by the piled up layers on both the top and the bottom of the middle layer.

The forming temperature of PA6 is 230 °C to 260 °C. The temperature of the CFRTP sheets is above 230 °C when the die is closed. This indicates that the pressure is applied at the forming temperature.

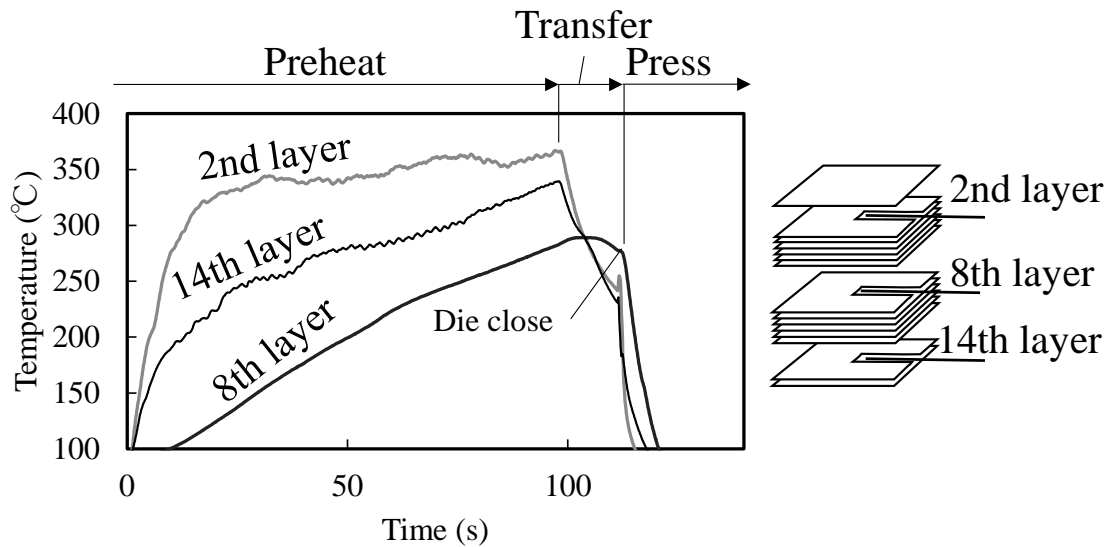


Fig. 6 Sheet temperature curve during preheat

3.2 Press load, sheet temperature, die temperature and pressure during pressing

The typical press load curve during pressing is shown in Fig. 7. The zero of the time in the diagram is when the press slide starts descending. The press load is maintained at 100 kN for 25 s. Every experiments were performed with this press load curve.

The temperature curve of the sheet and that of the die and the pressure curve detected by the pressure sensor are shown in Fig. 8. The temperature of the CFRTP sheets was recorded only once for each press condition. Pressure, die temperature and press load were recorded 5 times at each press condition.

The cooling rate were defined as the temperature gradient between 280 °C to 150 °C of 8th layer.

With the cooling rate of -26 °C/s (Fig. 8 (a)), the pressure increases up to 7 MPa with the increase in the press load, and then drops at 7 s. When a drop in the pressure

occurs, the 8th layer is at 160 °C. With the cooling rate of -6.6 °C (Fig.8 (b)), the pressure reaches 9 MPa, and then, drop after 13 s. When the pressure drops, the 8th layer is at 180 °C. With the cooling rate of -4.4 °C (Fig.8 (c)), the pressure reaches 9 MPa, and then, drop after 18 s. When the pressure drops, the 8th layer is at 190 °C. The trend of the pressure curve was the same for 5 repetitions of the experiments.

The pressure decrease after the first peak is likely caused by the thermal shrinkage of the resin. The pressure drop a few seconds after the first peak occurs when the 8th layer is at 190 °C. The molten PA6 solidifies with some degree of crystallization at 180 °C–190 °C when it is cooled [19]. Therefore, it is assumed that the deformation of the pressure detection area of the pressure sensor is relaxed because the volume of the resin shrinks when it is solidified, and hence, the pressure dropped. Owing to the load control program of the press machine, the press load applied to the entire area of the CF RTP sheets is kept constant.

To investigate the influence of the temperature on the pressure sensor, the examination was performed by heating the silicone rubber sheet in the 220 °C die, and then, cooling it at 5 MPa for 25 s; however, the value of the pressure sensor was constant. Therefore, the pressure drop shown in Fig. 8 is caused by the change in the state of the resin.

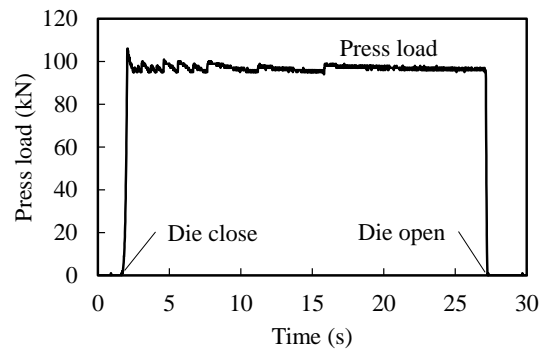


Fig. 7 Typical press load curve during pressing

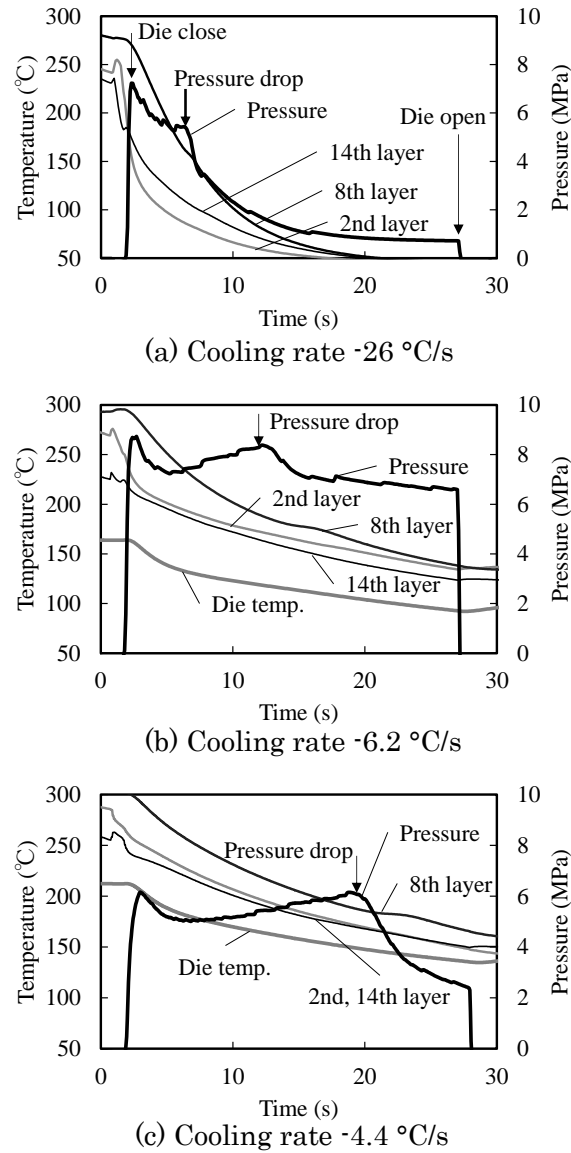


Fig. 8 Sheet and die temperature curve and pressure curve in different cooling rate

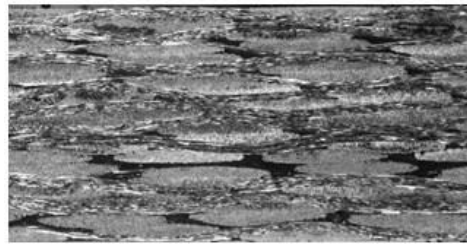
3.3 Internal structure

The internal structures of the flat sheet are shown in Fig .9. The interlayer is bonded well and voids were not observed either in the fibre bundles or the interlayer at every cooling rate. The process of consolidating the stacked CFRTP sheets in press forming can provide a composite laminate without voids.

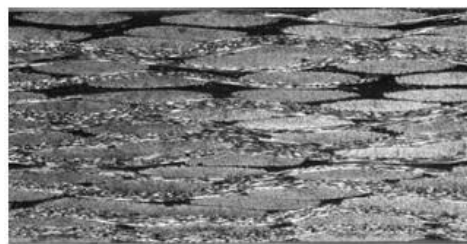


(a) Cooling rate $-26\text{ }^{\circ}\text{C/s}$

1 mm



(b) Cooling rate $-6.6\text{ }^{\circ}\text{C/s}$



(c) Cooling rate $-4.4\text{ }^{\circ}\text{C/s}$

Fig. 9 Comparison of internal structure with cooling rate

3.4 Flexural test

3.4.1 Flexural load curve

The three point bending test (based on JISK7074) was conducted on a specimen with dimensions of $100\text{ mm} \times 15\text{ mm}$ cut from the flat sheet. The testing speed was 3.6 mm/min and the span length was 80 mm . The surface touched lower die was the elongation surface. The typical load-stroke curve out of five samples are shown in Fig. 10.

As the cooling rate decreases, the rigidity and the maximum load increases, and the specimens are fractured in a more brittle manner. It is assumed that the low cooling rate caused better bonding between the fibre and the resin, consequently the load on the fibres increased.

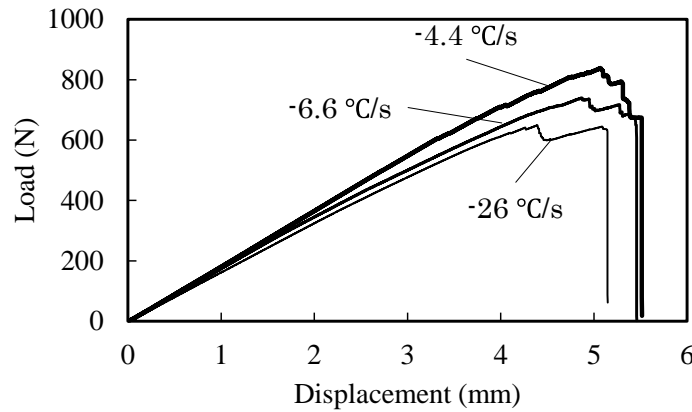


Fig. 10 Comparison of typical flexural load curve with cooling rate

3.4.2 Flexural strength

The flexural strengths calculated using the maximum testing load in Fig. 10 are shown in Fig. 11. The average value of 5 specimens is shown.

With the cooling rate of $-26\text{ }^{\circ}\text{C/s}$, the flexural strength is 536 MPa, which is the weakest compared to the others. With the cooling rate of $-6.6\text{ }^{\circ}\text{C/s}$, the flexural strengths is 631 MPa. With the cooling rate of $-4.4\text{ }^{\circ}\text{C/s}$, the flexural strength is 733 MPa. The cooling rate of the sheet affects the flexural strength.

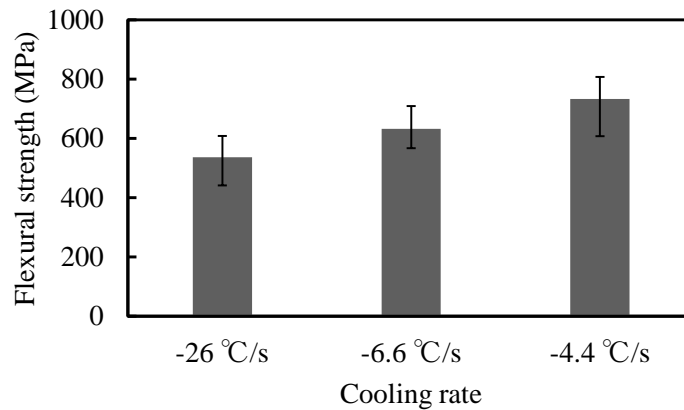


Fig. 11 Comparison of flexural strength with cooling rate

3.5 Relationship between flexural strength and pressure holding time

The relation between the flexural strength and the “pressure holding time” is shown in Fig. 12. The “pressure holding time” is defined in this study as the period from when the pressure of the pressure sensor shows the first peak to when it drops.

As the cooling rate decreases, the pressure holding time becomes longer. In addition, the flexural strength and the pressure holding time have a proportional relation.

The reason why the low cooling rate increases the flexural strength of these relationships can be estimated as follows. The low cooling rate keeps the resin in the molten state for a longer time. The pressure forces the molten resin to flow in the reinforcing fibre bundles and in between the stacked layers. Owing to that, the molecular chains entangle with the fibres well, and more molecular chains between the layers get entangled with each other [20], consequently, improving the bonding between the fibre-resin and the interlayers.

Another possible reason for this is the influence of the degree of crystallization. A slower cooling rate increases the degree of crystallization of PA6 [21].

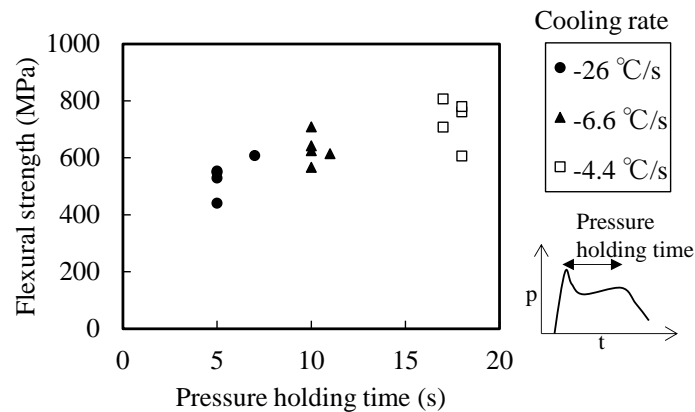


Fig. 11 Relationship between flexural strength and pressure holding time

4. Discussion

4.1 Temperature and pressure condition

For selecting the press machine and for achieving a short cycle time, the pressure and the pressing time were chosen as 5 MPa and 25 s, respectively, as described in section 2. To achieve a press forming cycle time within 1 min, the time consumed for the pressing and cooling would be 30 s, excluding the time taken to place the sheets in the die, to close and open the die and to remove the part from the die.

With the cooling rate of $-26\text{ }^{\circ}\text{C/s}$, and that of $-4.4\text{ }^{\circ}\text{C/s}$, the pressure holding times were 7 s and 18 s, respectively, and the flexural strength achieved was 536 MPa and 733 MPa, respectively. The pressure holding time was estimated to correspond to the period that the molten resin is solidified.

4.2 Die design to achieve the optimum cooling rate

There are two stages in press forming. (1) Deformation stage of the CFRTP sheets to follow the configuration of the die. Deformation includes changing the crossing angle of weft and warp, bending in the out-of-plane direction and interlayer slip. (2) Compression stage after the deformation stage. Compression stage includes compaction of fibre-resin and the interlayers. In the deformation stage, the die should be at high temperature so that the fibre-resin and the interlayers slip with low friction. On the other hand, in the compression stage, the die temperature should be controlled at a certain cooling rate to achieve both high mechanical property and the short cycle time.

In order to cool the molten CFRTP sheets to 190 °C in 18 s, as shown in Fig. 8 (c), the die surface should be cooled at a cooling rate of -3.9 °C/s ((150 °C–220 °C)/18 s).

Running the cooling medium in the cooling channel near the surface of the die is one of the methods to cool the die. In the case of cooling a die surface having an intricate configuration such as a sphere or a hat section using this cooling rate, a channel for the heat transfer medium near the die surface, a combination of heating and cooling medium or a combination of surface heating and cooling are required.

4.3 Press machine to achieve the pressure process

Owing to the need for maintaining a constant press load during pressing the CFRTP, hydraulic presses are generally used in the press forming of CFRTP. However, this study has demonstrated that a mechanical servo press can also provide the CFRTP part with high mechanical strength by using the load control program. The pressure in this study maintained a constant pressure of 5 MPa during the pressing. Moreover, a pressure that varies according to the state of the resin during the pressing will be desired in the future. The preheated CFRTP sheets lose their temperature quickly during the transfer; therefore, the die should be closed as quickly as possible. The high speed slide motion of the mechanical servo press benefits the press forming of CFRTP.

4.4 Influence of CFRTP sheet quality

The CFRTP sheets used in the experiments have a good impregnation quality. When using CFRTP sheets with an insufficient impregnation quality, an additional pressing time is required so that the molten resin can impregnate the fibre bundles; therefore, the pressing time would be more than 25 s. On the other hand, even if the CFRTP sheets have a perfect impregnation quality with a flat surface, pressure has to

be applied at least until the resin at the boundary of the adjacent layers entangles each other and solidifies.

Even if other thermoplastic resins are used for the matrix, pressing with relatively slow cooling rate would also provide a part with high mechanical strength.

The experiments demonstrated in this study is useful to find the optimum press forming conditions for CFRTP sheets with a different impregnation quality, or for CFRTP sheets with other matrix resins.

If the laminate sequence has a quasi-isotropic design, the valley-to-valley gap of the adjacent CFRTP sheets may increase because the geometric arrangement of the peaks and valleys caused by the woven pattern is different in adjacent layers. Therefore, pressing time would be longer to cause the resin to flow sufficiently in the interlayers.

4.5 Internal structure

The press-formed flat laminates have no visible voids in their internal structure for all cooling rates, as shown in Fig. 9. However, the flexural strength shown in Fig. 11 indicates that it differs with the cooling rate. This indicates that the factor that determines the flexural strength cannot be observed at the magnification shown in Fig. 9, but microscale phenomena such as the bonding degree between the fibre-resin, interlayer, and the degree of crystallization of the resin determines the flexural strength.

4.6 Consolidating CFRTP sheets in press forming

Consolidating stacked CFRTP sheets in press forming has an advantage that a variety of laminate sequences can be achieved in the lab or in the manufacturing factories without impregnating facilities. For example, a hybrid laminate consisting of CFRTP and glass fibre reinforced thermoplastic can be produced, and a tailored blank sheet that has a thickness distribution in the laminate can also be achieved. Thick laminates that cannot be produced with a double belt press can be achieved by stacking a large number of CFRTP sheets and press forming them. Thick laminates can also be achieved by stacking the multi-layered CFRTP sheets produced by double belt press and press forming them. The knowledge of the temperature and pressure conditions obtained in this study can also be applied to the production of such thick laminates.

5. Conclusions

The effect of the cooling rate in press forming on the mechanical property of the CFRTP was investigated.

A cooling rate of $-4.4\text{ }^{\circ}\text{C/s}$ under 5 MPa gives a flexural strength of 733 MPa for the flat laminate. Parts with high mechanical strength were achieved by consolidating CFRTP sheets in press forming less than 1 min cycle time.

In case of the press forming involves deformation of the CFRTP sheets, the temperature and pressure conditions found in this study can be applied to obtain the part with high mechanical property.

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