OPTIMUM CONSOLIDATION OF SELF-REINFORCING COMPOSITE AND ITS TIME DEPENDENT DEFORMATION

Kyoung Ju Kim[1], Woong-Ryeol Yu[1], Philip Harrison[2], Suk Jin Hong[1], Byoung Sun Lee[1], Hae Dong Park[1]

[1]Seoul National University-Department of Materials and Engineering, San 56-1, Sillim-dong, Gwank-gu, Seoul, 151-742, Korea
[2]University of Glasgow - Materials Engineering Group, Department of Mechanical Engineering, Glasgow, G12 8QQ, UK

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Abstract

Self reinforcing composites (SRC) are known to have a narrow processing window due to the same material system for matrix and reinforcement. In this study, a systematic study was carried out to determine the optimum consolidation process of SRC considering static and time dependent deformation behavior. It was shown that very little change in the processing condition influenced significantly the mechanical behavior of consolidated SRCs, e.g., the mechanical strength can be doubled within 10 degree change in temperature, the reason of which was scrutinized with structural analysis such as DSC and XRD and mechanical test such as T-peeling, tensile, and creep tests.

1. Introduction

Self reinforcing composite (SRC) is a new class of thermoplastic composite, in which the matrix and fibers are composed of the same polymer. SRCs are expected to retain good mechanical performance due to better interfacial bonding between fiber and matrix and furthermore to be free of recycling issues [1]. However, there has been little literature about processing and mechanical performance of this new kind of composite [2,3]. Therefore, this study aims at finding the optimized processing path to consolidate the SRCs and investigating their mechanical performance, in particular focusing on time dependent deformation.

The optimum consolidation of SRCs was studied using a hot press. Resulting molecular structure was determined using differential scanning calorimeter (DSC), wide-angle X-ray diffraction (WAXD), scanning electron microscope (SEM), and time-independent tensile test. Then, the time-dependent deformation behavior of the consolidated SRC was characterized, based on which a time dependent deformation theory is developed and applied to SRC, enabling to predict the performance of composite parts made from SRCs.

2. Experiments

2.1 Materials

In this study, woven preforms with self-reinforced polypropylene (PP) tapes from Don & Low Ltd were used. The tapes are a two component material consisting of dense and highly-crystalline drawn PP tapes (fibers), coated with a thin layer of low-crystalline PP co-polymer, which has a significantly lower glass transition and melting temperature than the drawn tapes and thus acts as the matrix phase of the SRC. In this consolidation process, the temperature control is a key factor because PP materials in the matrix should flow in-between fibers to surround the fibers and make continuous phase between fibers, which is the first criterion for effective composite performance.

2.2 Consolidated process of SRC

Self-reinforced composites were fabricated by firstly analyzing DSC thermogram in Fig. 1. It is found that the lower peak represents the melting of the copolymer used for the matrix, whereas the higher peak does the melting of PP homopolymer for the reinforcement. As shown in Fig. 1, candidate temperature for the consolidation process can be any one between the initial melting peak of copolymers and the one of the homopolymers, however, in the consolidation, a precise temperature control is required because the load transfer between matrix to fibers depends on the continuous matrix phase between fibers and the adhesion between matrix and fibers, which are determined by the flow behavior of the matrix.

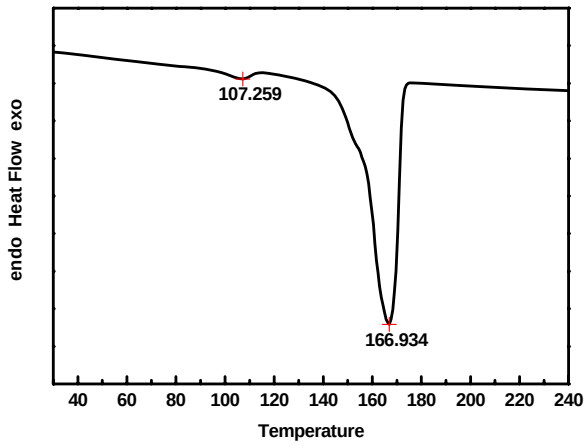

Fig. 1. DSC thermogram of SRC preform

After some preliminary tests, several consolidation temperatures (140℃, 147℃, 150℃ and 153℃) were selected, fixing other conditions such as pressure and time to be 100Kg/cm^2 and 30 minutes, respectively. Four layers of woven preforms ($[0]_4$) were then fabricated into a consolidated sheet using a hot-press. After the heat and pressure was applied to SRC preform, it was left in the mold for one hour inside two steel plates for stabilization. With these processing conditions, SRC performs were consolidated into composite sheet with 0.6 mm in thickness.

2.3 Characterization

2.3.1 Thermal and structural analysis

Calorimetric investigations were carried out through DSC thermogram. For this test, processed SRCs were heated from 30 to 250 ℃ under nitrogen atmosphere with a heating rate of 10 K/min. Wide angle X-ray diffraction (WAXD) analysis was also performed to investigate the crystal structure of polymers in consolidated SRCs. The SRC sheets were scanned from 5° to 30° in a 2Θ range at a rate of 5°/min using Model M18XHF-SRA X-ray diffractometer (Cu Kα radiation, λ = 1.54Å, generator voltage = 50kV, current = 200mA).

2.3.2 T-peel test

T-peel strength of laminated SRCs was determined by following ASTM-D1876 method. For the sample preparation of the peel test a thin film were introduced between SRC preform layers before consolidation. T-peel test was then performed by using a universal tensile tester (Instron 5543) at a rate of crosshead movement 120mm/min at room temperature. The peel strength (W) was calculated from

$$W = \frac{F}{L}$$

where F and L are the force applied to the joint and the width of the strip, respectively. The fracture surfaces of the peeled specimens were also examined by SEM microscope (JEOL Model JSM 5410LV).

2.3.3 Tensile test

Tensile properties were measured using the universal tensile tester at crosshead speeds of 10mm/min. The stress and strain curves were obtained for each SRC processed at different processing conditions. Anisotropic properties were also characterized by testing specimens at two directions (0 and 45 degree). The dumbbell-shaped specimens were prepared according to ASTM D638 and an extensometer was used for the accurate strain measurement.

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2.3.3 Creep test

Creep tests were conducted to investigate the time dependent deformation behavior of consolidated SRC, i.e. visco-elastic properties. A constant load (5MPa) was applied to SRCs for 10 hours at the room temperature. As in the tensile tests, anisotropy was investigated by performing creep tests in two directions, respectively.

3. Results and discussion

3.1 Thermal and structural properties

As shown in Fig. 2, two melting peaks around 106 ℃ and 166 ℃ were observed for DSC thermograms for each consolidated SRCs. The small peak at lower temperature represents the melting of matrix material while large peak at higher temperature does the melting of reinforcement homopolymer. Note that the processed SRCs show the doubling of the second peak, which is due to modified crystallite in the homopolymer by the thermal processing. The shoulder peak in the doubling moves to higher temperature as the processing temperature increases. This behavior can be explained by the secondary crystallization. In polymer, the structure may no be completely organized by only primary crystallization, i.e., the polymer structure can be improved by post-crystallization and re-crystallization processes, which are collectively designated as the secondary crystallization [4]. Since the second crystallization causes a peak to be doubled in general, SRCs used in this study seems to have undergone the second crystallization, which may be due to the crystallization of polymer molecules in the reinforcement fibers. This consideration can be confirmed by XRD analysis as follows.

In general, isotactic polypropylene (iPP) homopolymer has three crystal structure, i.e. α-, β- and γ-form, originated from molecular conformation. PP used in this study seems to have α-form crystal structure as shown Fig. 3. On the isothermal crystallization, α-modification may be formed, causing the doubling of the peak due to its crystallization [5].

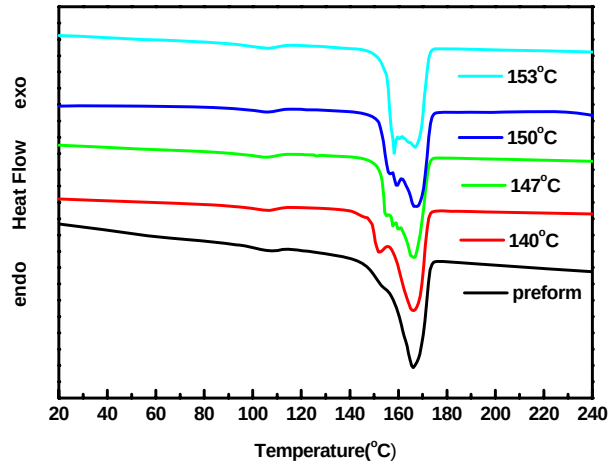

Fig. 2. DSC thermogram of SRCs according to the processing temperature

In Fig.3, WAXD profiles are compared for SRC preform and its consolidated sheets. Note that relative intensity of the second and third peaks based on the first one changes as the thermal processing was applied. The consolidated process seems to make the diffraction intensity around 21.3°(2θ) more relatively strong while it decreases the relative intensity around 21.3°(2θ). This intensity change may be due to the re-crystallization of PP by α-modification, which was also observed from the doubling peak in DSC thermogram.

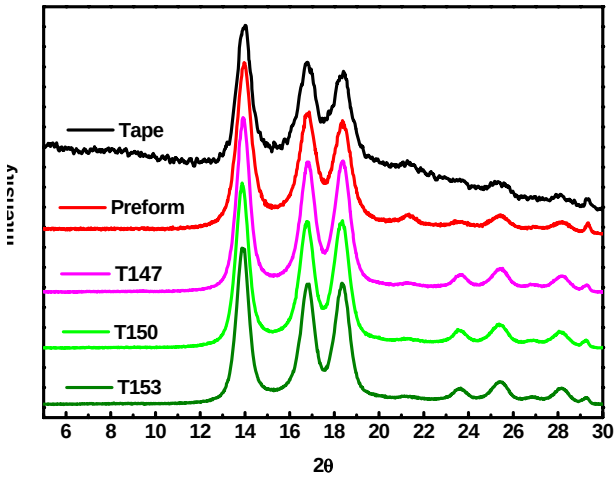

Fig. 3. WAXD patternof SRC preform and its consolidated sheet.

3.2 Interfacial property: T-peel test

Optimum consolidation process in the present work is a process that can make the copolymers in the preform melted out and flew in-between the reinforcement fibers. If a processing condition is close to the optimum process, it makes matrix interface between each preform layer continuous, resulting in good peeling strength. Thus it can be estimated by measuring the peeling strength whether a processing condition is preferable one or not. The peeling strengths were measured and compared for several consolidated SRCs in Fig. 4, demonstrating that the thermal processing at 150℃ brings better consolidation in SRC laminate than any other conditions.

The adhesion between each layer in SRC laminates can be assessed indirectly by analyzing the fracture surface of T-peel test specimens as shown in Figs. 5 and 6. Note that the surface morphology in Fig. 6 was obtained by magnifying the images in Fig. 5. There are two interfacial adhesions to determine the peel strength of the laminates: matrix-matrix and

matrix-fibers. If a temperature is not high enough to melt the surface coated polymers in the consolidation process, the adhesion between matrix and matrix of each layers becomes week, showing the matrix part more clearly than fibers on the fracture surface.

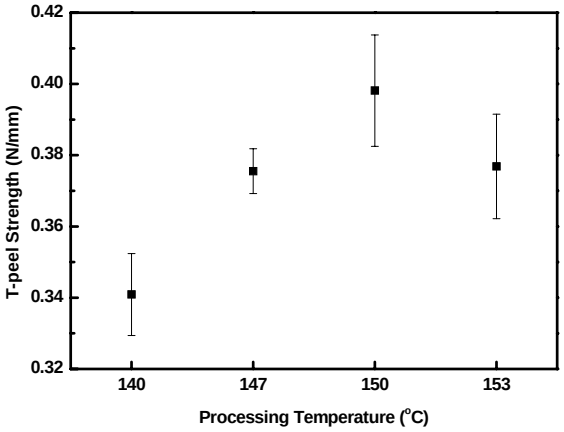

Fig. 4. T-peel strength for processing temperature

As shown in Fig. 5, higher temperatures than 150 ℃ seems to be required to ensure continuous matrix phase between each layer. Matrix and fiber adhesion should be strong enough for the applied load to be transformed onto the reinforcement fibers. As this adhesion gets stronger, the pulled-out fibers will be seen more frequently on the fracture surface. In this aspect, it can be concluded that thermal processing at 150 ℃ is a better consolidation condition than 153℃ because the pull-out fibers can be observed on the fracture surface of the SRC consolidated under the former condition.

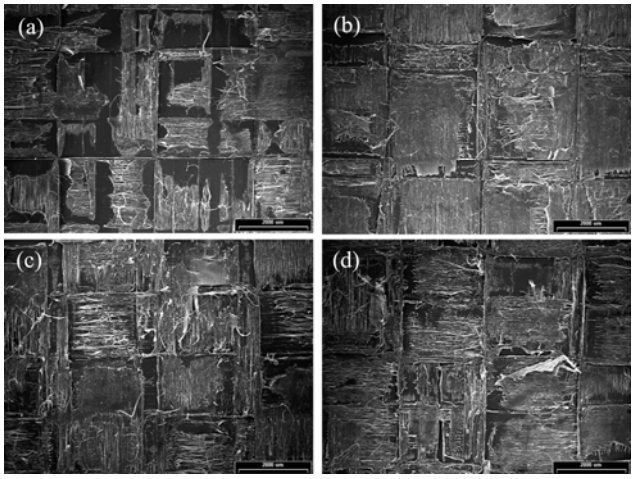

Fig. 5. Fracture surfaces of T-peel test specimens (x15). (a) 140℃, (b) 147℃, (c) 150℃, (d) 153℃

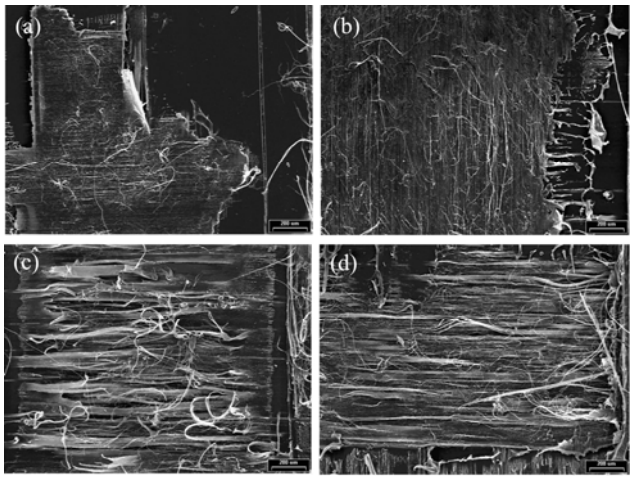

Fig. 6. Fracture surfaces of T-peel test specimens (x 75). (a) 140℃, (b) 147℃, (c) 150℃, (d) 153℃

3.3 Mechanical properties: tensile strength

The mechanical performance of consolidated SRC laminate was investigated by measuring tensile strength in two directions (0 and 45 degrees), respectively. As the processing temperatures were increased, the tensile strength in 0 degree direction was decreased (see Fig. 7). Note that the initial modulus was not presented in this paper because it was not varied that much according to the processing temperature. The decrease in the tensile strength can be explained by the relaxation of PP reinforcement fibers. It was observed that the thickness of consolidated SRCs was increased due to the shrinkage in the fiber direction as the temperature increased. During the shrinkage process, the re-crystallization was also involved (see double peaks in DSC thermogram in Fig. 2), reducing the molecular orientation in the PP fibers and subsequently tensile strength as well.

In 45 degree direction, however the trend is not the same as in 0 degree direction. This may be explained by observing the fracture mode. The fracture by tensile loading in 0 degree direction was governed by the matrix failure ecause the fibers inside composites was slipped and pulled out before their breakage in the 45 degree test. Note that all of the specimens used in the present study have two fiber orientations in 0 and 90 degree. As a result, the strength of consolidated SRC in 45 degree direction is determined not by fiber breakage but by matrix-matrix adhesion between each layer and also matrix-

fiber adhesion. Therefore the tensile strength in 45 degree direction follows the same tendency as those of the peel test and SEM analysis (see , Fig. 8), i.e. 150 ℃ is the optimum temperature among the candidate processing conditions.

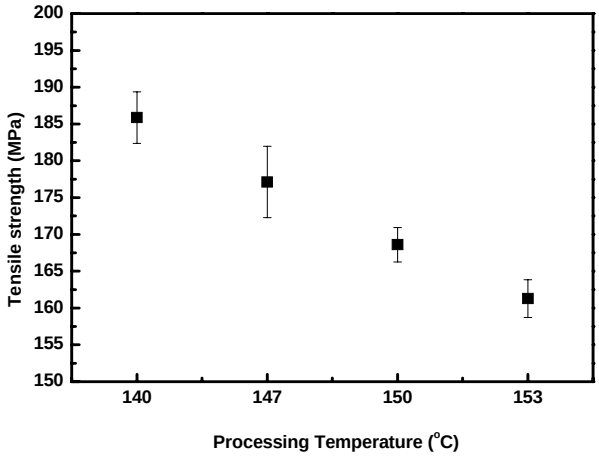

Fig. 7. Tensile strength of SRC in 0 degree direction according to the processing temperature

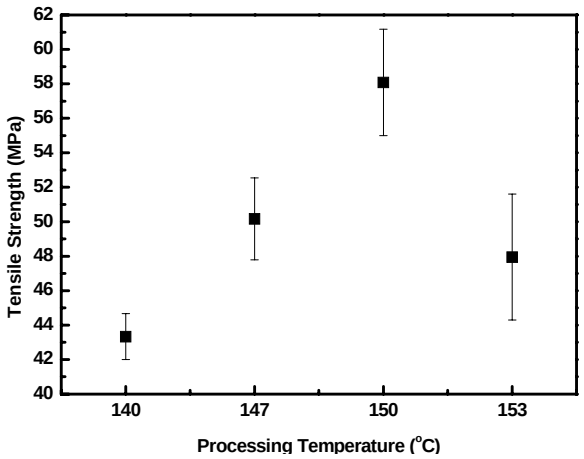

Fig. 8. Tensile strength of SRC in 45 degree direction for the processing temperature

3.4 Time dependent deformation: creep

Since SRCs in the present study consists of polymeric constituents, they are supposed to undergo time dependent deformation. The effect of processing conditions on the time dependent deformation was investigated by performing creep tests at a constant load of 5MPa, which was chosen to ensure that the creep deformation was occurred within the small deformation regime. As shown in Figs. 9 and 10, the creep resistance increased as the processing temperature increased. For 45 degree specimen, the same tendency as in the tensile and peel tests was exhibited because the main source of the deformation was the matrix elongation and shearing. Note that the creep resistance in 0 degree direction is the same as 45 degree case, which is not consistent with the tensile strength results. Since the current creep test was performed under the small constant load that can bring small deformation in SRC composites, the creep resistance in 0 degree direction seems to be determined by the matrix deformation.

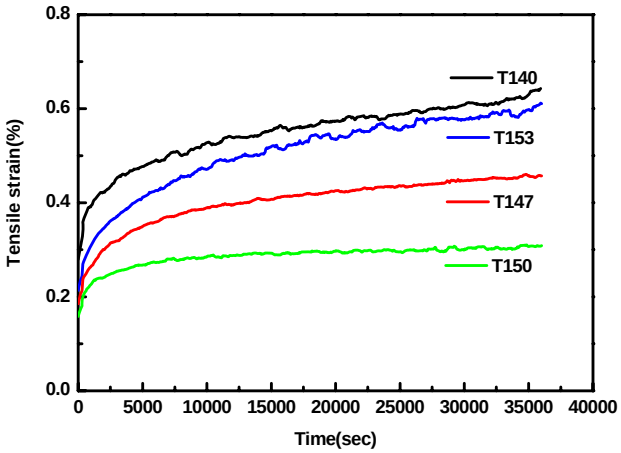

Fig. 9. Creep behaviors of SRC in 0 degree direction according to processing temperature.

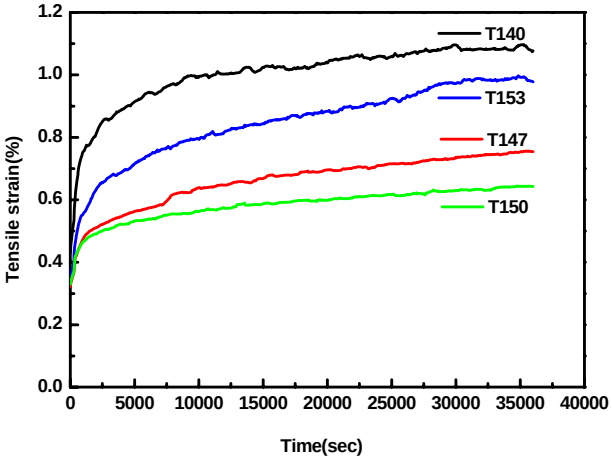

Fig. 10. Creep behaviors of SRC in 45 degree direction according to processing temperature.

4. Summary

Optimum consolidation process for SRC was investigated using DSC and XRD analysis, peeling and tensile tests, and creep test. Since thermal analysis such as DSC thermogram offers two broad

processing temperatures, other information should be provided to determine the optimum condition. In this study, it was shown that T-peeling test can be used to provide such information because it can reveal adhesions between SRC preform layers and between fibers and matrix, which is one of crucial factors to determine the mechanical performance of SRC composites. On the other hand, time dependent deformation behavior was observed to be anisotropic, which will be further characterized in order to formulate a visco-elastic model that can predict the time dependent deformation behavior of SRCs in multi-axial stress state.

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