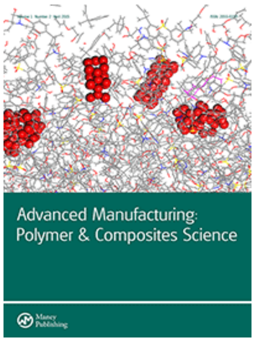

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Takushi Miyake & Satoshi Imaeda

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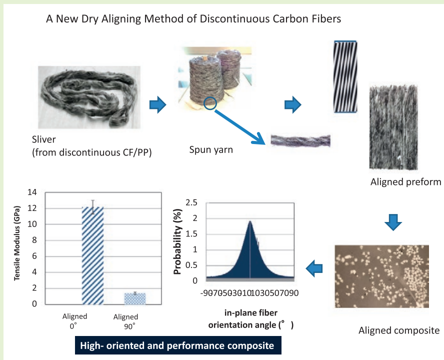
Original Research Paper

A dry aligning method of discontinuous carbon fibers and improvement of mechanical properties of discontinuous fiber composites

Takushi Miyake[1] and Satoshi Imaeda[2]

[1]Composite Materials Center, Gifu University, 1-1 Yangido, Gifu 501-1193, Japan
[2]Department of Mechanical Engineering, Gifu University, 1-1 Yangido, Gifu 501-1193, Japan

Abstract A novel method for dry alignment of discontinuous carbon fibers, such as recycled carbon fibers was developed by the modification of the yarn manufacturing process. In order to achieve high production rate and application for long fibers, fluffy synthetic fibers were used as the suspending and carrying media for discontinuous carbon fibers instead of a fluid medium generally used. Using the mechanical interaction in the dry alignment method, a mixture of 200 mm long discontinuous carbon fibers and fluffy polypropylene fibers was fabricated into an aligned and homogeneously comingled sliver by drafting. Then, the sliver was spun into yarn and an aligned yarn preform was obtained. Composites, with approximately 70% of the fibers aligned within an angle of ±14° with respect to the drafting direction, were successfully produced from the preform of the aligned spun yarn. The tensile modulus of the composite specimen along the aligned direction was about 10 times larger than that along the transverse direction, and about 3.5 times larger than that for the specimens obtained from the same yarn with random orientation.



Keywords Alignment of discontinuous fiber, Spun yarn, Dry method, Recycled carbon fiber

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Introduction

The increased use of carbon fiber reinforced composite materials has rendered the need for recycling the generated waste a crucial requirement, both economically and environmentally. In order to utilize recycled carbon fibers effectively, actualization of high performance composites reinforced by recycled carbon fibers is inevitable. Reuse of recycled discontinuous fiber in high performance structural composites requires a large fiber length, high alignment ratio, and high volume fraction of the fiber. In particular, a high alignment ratio is crucial for the improvement of mechanical properties. When discontinuous carbon fibers can be fabricated in a continuous aligned form with high production rate, then unidirectional carbon fiber/thermoplastic preform such as unidirectional tape will be easily made from them. Manufacturing by means of tape placement machines can be available for discontinuous fibers in the same manner as continuous fibers. Tailored design or local reinforcement using tape-shape discontinuous carbon composites will achieve reasonable improvement of the performance of composite parts with minimal increase of weight and expense.

In previous studies, different alignment processes for discontinuous fibers are reported.[1–4] Most of the proposed alignment methods use either the hydrodynamic or pneumatic effect to move and rotate discontinuous fibers. Wet processes, e.g. a modified papermaking technique and converging jet flow technique, have been developed and favorable results have been obtained. High modulus aligned composites have been successfully fabricated using discontinuous fibers shorter than 5 mm.[2–4] From the view point of mechanical properties, especially impact strength, longer fibers are desired. In case of long fiber, the spacing between the fibers is not enough for fibers to rotate their direction and fibers are easy to interact. Momentum arose by fluid media is not enough

*Corresponding author. email miyake@gifu-u.ac.jp

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Table 1 Specifications of the carbon fiber

	Unit	T700SC-12k-60E (Toray Co, Ltd.)
Cut length (cut from tows)	(mm)	200
Tensile strength	(MPa)	4900
Tensile modulus	(GPa)	230
Sizing	(wt%)	0.3

to rotate long fibers Therefore, the use of alignment methods based on hydrodynamic effects is limited to short fibers.[4] Additionally, use of a dilute suspension is inevitable to allow discontinuous fibers to rotate freely in a flow medium. This necessitates the use of a large amount of liquid medium or a high viscosity medium, and results in low production rate. Hence, a dry process with mechanical interaction is expected to help overcome these issues.

Therefore, the aim of this study is to develop a technique applicable for long discontinuous fibers via a dry procedure, with high productivity. To realize this aim, fluffy synthetic fibers were employed as the suspending and carrying medium for the discontinuous fibers, instead of a liquid medium. This is because synthetic fluffy fibers tend to crimp readily and hence can be easily drawn along a given direction by conventional textile techniques. Carbon fibers can be aligned together with the synthetic fibers if there is mechanical interaction between them.

This paper discusses the developed dry fiber alignment method that enables the handling of long discontinuous fibers by making use of synthetic fibers of thermoplastic as the carrier medium. In this study, mixture of discontinuous carbon fiber of 200 mm long and polypropylene fluffy fiber were processed following a traditional textile procedure of yarn fabrication, and comingled sliver was produced. Then yarn was spun from the sliver and finally aligned discontinuous fiber composite sheets were made from preform of stretching the spun yarn in one direction. The tensile stiffness and strength of the aligned composite specimens were tested and compared with those obtained from specimens of randomly oriented yarn composite. Fiber orientation in the composite specimen was characterized using X-ray CT images and was correlated with the tensile properties.

Experimental

Materials

PAN (polyacrylonitrile)-type carbon fibers of high strength (T700SC-12K Toray Co., Ltd.) were used in the study. The specifications of the carbon fibers are listed in Table 1. The thermoplastic matrix was fluffy polypropylene fiber (Daiwabo Polytec Co., Ltd.). The carbon fiber sizing agent is compatible with epoxy resin and 0.3 wt% in amount. About 200 mm long pieces of carbon fiber were cut from continuous tow to simulate the reclaimed discontinuous fibers. The pieces were opened in advance by a modified carding machine.[5] Polypropylene fiber was used as received.

Alignment process and apparatus

The mechanism of discontinuous carbon fiber alignment using fluffy synthetic fiber is shown in Fig. 1. The carbon fibers changed their orientation along the drafting direction by following the synthetic fibers used as the suspension medium, because the synthetic fibers can be easily stretched and aligned by drawing. Together with reduction in the thickness, uniform comingling of the carbon fibers was observed during the drawing process.

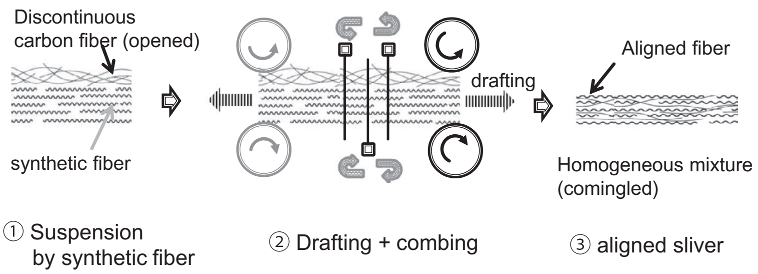

Figure 1 Schematic of the developed alignment method used for the fiber orientation

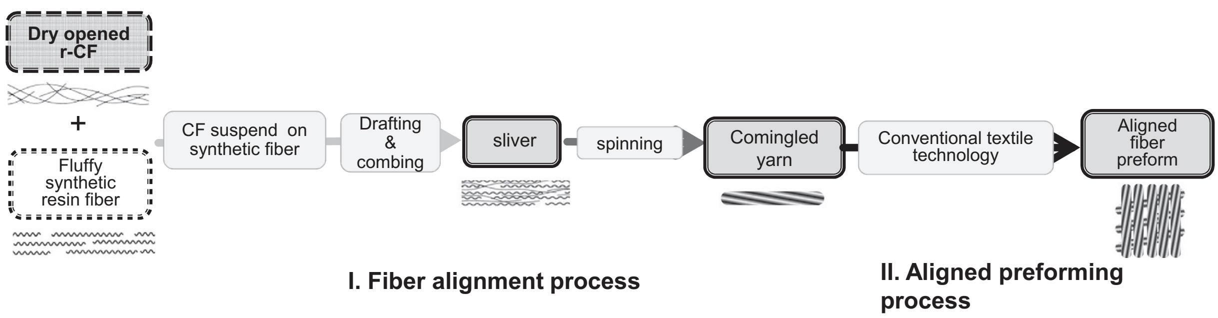

Figure 2 Flow chart of discontinuous fiber alignment process

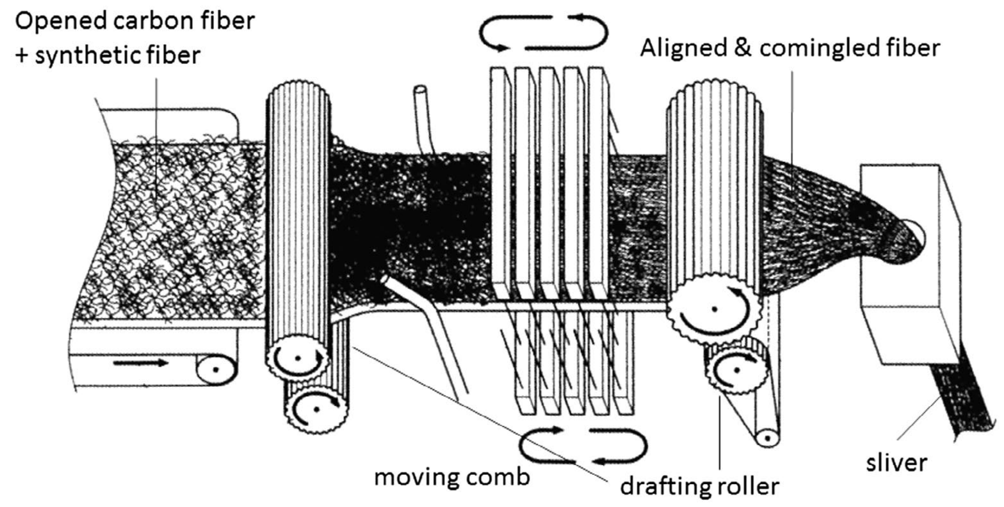

Figure 3 Schematic of the equipment used for making aligned sliver

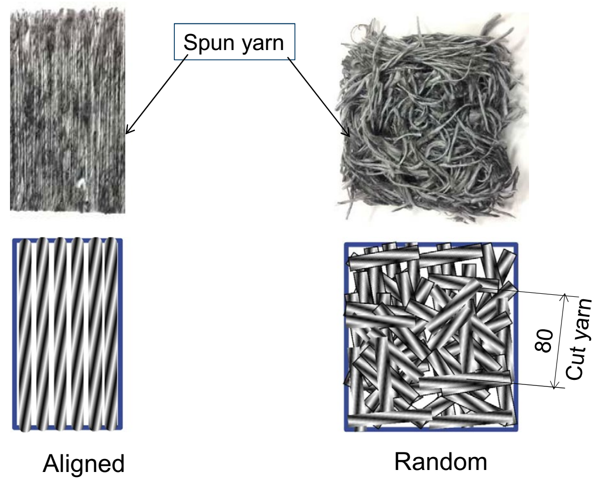

Figure 4 Illustration of unidirectionally and randomly oriented composite samples

The overall procedure includes the following steps, as illustrated in Fig. 2,[6]

(1) Opening of discontinuous carbon fibers.
(2) Suspension of the opened carbon fibers on fluffy synthetic fibers.
(3) Drawing the mixture of fibers together in one direction, and execution of the drawing process multiple times if necessary.
(4) Reducing the comingled mat into sliver.

Then, the sliver is spun into the yarn form, and discontinuous carbon fibers are more aligned in the axial direction by stretching during the process.

(5) Spinning the comingled sliver into yarn.

Once discontinuous fiber is made into yarn, the preform of the oriented discontinuous fiber, e.g. unidirection tape, is fabricated using traditional textile methods.

(6) Placing the yarn in one direction to form the aligned fiber preform.

Figure 3 shows the schematic illustration of the apparatus used for discontinuous fiber alignment. The apparatus consists of two pairs of rollers and moving combs, similar to a textile machine called "Gill", which was used for shaggy and rigid wool alignment. The two pairs of rollers show some difference in their rotation speeds, and they draw the mixture of discontinuous carbon fiber and fluffy thermoplastic fiber in the rotational direction. Moving combs push discontinuous carbon fibers into thermoplastic fibers and mix the comingled fibers uniformly. To ensure that the developed method is applicable to long fibers, the tangled long carbon fibers were opened by a modified special garnet machine combined with pneumatic action, instead of a usual carding machine,[5] before suspension on the synthetic fibers.

Fabrication of aligned discontinuous fiber composite specimens

Composite samples with different fiber orientations were prepared by hot-pressing the preform of unidirectionally and randomly placed spun yarn, as shown in Fig. 4. The preform of the unidirectional composites was fabricated by stretching the yarn in one direction and hot-pressing in a semi-closed 80 mm wide and 100 mm long mold. Random preforms were fabricated by dispersing 80 mm long cut yarn and hot-pressing in a closed mold of 115 mm × 115 mm. The hot press-molding was performed as the following to prevent the aligned fibers from disrupting their orientation. The material was placed in the mold heated at 200 °C and then pressed at the pressure of 2 MPa. After keeping for 10 min, the material was cooled down in the mold under the pressure. The volume fraction of the carbon fiber in the composites was evaluated by a combustion method, in which weight of residue after burning was estimated as weight of carbon fiber.

Determination of fiber orientation

Fiber orientations were determined using X-ray CT images, which revealed information related to the thickness direction of the composites, and not just information pertaining to the surface. Composite plates with sizes of 10 mm × 10 mm were cut for the X-ray CT imaging. Images of the plates were obtained using a micro X-ray CT (SkyScan 1172 Bulker) with a spatial resolution of ≈0.5 μm. Probability density functions

Figure 5 Photographs of fabricated (a) sliver and (b) spun yarn

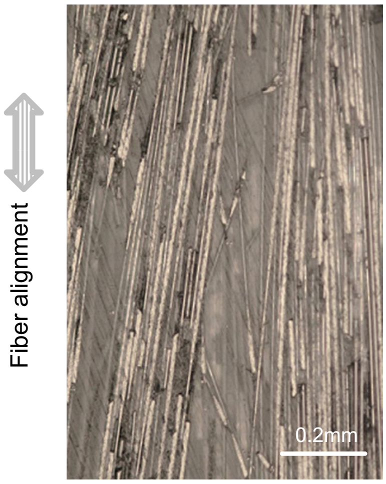

Figure 6 Microscope image of the surface of the aligned composite sample

for in-plane fiber orientation were obtained using composite material fiber analysis software (VGStudio MAX Volume Graphics).

Mechanical testing

Specimens used for 0°-direction tensile tests were cut from the hot-pressed plates along the fiber direction. 90° tensile tests along the transverse direction were performed using specimens cut along their longitudinal direction at 90° against the 0°-direction test specimen. Five specimens of each direction were tested in tensile. The tensile test specimens were 10 mm wide, 100 mm long, and 1 mm thick. End tabs were adhered on all the specimens. Tensile tests were performed using an electro-mechanical testing machine (3376 Instron) at a crosshead speed of 1 mm/min. The strain in the samples was measured by a contact-type extensometer with a gage length of 50 mm (2639-111 Instron). Load was measured with a 30 kN load cell (Instron) and calibrated by another load cell of 1 kN capacity.

Results and discussion

Structure of aligned composites

Figure 5 shows photographs of the fabricated sliver and spun yarn. The discontinuous carbon fibers and synthetic fibers were uniformly mingled (Fig. 5*a*). Fibers were separated individually and mingled with each other in the spun yarn (Fig. 5*b*).

Optical microscopy images of the surface and the cross section of the fabricated aligned composite are illustrated in Figs. 6 and 7. The image of the composite surface revealed that fibers maintained a length of a few millimeters. The mean fiber length in the composite was evaluated as 60 mm by a combustion method. The cross sectional image shown in Fig. 7 reveals that the carbon fibers were oriented perpendicular to the cross section and dispersed individually. Because of the low fraction of the carbon fibers, resin-rich portions were generated as well. The volume fractions of the carbon fiber were evaluated to be 13.3% in the aligned unidirectional composites. The volume fraction is comparable with the measured value from the cross sectional image shown in Fig. 7. The volume fraction in the randomly oriented composites was 12.8%, which was almost equal to that of the aligned composite samples.

Fiber orientation

Figure 8*a* shows an X-ray image of the plane along tensile direction of the unidirectional composite sample. Owing to the high resolution of the X-ray scanner, individual fibers in the composite were clearly observed. In Fig. 8*b*, the software analyzed result is shown. The evaluated direction angles are reasonably consistent with the observed fiber direction in the X-ray image.

Probability distribution of fiber orientation angle for each 1° was shown in Fig. 9. When the probability distribution was fitted with a Gaussian function, the standard deviation was calculated as 13.7°. In the case of the unidirectional composite specimens, approximately 70% of the fibers were in the range ±14°. The variation in fiber orientation was supposed to be attributed to the helicoidal change in the orientation

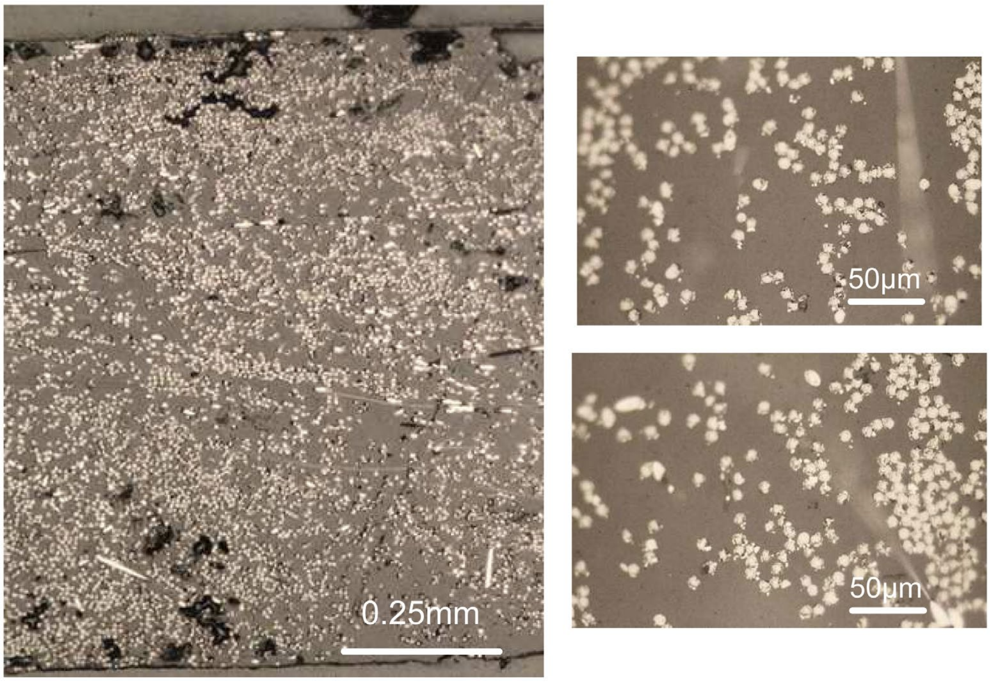

Figure 7 **Microscope images of the cross-section of aligned composite ample**

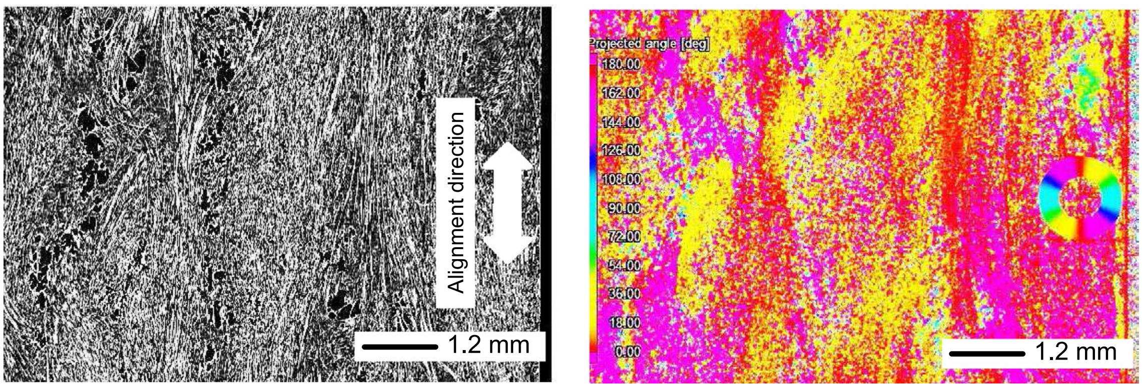

Figure 8 **In-plane images of aligned composite sample: (a) X-ray image and (b) result of fiber orientation analysis**

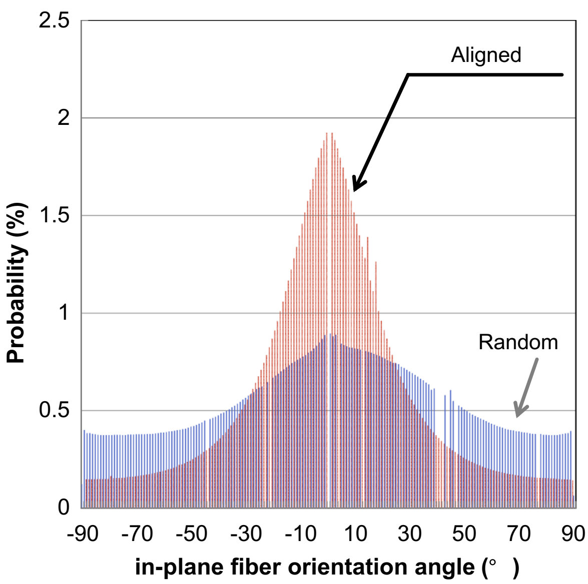

Figure 9 **Distributions of fiber orientation of aligned and randomly oriented composite specimens**

of the discontinuous fibers during the spinning process. The fiber angle distribution evaluated by the same procedure for random composite specimens is also shown in Fig.9. In contrast to the case of the aligned specimens, the distribution of the randomly oriented fibers is more uniform, as expected.

Mechanical properties

Figure 10 shows the tensile modulus and tensile strength results of 0°-direction tests (along the aligned fiber) and 90°-direction tests (perpendicular to the aligned fiber). The interface between the carbon fibers and the matrix resin is weak due to the poor adhesion of polypropylene to the carbon fiber surface. The stress–strain curve shown in Fig. 11 and the fracture surface of the aligned composite in Fig. 12 indicate that the fiber pull-out played a dominant role in the fracture strength of the composite. In addition, taking the low volume fraction of the carbon fibers into account, composite strengths are deemed unsuitable for further discussion; therefore, the discussion was focused on the composite moduli.

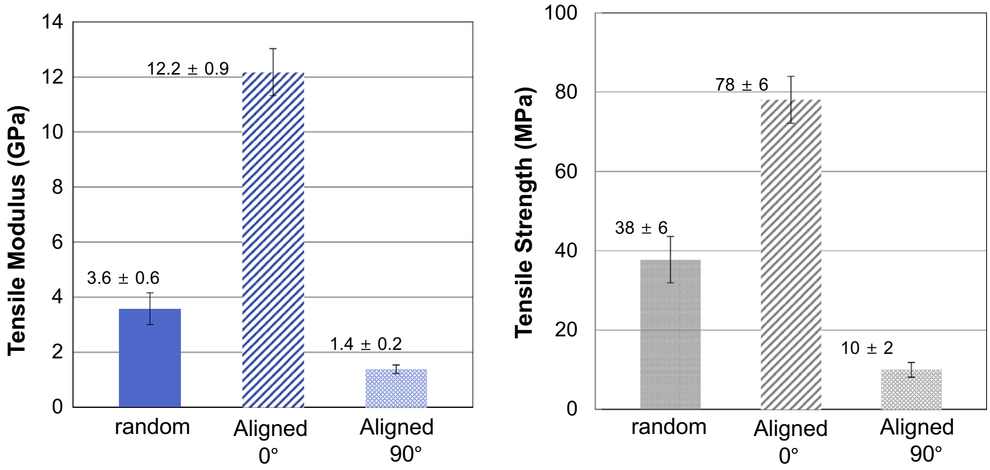

Figure 10. **(a) Tensile modulus and (b) tensile strength for the 0°- and 90°-direction of aligned and randomly oriented composites**

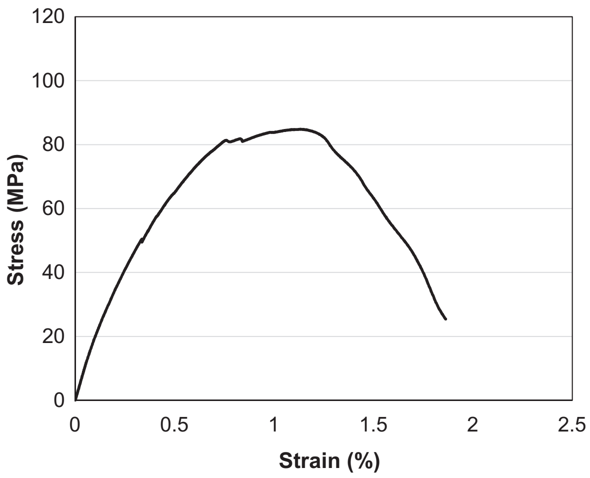

Figure 11 **Stress–strain curve of aligned discontinuous carbon/Polypropylene composite**

Fairly large anisotropy of the tensile moduli was found in tensile test results, as shown in Fig.10. The ratio of the moduli for the 90°–0° directions was about 1/10. This value was a little higher than that of the completely aligned composites reported in the other study, 0.06–0.08.[7] The modulus of the aligned composites is approximately 3.5 times higher compared to the randomly oriented composite fabricated from the same yarn. These results are attributed to the relatively wide distribution of fiber orientation, where 70% of the fibers are within ±14°. Therefore, the tensile modulus is expected to be improved when the distribution of the fiber alignment angle is narrowed down.

Conclusions

In this paper, a new dry alignment method for recycled discontinuous carbon fibers is introduced. In order to achieve high production rates and enable the use of this method for long fibers, fluffy thermoplastic fibers were used as the suspending and carrying medium for the discontinuous carbon fibers, instead of fluid media. A mixture of 200 mm long carbon fibers and polypropylene fluffy fibers was processed to form a comingled spun yarn by means of a modified textile machine. Aligned composites were fabricated from the yarn by placing the yarn in one direction and hot-pressing at 200 °C.

The tensile modulus of the aligned composites between the aligned direction and the transverse direction was in the ratio 10:1. The tensile modulus of the aligned composites was approximately 3.5 times higher as compared to that of the randomly oriented composite fabricated from the same spun yarn. The fiber orientation distribution determined by X-ray CT images showed a near-Gaussian distribution. Almost 70% of the fibers were distributed in the range of ±14°.

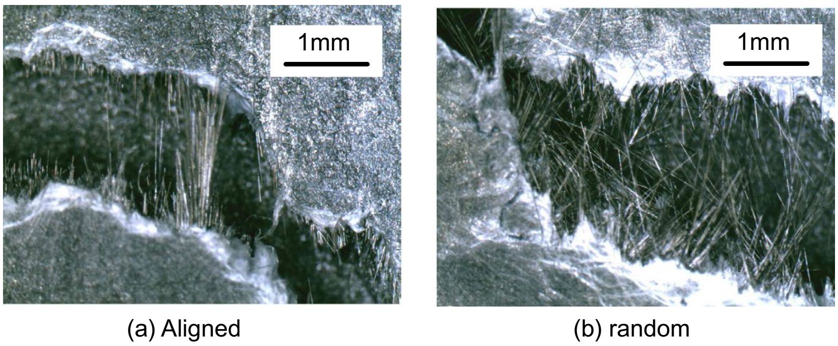

Figure 12 **Tensile fracture surface of (a) aligned composite specimen and (b) randomly oriented composite specimen**

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Disclosure statement

No potential conflict of interest was reported by the authors.

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