

## IN-SITU LASER ASSISTED DIRECT IMPREGNATION, LAMINATION AND PLACEMENT OF CFRTP

Tadashige Ikeda<sup>1</sup>, Takeharu Sakai<sup>2</sup>, Ayako Yokosu<sup>3</sup>, Yasunari Fujii<sup>3</sup>, Hiroki Takenami<sup>3</sup>,  
Masahiro Arai<sup>3</sup> & Takashi Ishikawa<sup>4</sup>

<sup>1</sup> Department of Astronautics and Aeronautics, Chubu University, Japan

<sup>2</sup> Department of Engineering, Tottori University, Japan

<sup>3</sup> Department of Aerospace Engineering, Nagoya University, Japan

<sup>4</sup> National Composite Center, Nagoya University, Japan

### Abstract

To develop lower-cost and higher-strength carbon fiber reinforced thermoplastic (CFRTP), in-situ laser assisted direct impregnation, lamination and placement method has been proposed, which is referred to as automated fiber placement direct (AFP-D). A prototype model of the AFP-D system consists of a diode laser, a compaction roller, air cylinder and a linearly movable stage. A thermoplastic film and a spread carbon fiber (CF) tow are independently supplied into the AFP-D system. The diode laser is used to locally heat the CFs. The heated CFs melt the thermoplastic film and are impregnated with the melted thermoplastic resin via a compaction roller. The impregnated CFs are directly laminated on already laminated CFs on a mold. This paper shows fundamental analysis of the AFP-D manufacturing process and its demonstration. In the analysis it was confirmed that the spread CF tow with a thickness of 35 $\mu$ m could be completely impregnated with PA6 resin within 0.06s at a possible temperature and pressure of the prototype AFP-D system. Since the pressing period would be 0.15s at a feed speed of 30mm/s in the experiment, the CF tow could be impregnated completely. Then, the demonstration of the AFP-D system was carried out. The CF tow seemed to be able to be impregnated with the PA6 resin completely and laminated. Bending strength of the fabricated specimens was higher than the ones prepared by a conventional heat press method. This shows feasibility of the proposed AFP-D method.

**Keywords:** CFRTP, automated fiber placement, laser, direct processing, impregnation

### 1. Introduction

Carbon fiber reinforced plastic (CFRP) came to be used as primary structures of commercial airplanes because of their high specific strength and corrosion resistance. CFRP used in the airplane structures are mainly made from laminated thermoset preregs. They are laid up by an automated tape/fiber placement system and then are consolidated by heating while vacuuming and pressing them in an autoclave with an adequate size fitted to the products. Accordingly, the manufacturing process of the CFRP is time-consuming and the products are limited in size and costly. Therefore, development of a low-cost, high-cycle, and less-size-limitation manufacturing technology is one of the challenges for expanding application fields of the CFRP. To overcome these issues, an in-situ automated fiber/tape placement (AFP/ATP) technology of manufacturing a three-dimensional carbon fiber reinforced thermoplastic (CFRTP) body has been studied [1-4]. Unlike the above mentioned autoclave manufacturing methods, a unidirectional CFRTP prepreg tape is laminated on a mold by locally heating, pressing and consolidating it in this method. This in-situ lamination by using the AFP method can reduce the overall process time of CFRTP components and has less-size-limitation without the autoclave of a closed container.

By using a laser as a heat source, instead of a hot gas torch used for the conventional AFP, a broader range of processing speeds can be possible because the laser provides a high power heat source with good on-off performance. This aspect can be effective to shape a more complicated three-dimensional component.

Qureshi et al. [1] examined the relationship between mechanical properties and process parameters

such as compaction force, tape feed speed and tool temperature for a CF/PEEK tape. In this study strength of the samples processed by the current AFP with a hot gas system as a heat source was also compared to those processed with the state-of-art AFP with a laser system. It was found that the samples processed by the hot gas AFP achieved 55% interlaminar shear strength of the samples prepared by the autoclave while the samples processed by the laser AFP achieved 85% strength of the autoclave ones. Comer et al. [2] also compared the mechanical properties of CF/PEEK samples processed by a laser AFP and an autoclave. The interlaminar shear strength of the laser AFP samples was 70% of the autoclave ones. To optimize the AFP process H  l  non et al. [3] performed finite element analysis. In the fine element model, slit tapes were pressed by a compression roller and tape thickness and gap width between the tapes after the compression were calculated. Stokes-Griffin and Compston [4] showed that temperature history of a material of the resin during the subsequent tape placements is important for the mechanical properties of the composite. The temperature must increase beyond the melting point and keep less than the pyrolytic temperature. The lower placement rate showed better fiber-matrix adhesion and less ductile failure, since the lower placement rate experiences a large cumulative time in the crystallinity temperature range during the placements. Although engineering and academic research on the AFP has been carried out as described above, that is still on the way.

The conventional AFP uses the slit prepreg tapes, which are harder than dry tows. Accordingly, it is not easy to steer the prepreg tapes and tape puckers may arise at high curvature region [5]. Since the puckers flatten by application of subsequent plies, local crimps may occur and reduce strength of the composite. Furthermore, the slit prepreg tapes are inevitably more costly than the raw materials.

To improve the steering property of the CFRTP tape and reduce the cost of the CFRTP products, an in-situ laser assisted direct impregnation, lamination and placement method has been developed [6], which is referred to as automated fiber placement direct (AFP-D). In this method, the costly CFRTP prepregs are not used but dry CF tows easier to be steered and a thermoplastic film are used. A prototype model of the AFP-D system consists of a diode laser, a compaction roller, air cylinder and a linearly movable stage. A thermoplastic film and a spread dry CF tow are independently supplied into the AFP-D system. The diode laser is used to locally heat the CFs. The heated CFs melt the thermoplastic tape and are impregnated with the melted thermoplastic resin via the compaction roller. The impregnated CFs are directly laminated on already laminated CFs on a mold.

Section 2 describes the prototype model of the proposed AFP-D system. In Section 3 fundamental analytical study of the AFP-D manufacturing process shows feasibility of the system. In Section 4 effects of the process parameters on impregnation are investigated and the specimens fabricated by the AFP-D system are compared in the bending strength and modulus with the specimens fabricated by using a conventional heat press method.

## 2. Prototype Model of AFP-D System

The AFP-D system developed in the present study is shown in Fig. 1. This prototype system mainly consists of a diode laser (LDF, 3000-100, Laserline GmbH), a compaction roller and a linearly movable stage. The specification of the system is summarized in Table 1. Laser beam is delivered to the compaction roller through an optical fiber and a zoom optics equipment. The zoom optics equipment arranges the shape of the laser beam to be rectangular, variable from  $4 \times 4 \text{ mm}^2$  to  $38 \times 38 \text{ mm}^2$ , and the spatial distribution of its intensity to be uniform. An incident angle of the laser beam can be controlled manually. The compaction roller device mainly consists of a steel roller with a diameter of 30 mm and an air cylinder. The air cylinder presses the roller to the stage. The stage moves linearly by 200 mm with a speed of 1 to 300 mm/s driven by a linear actuator. A rubber sheet (Kalrez   7075UP, DuPont) with a thickness of 1mm is wrapped around the compaction roller to expand a contact region between the roller and a specimen which is referred to as the nip length. The rubber sheet is fixed with a polyimide film covered. The nip length of the compaction roller increased to 4.0 to 5.0 mm with the rubber sheet wrapped from 1.5 to 1.8mm without the rubber sheet, where a pressure sensitive paper (Prescale 4LW, Fujifilm) was used to measure the nip length.

A non-oriented PA6 film (DIAMIRON   C-Z, thickness 25  $\mu\text{m}$ , Mitsubishi Plastics) and a spread CF tow (15K, TR-50S, Mitsubishi Rayon, 24 mm width, SAKAI OVEX) were used in the present study. Because the thickness of the spread CF tow was about 35  $\mu\text{m}$ , it was expected that a time for resin

impregnation could be shortened. Long strips with a width of 20 mm were cut out from the PA6 film and the spread CF tow. The two strips and a polyimide tape were rolled up around a bobbin. The polyimide film was used to easily release the PA6 from the compaction roller. A prepreg sheet (TC910 PA6, Tencate) was fixed on the stage as a base substrate. The strips were delivered to the compaction roller from the bobbin and irradiated by the laser. The spread CF tow was heated by the laser and melts PA6. It is impregnated with the melted PA6 and laminated on the substrate by being pressed with the compaction roller. The CF-PA6 laminate was cooled down and consolidated in the nip region. Although the polyimide tape must be removed after every placement in this experiment, the tape would not be used in practical application. The subsequent fiber placement was done in a similar way. As described above the impregnation, lamination and consolidation are done in-situ directly in this proposed AFP-D method.

Feasibility of the proposed AFP-D method was examined for several process parameters listed in Table 2. The values followed by an asterisk were assumed as control values.

Table 1 – Specifications of the prototype model of AFP-D system.

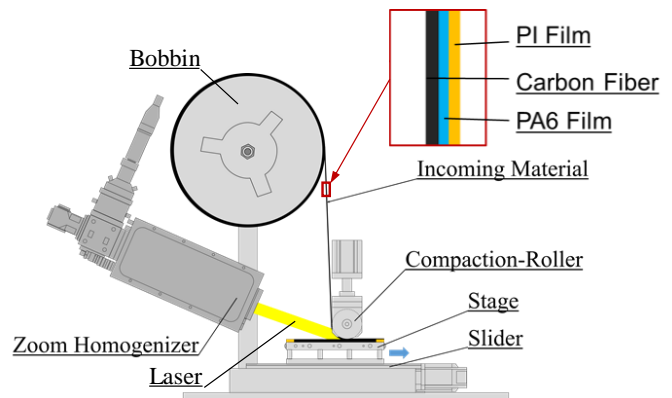
Specification		Value
Diode laser	Power	100~3000 W
	Minimum beam quality	20 mm·mrad
	Wavelength	900~1070 nm
	Focal distance	200 mm
	Beam shape	4×4~38×38 mm <sup>2</sup>
Air cylinder	Press force	40~ 784 N
Compaction roller	Material	Stainless steel
	Diameter	30 mm
Stage	Speed	1-300 mm/s

Table 2 – Examined process parameters.

Process parameter	Values
Lase power	160W, 180 W*, 200W
Laser irradiation angle	25°*, 30°, 35°
Rubber sheet	With* or without
Sizing agent removal treatment	Without* or with
Roller press force	784N
Stage speed	30 mm/s



(a)



(b)

Figure 1 – Prototype model of AFP-D system.

(a) Photograph and (b) schematic diagram of the AFP-D system.

### 3. Fundamental Analytical Study of AFP-D Manufacturing Process

Before the AFP-D experiment, fundamental study of the AFP-D manufacturing process was performed to show feasibility of the AFP-D system. Figure 2 shows the measured and the calculated temperature of the CF spread tow heated by the diode laser at a power density of  $1.39 \times 10^5 \text{ W/m}^2$  ( $200 \text{ W}/(38 \times 38 \text{ mm}^2)$ ) for a period of 0.3s. The reflectivity was obtained by the ray tracing analysis, which value was 0.23 to 0.27 for 0.4 to 0.6 of fiber volume fraction,  $V_f$ . The value of  $V_f$  was  $0.52 \pm 0.08$  from SEM (scanning electron microscope) observation of the CF tow. It is seen from the figure that the calculated temperature is in good agreement with the experiments and that the temperature increases to approximately 600K (327°C), which is higher than the melting temperature of the PA6 (225°C).

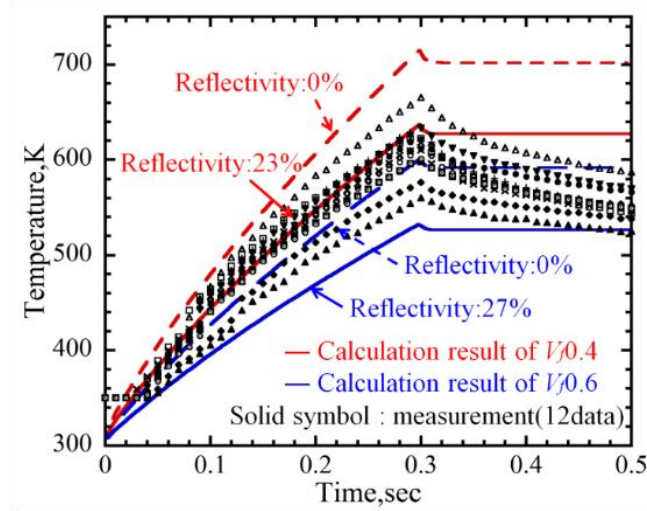


Figure 2 – Temperature of carbon fibers heated by a diode laser.

Then, impregnation of the PA6 into the CF tow was examined analytically by assuming that thermoplastic resin flow was isothermal and incompressible. By using the one-dimensional mass conservation equation and the equation for Darcy's law, an equation for the depth of the resin impregnation,  $x_f$ , can be obtained as follows [7]:

$$x_f = \sqrt{\frac{2K_{xx}(P_e - P_v)t}{(1 - V_f)\mu}} \quad (1)$$

where  $K_{xx}$ ,  $\mu$ ,  $P_e$  and  $P_v$  denote the permeability, the viscosity, the pressure applied to the resin and the pore pressure, respectively. The value of the permeability was calculated with the following formula [8] by specifying the diameter of CF,  $\phi$ , and  $V_f$ .

$$K_{xx} = \frac{(1 - V_f)^3 \phi^2}{96V_f^2} \quad (2)$$

The temperature of the melted PA6 was assumed to be a constant value at 523 K and the value of the viscosity was set to be 100 Pa.s. It is noted that the viscosity varies depending on not only temperature but also shear strain rate, that was not considered here. The value of the applied pressure was calculated from the press force 784N and the area of the test specimen used in the experiment  $20 \times 4.5 \text{ mm}^2$ . The pore pressure was taken to be an atmospheric pressure 0.1MPa. The fiber diameter was assumed to be  $7 \mu\text{m}$ . The material constants and the process parameters are summarized in Table 3. Figure 3 shows calculated aspect of the impregnation. When the value of  $V_f$  is assumed to be between 0.4 and 0.6, the CF spread tow with a thickness of  $35 \mu\text{m}$  is completely impregnated with the PA6 within 0.06s. Since the pressing period is 0.15s when the nip length is assumed to be 4.5mm and the feed speed to be 30mm/s, the tow could be impregnated with the PA6 completely. It is noted that wettability between a fiber and a viscous fluid affects the impregnation. The CF tow was coated with a sizing agent supposed to be adequate for PA6. The effect of the sizing agent will be mentioned in Sections 4.4 and 4.5.

Table 3 – Material constants and process parameters for analysis.

Material constant/ process parameter	Symbol	Value
Viscosity of melted PA6	$\mu$	100 Pa·s
Applied pressure	$P_e$	8.71 MPa
Pore pressure	$P_v$	0.1MPa
Diameter of CF	$\phi$	7 $\mu$ m

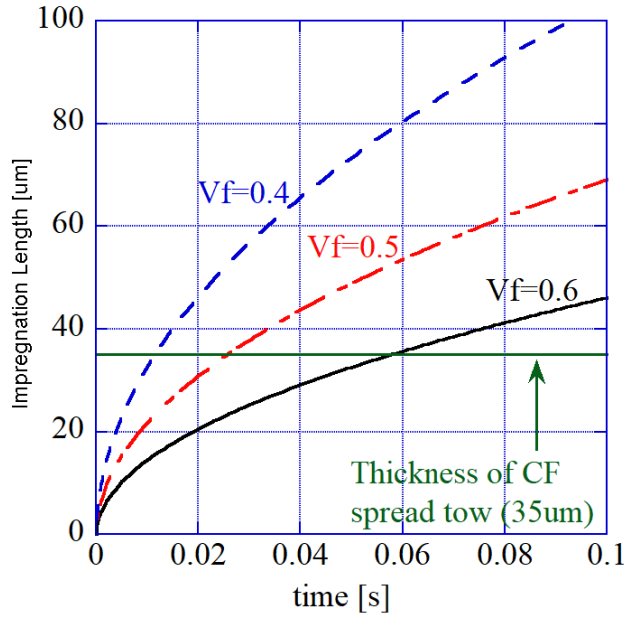


Figure 3 – Impregnation analysis.

## 4. Experimental Results and Discussions

At first, effect of process parameters of the AFP-D listed in Table 2 on impregnation is examined by observing cross-sectional images. Then, strength and stiffness of the specimens fabricated by the AFP-D are evaluated by comparing to the specimens fabricated by a heat press method.

### 4.1 Effect of Rubber Sheet

Effect of the rubber sheet around the compaction roller expanding the nip length on the impregnation was examined. The process parameter values listed in Table 2 with the asterisk were used for parameters other than the rubber sheet. Figures 4(a) and 4(b) show cross-sectional images of the specimens fabricated without and with the rubber sheet, respectively. One can see the prepreg base sheet, the first CF-PA layer and the second CF-PA layer from the bottom.

It is seen that there is almost no void and the CF tows are impregnated with the resin in the specimen fabricated with the rubber sheet shown in Fig. 4(b). While one can see the cracks and the CF tows are hardly impregnated in the specimen fabricated without the rubber sheet shown in Fig. 4(a). This difference comes from that the rubber sheet can expand the nip length and press nearly uniformly against even somewhat uneven surface of the CF tow.

### 4.2 Effect of Laser Power

Figures 5(a) to 5(e) show the cross-sectional images of the specimens fabricated at a laser power of 160 to 200 W, respectively. It can be seen from Fig. 5(a) that the resin can not be melted well at 160 W and the CF tows can not be impregnated with the resin. We can see the crack-like voids even in the specimen fabricated at 170 W. For the specimens fabricated at the laser powers more than



180W, the CF tows are well impregnated with the resin in the first and second CF-PA6 layers. However, voids appear in the base layer for the specimens fabricated at 190 and 200W. Therefore, the laser power of 180W is the best of the five powers in this experiment.

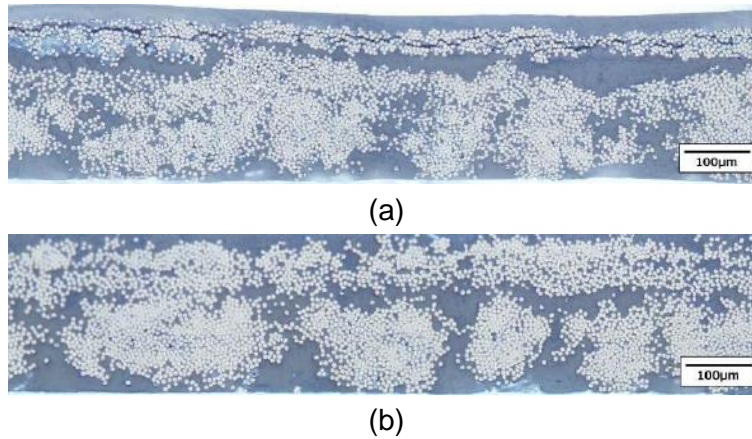


Figure 4 – Cross-sectional images of the specimens fabricated (a) without and (b) with the rubber sheet around the compaction roller.

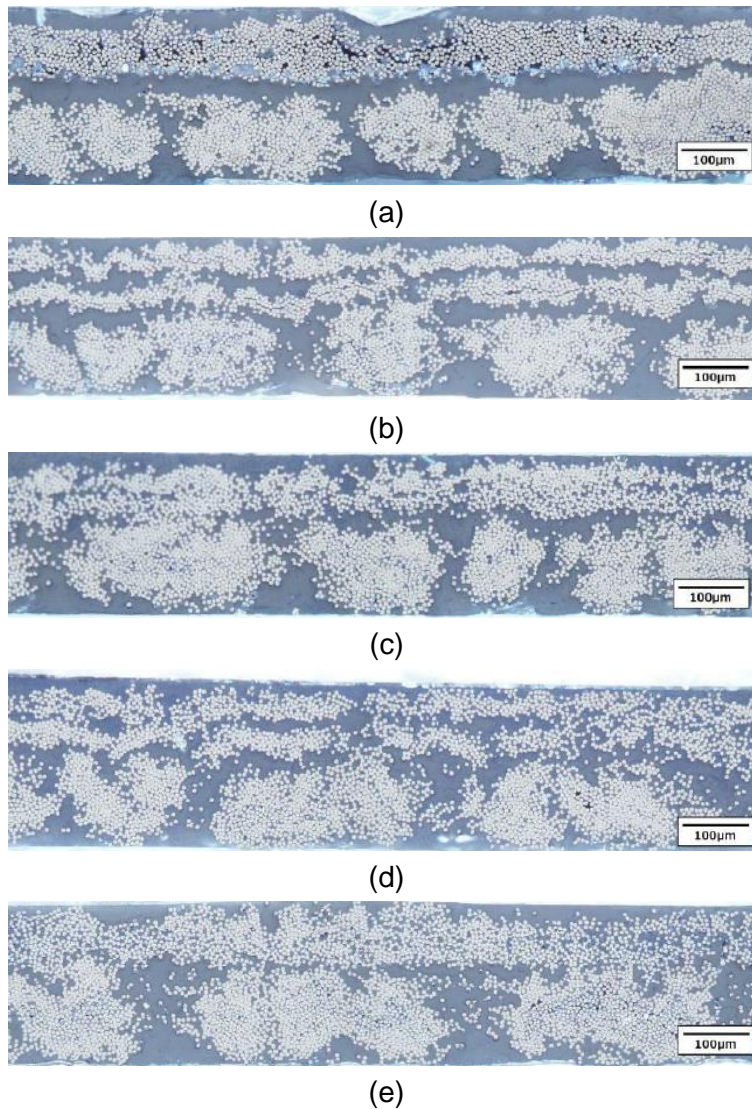


Figure 5 – Cross-sectional images of test specimens fabricated at a laser power of (a) 160W, (b) 170W, (c) 180W, (d) 190W and (e) 200W.

### 4.3 Effect of Laser Irradiation Angle

As increasing the laser irradiation angle, the region shaded by the compaction roller increases as shown in Fig. 6. Temperature of the specimen decreases significantly in the shaded region especially when the temperature before the shade is high. Hence, effect of the irradiation angle was examined. Figures 7 (a), 7(b) and 7(c) show cross-sectional images of the specimens fabricated at a laser irradiation angle of 25°, 30° and 35°, respectively. The specimen can be well fabricated at the laser irradiation angle of 25° and no void exists in the specimen. We can see the crack-like voids in the first and second CF-PA6 layers and also large voids in the base prepreg layer in the specimens fabricated at the laser irradiation angles of 30° and 35°. The crack-like voids come from the temperature decrease due to the shade and the voids in the base prepreg layer come from the overheat. The overheat is caused by high power per unit area due to the high irradiation angles.

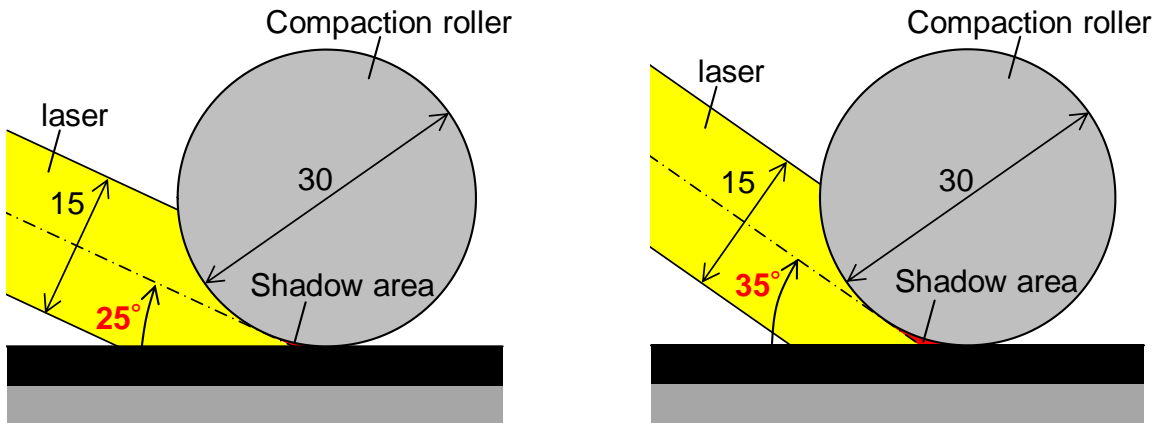


Figure 6 – Shaded area dependent on the irradiation angle.

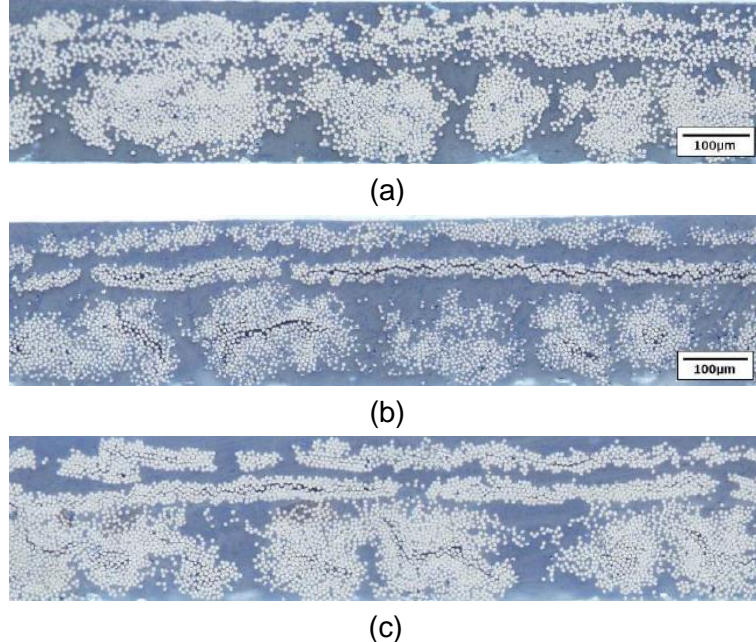


Figure 7 – Cross-sectional images of the specimens fabricated at an irradiation angle of (a) 25°, (b) 30° and (c) 35°.

### 4.4 Effect of Sizing Agent Removal Treatment

To improve interface strength between fibers and resin and to make easy to handle a fiber bundle, the fibers are usually coated with a sizing agent. This sizing agent is not necessarily adequate for the present resin and the present fabrication method. In this experiment the CF tow was heated at 400°C for 10 minutes to remove the sizing agent partially because the fully sizing removed fibers are



difficult to handle. Figures 8(a) and 8(b) show the cross-sectional images of the specimens fabricated at the laser power of 160W and 180W after the sizing agent removal treatment, respectively. It is seen that the fibers can be impregnated with the resin well even for the laser power of 160 W by removing the sizing agent.

There were almost no differences between with and without the sizing agent removal treatment at the laser power of 180W. The sizing agent can be considered to be removed when the laser heats the CF tows at 180W.

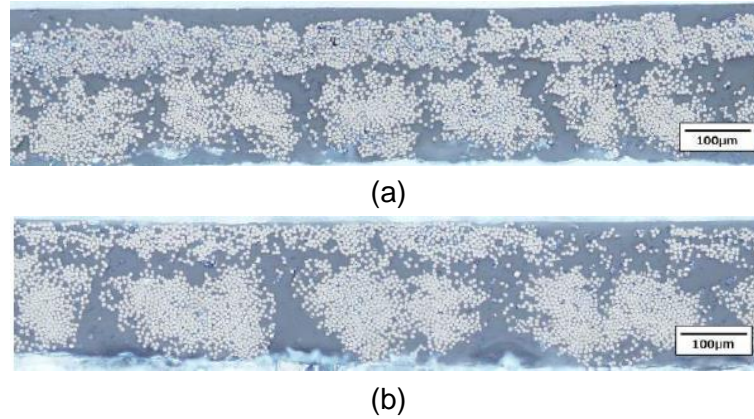


Figure 8 – Cross-sectional image of the specimens fabricated after the sizing agent removal treatment at a laser power of (a) 160W and (b) 180W.

#### 4.5 Mechanical Properties of AFP-D Specimens

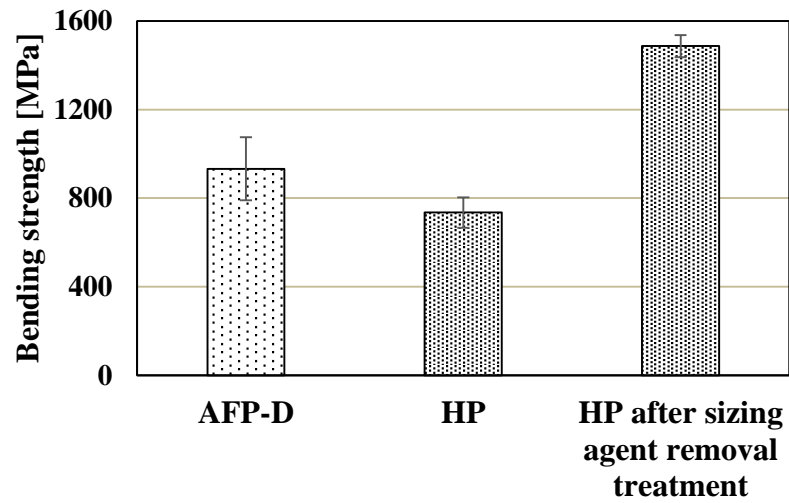
To evaluate mechanical properties of the specimens fabricated by the proposed AFP-D methods, bending strength and modulus of the specimens are compared to the specimens prepared by a conventional heat press (HP) method. The AFP-D specimens for the bending test were prepared by laminating twenty CF-PA layers on the base prepreg layer for an optimal fabrication condition, which are the same as the control values followed by the asterisks in Table 2. The HP specimens were prepared by charging twenty pairs of the CF tows and the PA6 films on the base prepreg sheet into a mold and heating and pressing them at 230°C and 2.5MPa for 15 minutes. Two types of CF tows were examined for the HP specimens, which were subjected to the sizing agent removal treatment described in Section 4.4 or not.

Three-point-bending test was performed following JIS K7074 [9]. Figures 9(a) and 9(b) show the bending strength and modulus, respectively, for the AFP-D specimens and the HP specimens without and with the sizing agent removal treatment. The properties for the AFP-D specimens have larger error bars since some of the process parameters of the prototype AFP-D were controlled manually although “automated” is included in the name. The bending strength for the AFP-D specimens, the HP specimens and the HP specimens with the sizing agent removal treatment are 933MPa, 735MPa and 1487MPa, respectively, and their bending modulus are 84GPa, 90GPa and 83GPa, respectively. The averaged values of the bending modulus are within the error bars for the three types of specimens while some amount of differences appears in the bending strength. It is noted that the strength of the AFP-D specimens is higher than the HP specimens when the sizing agent removal treatment is not subjected.

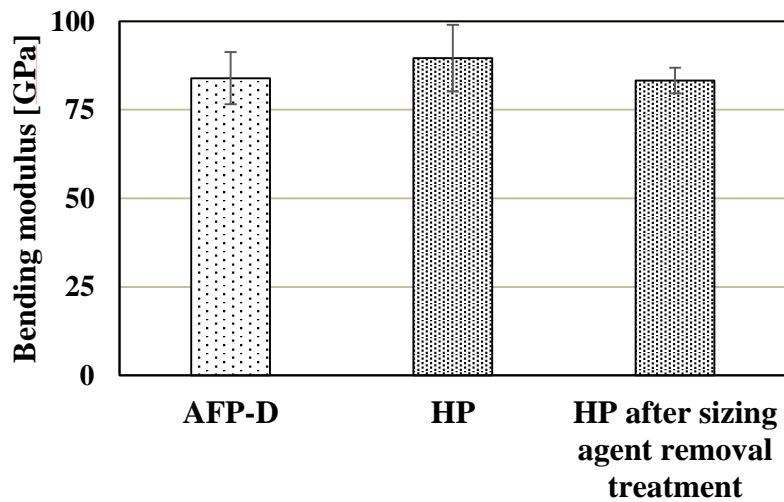
To discuss the reason of the difference in the strength, cross sectional images for the three types of specimens were observed. Figures 10(a), 10(b) and 10(c) are the cross-sectional images for the AFP-D specimens and the HP specimens without and with the sizing agent removal treatment. Almost no void exists and the CF tows are well impregnated with the resin for all the types of specimens. Crack-like defects appear in the top layers for the AFP-D specimen. Temperature in the top layers of the specimen varies, since the distance and the angle between the laser and the top layer changes increasing the number of the layers during the placements. Degree of homogenization for the AFP-D specimen and the HP specimen with the sizing agent removal treatment are better than the HP specimen without the sizing agent removal treatment. Accordingly, the interlaminar



strength is also higher for the two types of specimens than the HP specimens without the sizing agent removal treatment.

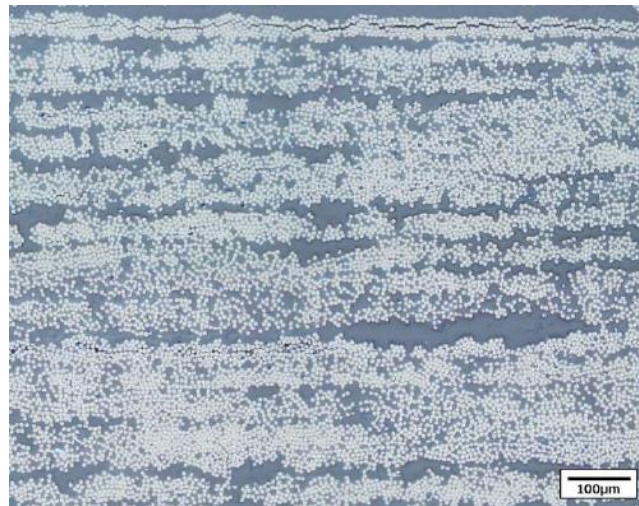


(a)

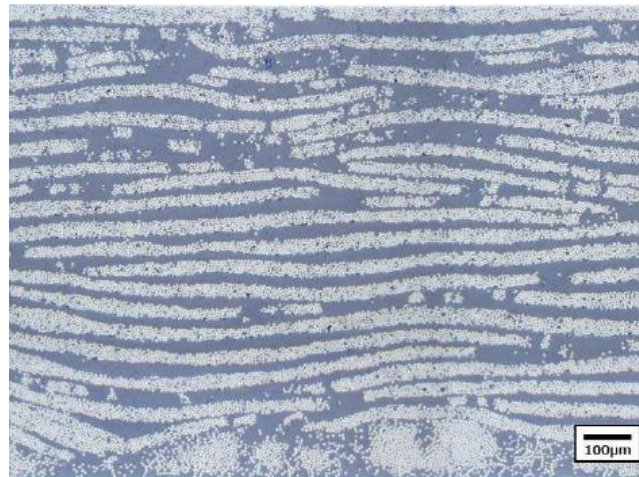


(b)

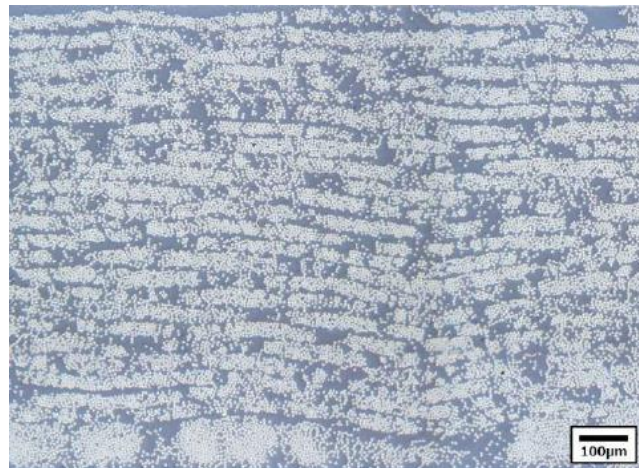
Figure 9 – Mechanical properties of the specimens fabricated by the AFP-D, the heat press without and with the sizing agent removal treatment. (a) Bending strength and (b) bending modulus.



(a)



(b)



(c)

Figure 10 – Cross-sectional images of the specimens fabricated by (a) AFP-D, (b) heat press without and (c) heat press with the sizing removal treatment.

## 5. Conclusions

The AFP-D method was proposed, where a thermoplastic film and a spread CF tow are independently supplied into the AFP-D system, and the CF tow is impregnated with the thermoplastic resin and laminated in-situ directly. The results of the fundamental analysis of the AFP-D manufacturing process and its demonstration are summarized below.

- (1) Fundamental analysis shows that the prototype AFP-D system increases the temperature of the CF tows up to 600K, which is higher than the melting temperature of the PA6, and the CF spread tow with a thickness of 35 $\mu$ m and a volume fraction of 0.6 is completely impregnated with the PA6 within 0.06s.
- (2) A compaction roller should be wrapped with a rubber sheet.
- (3) There exists an optimal set of process parameters such as laser power, laser irradiation angle and so on.
- (4) A sizing coating is not needed to be removed in the AFP-D method.
- (5) Bending strength of the AFP-D specimens is higher than the HP specimens without the sizing agent removal treatment but lower than the HP specimens with the sizing agent removal treatment.

This paper is a kind of progress report on our on-going work. A continued effort will be made in order to improve quality of CFRTP products.

## 6. Contact Author Email Address

mailto: [ikeda@isc.chubu.ac.jp](mailto:ikeda@isc.chubu.ac.jp)

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