Impregnation Quality Of Carbon Fiber Reinforced Waste Polypropylene With Variation Of Fiber Treatment And Matrix Recycling Cycles

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Carbon Fiber Reinforced Thermoplastics offer a promising alternative engineering material characterized by high stiffness, strength, lightweight, and superior formability compared to traditional metal counterparts. However, the high viscosity of the thermoplastic matrix poses a significant challenge in achieving optimal impregnation quality. The quality of impregnation plays a crucial role in determining the mechanical properties of the composite. Therefore, this research aims to investigate the impregnation quality in carbon fiber-reinforced recycled polypropylene composites. Recycled polypropylene with recycling cycles of 1, 3, and 5 times was utilized as the matrix material. Carbon fibers underwent three different treatment variations: immersion in liquid nitrogen, heating to 600°C followed by immersion in liquid nitrogen, and treatment with a silane coupling agent. The interfacial shear strength was assessed using a pull-out test, and the microstructure was examined through scanning electron microscopy (SEM). The research findings reveal that the highest interfacial shear strength value of 14.5 MPa was achieved in the recycled polypropylene material with a recycling cycle of 5 times, employing the liquid nitrogen immersion treatment for the carbon fibers. Conversely, the lowest interfacial shear strength value of 8.6 MPa was observed in the recycled polypropylene material with a recycling cycle of 1 time, involving the coupling agent treatment for the carbon fibers. These results were further substantiated by microstructure observations using SEM.

Keywords: Interfacial shear strength; Cryogenic treatment; Silane coupling agent; Recycled polypropylene

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1. Introduction

Composite materials, particularly those reinforced with carbon fibers, have gained significant attention in the field of engineering due to their exceptional mechanical properties and lightweight nature. Unterweger et al. [1] stated that high specifications of carbon fibers, particularly in terms of stiffness and strength, enable significant weight reduction. Carbon Fiber Reinforced Polymers (CFRPs) offer a viable alternative to traditional materials in various industries, including automotive, aerospace, and construction [2]. However, achieving optimal impregnation of carbon fibers within the polymer matrix is crucial to ensure superior mechanical performance and structural integrity of the composites [3].

In recent years, there has been a growing emphasis on recycling waste materials and reducing environmental impact. Waste polypropylene (PP) has garnered attention as a potential matrix material for carbon fiber composites, offering both cost-effectiveness and sustainability advantages. By utilizing recycled waste PP, the composite industry can contribute to reducing landfill waste and conserving resources.

The impregnation quality of carbon fiber-reinforced waste polypropylene composites depends on multiple factors, including fiber treatment and matrix recycling cycles. It is proposed that the interfacial adhesion between the fiber and the resin is improved in the recycled composite compared to conventional materials due to the effective impregnation of carbon fibers in the waste of carbon-fiber-reinforced thermoplastics [4]. Various fiber treatment methods have been investigated to enhance the wetting and bonding between the carbon fibers and the waste polypropylene matrix [5]. These treatment methods typically involve surface modifications of the carbon fibers to improve their compatibility with the polymer matrix, such as cryogenic treatment [6], thermal treatment [7], or the use of coupling agents [8, 9].

Furthermore, the recycling cycles of the waste polypropylene matrix also play a crucial role in impregnation quality [10]. The recycling process introduces changes in the polymer's molecular structure and rheological properties, potentially affecting the impregnation behavior and interfacial adhesion between the fibers and the matrix [11].

This paper aims to investigate the impregnation quality of carbon fiber-reinforced waste polypropylene composites, considering variations in fiber treatment and matrix recycling cycles. The fiber treatment methods under study include cryogenic treatment, thermal treatment, and coupling agent treatment, while the waste polypropylene matrix undergoes different recycling cycles. The impact of these variations on the impregnation quality will be evaluated through comprehensive experimental analysis.

Key aspects of the research will include assessing the Interfacial Shear Strength (IFSS) to quantify the interfacial bonding between the carbon fibers and the waste polypropylene matrix. Additionally, microstructural analysis, such as scanning electron microscopy (SEM), will provide insights into the fiber-matrix interaction and interfacial characteristics.

The findings of this study will contribute to a deeper understanding of the impregnation behavior in carbon fiber-reinforced waste polypropylene composites, shedding light on the influence of fiber treatment and matrix recycling cycles. These insights can guide the optimization of composite manufacturing processes, leading to the development of high-performance, environmentally friendly composites for various applications [10].

2. Materials and methods

The matrix material used in this research was polypropylene waste. The recycling cycles of the matrix were varied between 1 cycle, 3 cycles, and 5 cycles. The reinforcing fiber used was T700 SC 12K carbon fiber, produced by Toray Composite Materials America, Inc., Tacoma, WA, USA [12]. The carbon fibers were subjected to different treatments to investigate their effects on impregnation quality. Three treatment variations were applied: cryogenic treatment, thermal treatment, and silane coupling agent treatment. In the cryogenic treatment, the carbon fibers were immersed in liquid nitrogen at a temperature of -196°C for 10 minutes [13]. For the thermal treatment, the fibers were heated to a temperature of 600°C for 10 minutes and then immersed in liquid nitrogen [14, 15]. The silane coupling agent treatment involved immersing the carbon fibers in a solution of aminopropyltriethoxysilane (APTS) with a concentration of 1 wt.% for 20 minutes [8, 16, 17].

The fabrication of specimens was conducted using the MEIKI injection molding machine (manufactured by MEIKI & Company, Ltd Japan) with a 70-ton clamping capacity. Previously, the resin underwent a drying process with hot air circulation at a temperature of 80 ^{0}C for 4 hours to minimize the occurrence of voids resulting from residual moisture. The melt temperature value was set by the barrel heater bands and the temperature profile along the barrel was 200 ^{0}C. The injection molding setting parameters are shown in Table 1 [18, 19]. Configuring these parameters will also help minimize the disparity of voids in the injection-molded specimens. The melt temperature must be controlled within the recommended range for polypropylene. Too high or too low temperatures can lead to voids. High injection pressure at slow speed can reduce the chances of voids by ensuring complete filling of the mold.

Table 1. Injection molding setting parameters.

Parameters	Value	Unit
Screw rotation	24	rpm
Injection pressure	140	MPa
Holding pressure	25	MPa
Barrel temperature	200	°C
Mould temperature	40	°C
Holding time	12	sec
Cooling time	12	sec

The unidirectional carbon fiber was placed into the mold and the plastic melt was injected to cover the fiber and to produce composite specimens. The specimens were carefully prepared for IFSS testing, as shown in Fig. 1.

To assess the impregnation quality and interfacial shear strength, a pull-out test was performed as described in Fig. 2. The procedure entailed the extraction of a fiber bundle that was partially embedded within matrix sleeves. All samples were subjected to a 24-hour conditioning period at

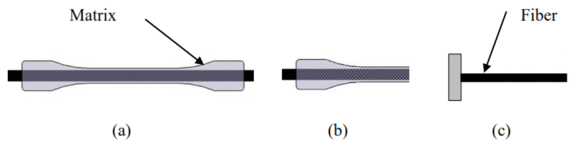


Fig. 1. Specimen preparation: (a) Molded; (b) Half cut; (c) Pull-Out test specimen.

a temperature of 23°C and a relative humidity of 50% before testing. The testing was conducted using a Zwick/Roell universal testing machine in a room maintained at 23°C and 50% relative humidity. The testing machine operated with a crosshead movement speed of 2 mm per minute, deliberately maintained at this low strain rate to prevent issues related to compliance. The controlled force pulled the carbon fibers out of the resin matrix, and the maximum force required was recorded as the interfacial shear strength (IFSS) [13, 20]. The IFSS (τ) can be determined as the maximum applied load (F) divided by the contact area using the Eq. (1) [21]. Where d and L_b are the width and the bond length of the contact area. To ensure the accuracy of the surface area measurements, the length and width of the bonding on each specimen's cross-section are measured using a vernier caliper with a precision of 0.02 mm. Multiple tests were performed on representative specimens (5 specimen for each run) to ensure consistency and reliability in the IFSS measurement. Averaging the results from several tests can provide a more accurate value.

$$\tau = \frac{F}{d \times L_b} \tag{1}$$

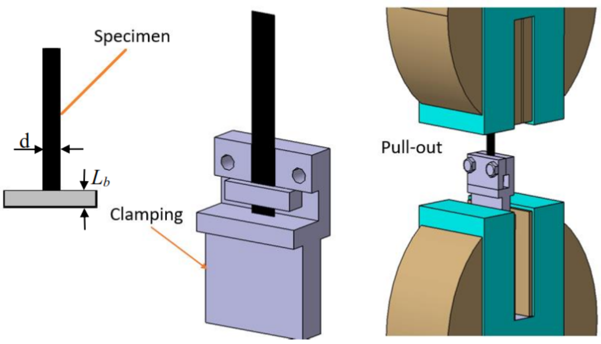


Fig. 2. Pull-out test.

Microstructural analysis of the specimens was carried out using a scanning electron microscope (SEM). This allowed for the observation of the fiber-matrix interface and provided insights into the impregnation quality and interfacial characteristics.

The experimental setup involved multiple samples for each treatment variation and recycling cycle, ensuring statistical validity and reliable data analysis. The Taguchi Design of Experiments (DOE) approach was employed. The Taguchi method involves three main steps: parameter design, orthogonal array selection, and signal-to-noise ratio (SNR) analysis [22]. The key factors affecting the impregnation quality of the carbon fiber-reinforced waste polypropylene were recycling cycles and fiber treatment variations, each factor divided into three levels, as can be seen in Table 2. Here, an L_9 (3^2) orthogonal array was selected to design the experimental matrix. The orthogonal array ensures a balanced and efficient distribution of the parameter combinations, allowing for effective analysis of the main effects and interactions.

3. Result and discussion

The IFSS testing was conducted on 5 specimens for each variation of factors and levels, and the average IFSS values are presented in Table 3 and Fig. 3. The lowest IFSS result was observed in run 3 which was 8.6 MPa, while the highest IFSS result was obtained in run 7 which was 14.5 MPa. Higher values of IFSS correspond to better adhesion between the matrix and carbon.

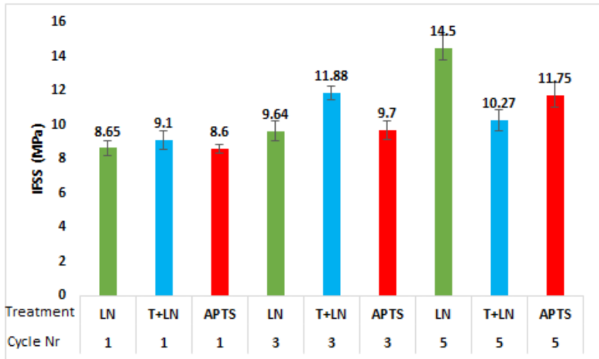


Fig. 3. Effect of number of recycling cycle and fiber treatment on IFSS.

The IFSS values were derived from the calculation of pull-out force versus displacement data using Eq (1). The evolution of force during pull-out for the highest and lowest IFSS is illustrated in Figure 4. In Figure 4(a), the fiber-matrix bond exhibits a higher resistance to applied force compared to Figure 4(b) before the occurrence of pull-out. This suggests that in Figure 4(a), the interface between the fiber and the matrix is stronger and more resilient, resulting in a higher IFSS value. Conversely, in Figure 4(b), the interface is less robust, leading to an easier pull-out and a lower IFSS value. These observations reflect the varying

Table 2. Factors and levels.

Factors	Level 1	Level 2	Level 3
Number of Recycle	1	3	5
Fiber treatment	Liquid Nitrogen	Thermal + Liquid Nitrogen	APTS

Table 3. Average IFSS and SNR.

RUN	Number of recycling	Fiber treatment	Average IFSS (MPa)	Signal-to-Noise Ratio
1	1	Liquid nitrogen	8.65	18.75
2	1	Thermal + Liquid nitrogen	9.10	19.18
3	1	APTS	8.60	18.69
4	3	Liquid nitrogen	9.64	19.68
5	3	Thermal + Liquid nitrogen	11.88	21.49
6	3	APTS	9.70	19.74
7	5	Liquid nitrogen	14.50	23.23
8	5	Thermal + Liquid nitrogen	10.27	20.23
9	5	APTS	11.75	21.40

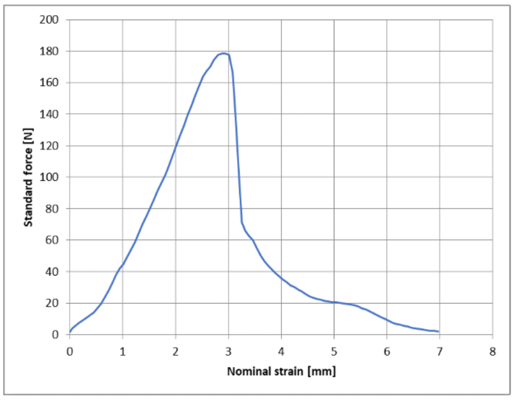
strengths of the fiber-matrix bonds and their impact on IFSS.

From the experimental results above, a Signal-to-Noise Ratio (SNR) analysis was performed. The SNR is a statistical measure used to evaluate the performance of a system or process by quantifying the ratio of the signal (desired response) to the noise (unwanted variation) [23]. In this analysis, the SNR was calculated based on the obtained IFSS values for each experimental run. The goal was to maximize the SNR, indicating a higher signal (IFSS). The Signal-to-Noise (S/N) ratio is categorized into three types: larger is better, smaller is better, and nominal is the best. The selection of the appropriate type of S/N ratio depends on the objective requirement. In this study, the larger is better type of S/N ratio, as described by Eq. (2), was utilized for the interfacial shear strength [24]. In this equation, yi represents the response value of a specific treatment with i replications, and n denotes the total number of replications. The results of the experiment is presented in Table 3.

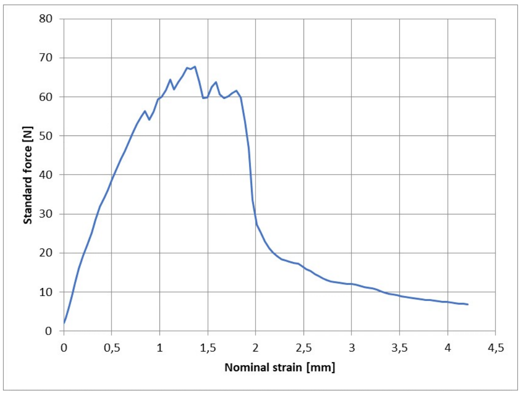
$$S/N = -10 \log \left((1/n) \Sigma \left(1/y^2 \right) \right) \tag{2}$$

In which y_i is the i^{th} experiment at the best test, n is the number of trials.

The main effect derived from the Signal-to-Noise Ratio (SNR) analysis provides insights into the individual contribution of each factor to the response variable. By evaluating the SNR values, it is possible to identify the relative importance of the factors influencing the response. The main effects can be obtained by comparing the average response values across different levels of a specific factor. A larger absolute value of the main effect indicates a more



(a)



(b)

Fig. 4. Evolution of force during pull-out: (a) Highest IFSS; (b) Lowest IFSS.

significant influence of the factor on the response. Table 4 presents the calculated main effects based on the analysis conducted.

Table 4. The main effect on IFSS based on SNR.

Level	Number of recycling	Fiber treatment
1	18.87	20.55
2	20.30	20.30
3	21.62	19.94
Delta	2.75	0.61
Rank	1	2

Based on the main effect analysis, the optimum contribution of parameter and their levels for achieving maximum IFSS are the recycling cycle at level 3 (5 times) and fiber treatment at level 1 (liquid nitrogen immersing). Once the optimal combination of process parameters and their level was obtained, the final step is to verify the estimated result. Confirmation testing was conducted using an injection molding machine with a matrix that underwent 5 recycling cycles, along with carbon fiber treatment involving immersion in liquid nitrogen. Table 5 represents the results of the confirmation testing compared to the highest result obtained in the previous experiments (in this case, Run 7). There is no significant difference between these two results, indicating that the combination of parameters can indeed yield the maximum IFSS (Interfacial Shear Strength).

The next step is to perform an Analysis of Variance (ANOVA). ANOVA operates under the fundamental concept of examining whether the variation in the outcome can be attributed to the distinct categories of the factors [25]. This enables us to conclude the meaningfulness of categorizing both factors in Table 4 based on their impact on the IFSS. The results of the ANOVA are presented in Table 6. The calculation included degrees of freedom (Df), a sequential sum of squares (Seq SS), an adjusted sum of squares (Adj SS), an adjusted mean square (Adj MS), an F-statistic from the adjusted mean square, and percentage contribution (p%).

From Table 6, one can observe the contribution of factors to the IFSS. The fiber treatment factor contributes 84% to the variation, which is larger than the contribution of the recycling factor. The fiber treatment can be attributed to the increased surface roughness of the carbon fiber, leading to improved adhesion between the carbon fiber and the matrix, thus resulting in a higher IFSS. Previous research by Budiyantoro et al. [13] has also indicated that the use of liquid nitrogen for carbon fiber surface treatment enhances surface roughness, leading to improved bonding between the fiber and the matrix.

Morphological observations were conducted to further confirm the above results. Figure 5 displays the scanning electron microscope (SEM) images of the post-pull-out test specimens. The specimen in Figure 5a was fabricated using a matrix recycled 5 times with carbon fiber reinforcement immersed in liquid nitrogen. It can be observed that the matrix effectively coats the fibers with minimal voids, which can be attributed to the rough surface of the carbon fibers after the treatment with liquid nitrogen. As a result, the matrix adheres well to the fibers, leading to a high IFSS value of 14.5 MPa.

Figure 5b illustrates the SEM results of the specimen with 3-time recycling and a combination treatment of initial heating and liquid nitrogen immersion. In this condition, the obtained IFSS value is only 11.8 MPa. Figure 5c displays the SEM results of the specimen with 1-time recycling and APTS fiber treatment, which exhibits the lowest IFSS value. Only a small amount of matrix adheres to the fibers, indicating poor fiber coating by the matrix. This can be attributed to the lack of surface roughness on the fibers, resulting in insufficient bonding between the matrix and the fibers. In this case, the observed phenomenon only yields an IFSS value of 8.6 MPa.

4. Conclusions

In conclusion, based on the conducted research, the following conclusions can be drawn:

- The impregnation quality of carbon fiber-reinforced waste polypropylene was investigated with variations in fiber treatment and matrix recycling cycles.
- The highest IFSS value of 14.5 MPa was achieved in the recycled PP material with a recycling cycle of 5 times, where the carbon fibers were immersed in liquid nitrogen. This indicates that the impregnation of carbon fibers using liquid nitrogen treatment resulted in better interfacial adhesion between the fiber and the resin.
- The lowest IFSS value of 8.6 MPa was found in the recycled PP material with a recycling cycle of 1 time, where the carbon fibers underwent a coupling agent treatment. This suggests that the coupling agent treatment did not effectively enhance the interfacial adhesion between the fiber and the resin.
- The SEM analysis confirmed that the liquid nitrogen treatment led to better impregnation of carbon fibers in the recycled matrix, resulting in improved adhesion between the fiber and the resin.
- The analysis of variance (ANOVA) indicated that the treatment of the fiber had the highest contribution

Table 5. Confirmation test results.

	Number of recycle	Fiber treatment	IFSS (MPa)
Run 7	5	Liquid nitrogen	14.50
Confirmation test			13.98

Table 6. Factor contribution to IFSS.

Factor	DF	Sq	Mq	F-ratio	p(%)
Number of recycle	2	11.325	5.662	3.61	12.7
Fiber treatment	2	1.473	0.736	0.18	84.4
Error	4	6.247	1.568		
Total	8	18.155			

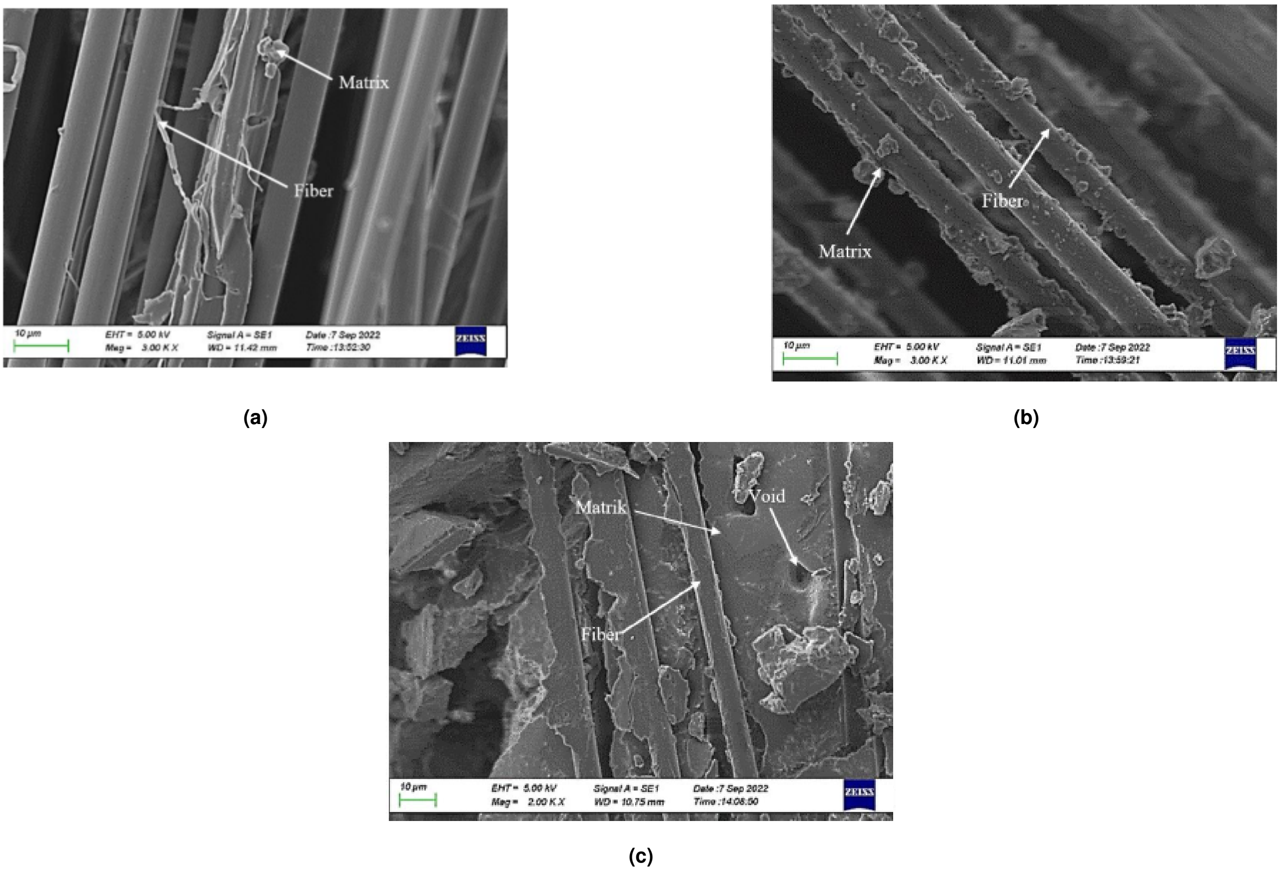


Fig. 5. Scanning images of pull-out test specimen: (a) 5 times recycled, liquid nitrogen; (b) 3 times recycled, Thermal + Liquid nitrogen; (c) 1 time recycled, APTS.

(84.4%) to the IFSS value, emphasizing its significant influence on the impregnation quality.

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