A NEW METHOD FOR CONTINUOUS PRODUCTION OF RAMIE YARN REINFORCED COMPOSITES

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Abstract

We focus on long fiber-reinforced composite strands and tapes using continuous ramie yarns, in which polypropylene including MA-PP (Maleic anhydride grafted polypropylene) was used as a matrix material. The composite strands and tapes were fabricated by a new method called Multi-pin-assisted Resin Impregnation (M-PaRI) process, of which the equipment was newly attached after conventional extrusion process. Tensile strength and Young's modulus of short ramie/PP reinforced composites and ramie/PP composite tapes through M-PARI process were investigated.

1 Introduction

Recently, Carbon fiber reinforced plastics (CFRP) and glass fiber reinforced plastics (GFRP) have been developed in engineering applications because of their excellent mechanical properties and huge demands in industry. However, CFRP and GFRP have also disadvantages such as poor recycling properties, non-biodegradability and potential health problems. Global environmental problems and energy shortage have brought the focus of making alternative materials from biomass resources. Natural fibers offer many superior benefits to glass fibers. They are abundant, biodegradable, renewable, and exhibit similar specific superior properties to glass fibers. Ramie fiber is one of the strongest natural fibers with high cellulose and low lignin contents. Mechanical properties of short fiber-reinforced composites depend on micro-structural parameters such as fiber length distribution, the alignment and packing arrangement of fibers. On the other hand, high mechanical properties are exhibited in long fiber-reinforced composites consisting of unidirectionally fiber-oriented layers or laminate stacked up in a tailored arrangement.

The main problem on short fibre composite materials is that the fibres are often broken during the fabrication processes such as extrusion and injection moulding, resulting in shorter fibre length than the critical one. In order to maintain appropriate mechanical properties of the composites, as is well-known, the fibre length has to be longer than the critical value. One of popular methods is to use pultrusion technique developed by ICI for the manufacture of Verton long fibre moulding materials. Thomason et al [1] also introduced a coating process using jute yarns to produce composite strands. However, poor performance of tensile properties on injection-moulded composite materials was reported, which is caused by low

Density (g/cm^3)	Microfibril angle (°)	Moisture content (wt%)	Chemical composition (wt%)				
			Cellulose	Lignin	Hemicellulose	Pectin	Wax
1.50	7.5	8.0	68.6-76.2	0.6-0.7	13.1-16.7	1.9	0.3

Table 1. Physical and chemical properties of ramie fibres [4]

level of impregnation between fibres. To solve this drawback, Bledzki et al [2] tried two-step extrusion process. Recently, a new technology for production of jute twisted yarn-reinforced composites has been reported by Tanaka and Hirano [3], in which both complete impregnation and long fibres length were achieved. However, the disadvantage of this technology is that the fabrication procedures are complicated, that is to say, jute yarns must be untwisted separately to facilitate resin impregnation process.

In this study, thus, a continuous ramie single yarn reinforced polypropylene (PP) composite strand was developed using a new and relatively simplified multi-pin-assisted resin impregnation (M-PaRI) process. The ramie/PP strands were pelletized and injection-moulded, and their tensile properties were measured. In addition, ramie/PP composite tapes were fabricated by using the M-PaRI process with the aim of application to a plain woven fabric or composite like a glass roving cloth.

2 Experimental

2.1 Materials

Continuous ramie single yarns, having a fineness of 95 tex, Type No. 16 (TOSCO, Co., Ltd., Japan) and polypropylene (Prime Polymer Co., Ltd., Japan) were used as a reinforcement and matrix material, respectively. Physical and chemical properties of ramie fibres are listed in Table 1. Maleric anhydride modified poly-propylene (MA-PP, Kayaku Akzo Co., Ltd., Japan) was also used as a coupling agnet to promote better fibre-matrix interaction in this study.

2.2 Fabrication procedures

Figure 1 shows schematic view of the present fabrication system. Continuous ramie single yarn/PP composites were produced through a new combined technique proposed in this study, which consists of resin coating process (Process A) and Multi-pin-assisted resin impregnation (M-PaRI) process (Process B) as shown in Figure 1. The continuous ramie single yarns were first delivered via preheating process into a cross-head die attached to a Φ 15mm single screw

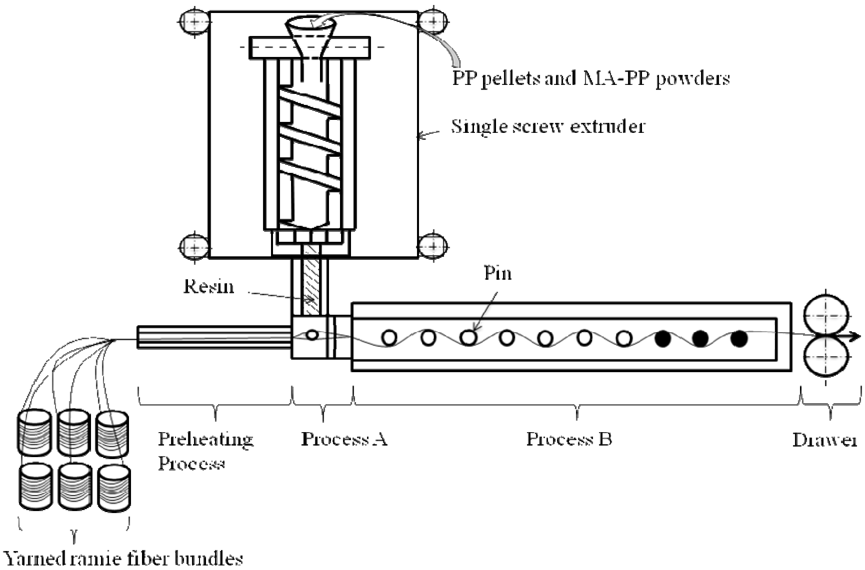


Figure 1. Schematic view of the present fabrication system (Process A: Resin coating process, Process B: Multi-pin-assisted resin impregnation (M-PaRI) process).

extruder (Spinning machine, Musashino Kikai Co., Ltd., Japan), into which PP pellets and MA-PP powders were fed at the same time. The mixed resin was coated on the ramie yarns in the die at Process A. After that, it was impregnated into inter-fibres through multi-pin system, as shown in Process B of Figure 1. The number and diameter of pins used here were 22 and 5 mm, respectively. Temperatures of the single screw extruder were all set at 190^{o}C with a screw speed of 7.0 rpm. A motor was set to draw the composite strand with a screw speed of 45.0 rpm.

The continuous ramie/PP composite strands containing six ramie yarns were chopped into pellets of 2mm length. Set temperature(s) was 190^{o}C at Process A, and were in the 160-225 ^{o}C range for Process B. The pellets were moulded into a tensile specimen on an injection moulding machine (BeVel 20, Shinko Sellbic Co., Ltd., Japan). The temperatures were set at 180-185 ^{o}C for injection moulding. The dimension of the specimen was 2 mm × 3 mm cross-section and 18 mm gauge length. Fibre content of specimens was 30 wt%.

Figure 2 shows schematic representation of fabricating ramie/PP composite tapes containing thirty-three ramie yarns. 7×1 mm^2 rectangular die with 2 mm length was additionally attached to control the shape of strands at the end of M-PaRI process. Composite tapes were continuously pulled as shown in Process C of Figure 2, in which rollers were used to produce two different types of ramie/PP composite tapes. Type II was produced into a flatter tape because its roller can yield a higher tension than that of Type I. Fibre content of the specimen was 70 wt%.

2.3 Tensile test

Tensile tests were conducted using a universal testing machine (Ritorusensuta small desktop tester, JT Tosi Co., Ltd., Japan) for short ramie/PP specimens at a crosshead speed of 10 mm/min. Cross-sectional area was measured on three locations along the longitudinal direction using a micrometer and its average was taken.

In the case of ramie/PP tape specimens, tensile tests were carried out using a instron-type universal testing machine (Autograph IS5000, Shimadzu Corporation, Japan). The gauge length and tensile speed of tensile specimens were 50 mm and 1 mm/min, respectively. The cross-sectional area was measured using a digital microscope (KH-1300, Hirox Co., Ltd., Japan). Five specimens were evaluated for each condition. Ultimate stress, stiffness (defined as linear region between strain 0.05 and 0.25) and fracture strain were determined from the stress-strain curves for each specimen.

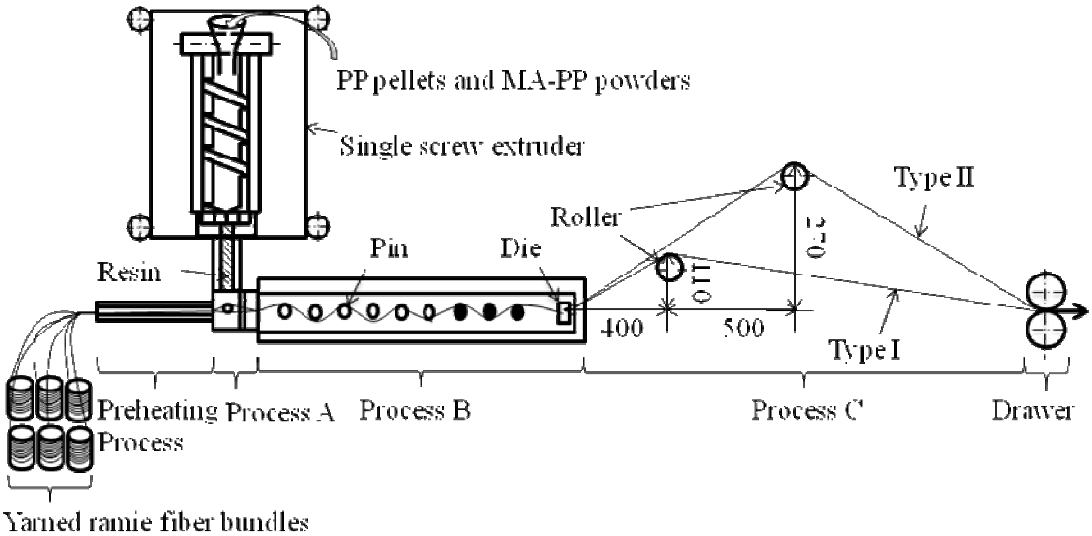
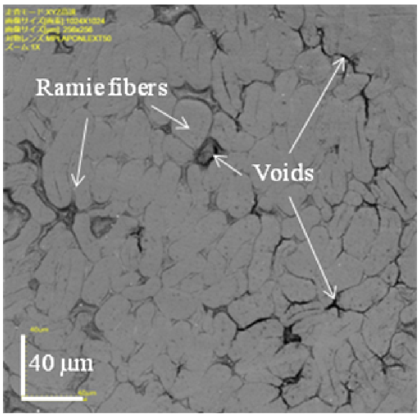
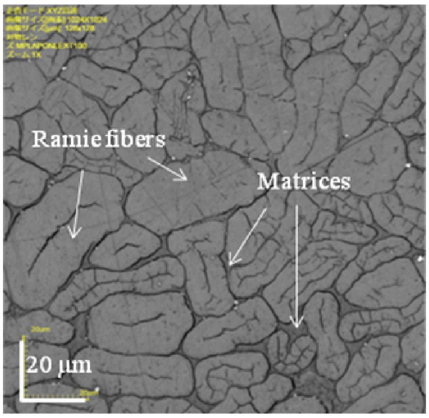


Figure 2. Schematic representation of fabricating ramie/PP compoiste tapes. (Process A: Resin coating process, Process B: Multi-pin-assisted resin impregnation (M-PaRI) process, Process C: Flattening process).





(a) (b)

Figure 3. The cross-sectional images of ramie/PP composite strands: (a) before multi-pin-assisted resin impregnation process and (b) final product.

3 Results and Discussion

3.1 Evaluation of impregnation in continuous strands by the proposed combined technique

The microscopic cross-section images of ramie/PP composite strands before and after M-PaRI process are shown in Figure 1. In this case, the set temperature of Process B was 195^{o}C. It can be observed from Figure 1(a) that not only resin is locally infiltrated, but relatively large amount of voids can be seen between fibres. Final product in Figure 1 (b) shows that no void occurs. It is verified that the attached multi-pin system completely assists in impregnating the resin into inter-fibres. The mechanism of this impregnation is estimated such that continuous contacting and rubbing with multi-pins makes the resin-coating yarns flat, and as a result the inter-fibre spaces are widened sufficiently to be well impregnated. On the other hand, occurrence of voids was also observed in the strands produced at Process B temperatures less than 195^{o}C.

3.2 Tensile properties

Figure 4 shows typical tensile stress-strain curves of short ramie/PP reinforced composite specimens containing 30 wt% fibres. The result shows that the tensile behavior depends on the set temperature at Process B. In other words, it can be seen that there is an optimal temperature giving higher strength and elastic modulus. Table 2 shows the result of tensile properties of short ramie/PP reinforced composites. Tensile strength and Young's modulus increase with increasing temperature in the range of 160-195^{o}C. In the composites specimens

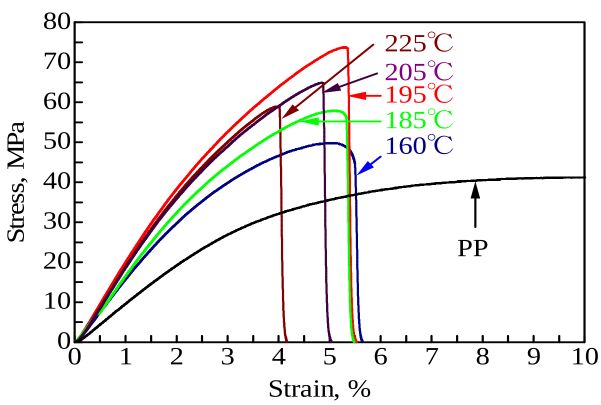


Figure 4. Typical stress-strain curves of short ramie/PP reinforeced composites at different temperatures during multi-pin-assisted resin impregnation (M-PaRI) process.

Material	T_{rip} °C	Young's modulus (GPa)	Tensile strength (MPa)	Fracture strain (%)
PP (0wt%)	-	1.04 (7.12)	42.4 (2.70)	>200
Composite	160	1.45 (11.3)	50.6 (5.29)	5.65 (8.28)
	185	1.64 (8.03)	55.1 (6.58)	5.32 (7.65)
	195	1.94 (7.16)	69.2 (4.57)	5.37 (3.97)
	205	1.72 (11.8)	67.7 (3.20)	5.26 (3.88)
	225	1.72 (8.62)	53.1 (8.60)	4.04 (9.25)

$T_{rip:}$ Temperature at resin impregnation process
The number in parentheses indicates coefficient of variation (%)

Table 2. Tensile properties of short ramie/PP composite specimens.

of 195°C, not only tensile strength is improved 1.63 times higher than PP specimens, but Young's modulus is increased up to 1.87 times. These results are consistent with the above-mention on the complete resin impregnation between fibres by means of additional Process B. On the other hand, there is a gradual decrease in tensile strength after the temperature exceeds 195°C. It is guessed that over-heating causes degradation of ramie fibres during the M-PaRI process. Figure 5 shows typical stress-strain curves of ramie/PP composite tapes. It can be seen that tensile strength and Young's modulus of Type II ramie/PP composite tape specimens were 181.1 MPa and 20.6 GPa, respectively, increased 1.35 % and 16.6 % in comparison with Type I specimens.

4 Conclusions

A new combine technique, resin coating and multi-pin-assisted resin impregnation (M-PaRI) processes, was introduced to produce continuous ramie single yarn reinforced polypropylene (PP) composite strands and tapes, using a single screw extruder. We found that the resin can completely be impregnated into inter-fibres of the yarns. Tensile test of injection-moluded composites was conducted by use of the strands produced at different temperatures of M-PaRI process. It was also confirmed that the maximum mechanical properties of composites can be

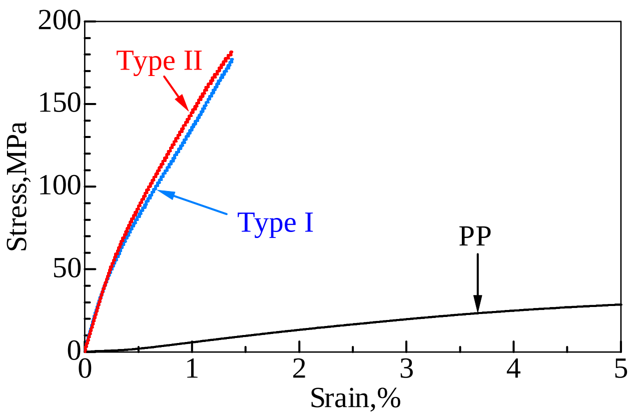


Figure 5. Typical stress-strain curves of ramie/PP composite tapes.

obtained at 195^{o}C, which is corresponding to the lowest of all temperatures used in completely impregnated composite strands. Tensile test of ramie/PP composite tapes was also carried out by using two different types of fabrication method. It can be noted that the tape specimens had better mechanical properties by applying a relatively higher tension after M-PaRI process. It is expected that ramie/PP composite tapes can be used as a semi-finished material for woven fabric composites.

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References

[1] J.L. Thomason, Dependence of interfacial strength on the anisotropic fiber properties of jute reinforced composites, *Polymer Composites*, **Vol. 31**, Issue 9, pp. 1525-1534, (2010).
[2] A.K. Bledzki, A. Jaszkiewicz and D. Scherzer, Mechanical properties on PLA composites with man-made cellulose and abaca fibres, *Composites*, *Part A* **Vol. 40**, Issue 4, pp. 404-412, (2009).
[3] T. Tanaka and Y. Hirano, *Kobe steel engineering reports*, **Vol. 51**, No. 2, pp. 62-66, (2001).
[4] K. Goda, M.S. Sreekala, A. Gomes, T. Kaji and J. Ohgi, *Composites, Part A*, **Vol. 37**, pp. 2213-2220, (2006).