

EFFECTS OF LAMINA CONSOLIDATION TEMPERATURE ON THE MECHANICAL PROPERTIES OF ALL-POLYESTER SELF-REINFORCED COMPOSITES

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Abstract

In this study, we proposed a modified film-stacking method, in which two-step consolidation for lamina and composite preparation was considered. Effects of the consolidation temperature for lamina and laminated srPET composites on the mechanical properties were examined. The mechanical properties were tensile, flexural and Izod impact tests.

1 Introduction

The self-reinforced polymeric composite material possesses many advantages and features, such as thermo-formability, high stiffness, high tensile strength, outstanding impact resistance at low density, and containing no glass [2–5]. Because the reinforcement and the matrix are compatible chemically; therefore, they usually have no interfacial problems. Additionally, the waste/ scrap materials can be recycled by melting which satisfies the demand for green material.

As we know that there is temperature gradient throughout thickness of the laminated composites during molding due to low thermal conductivity. Extending molding time to reach temperature equilibrium is often utilized. This consideration is important for molding the highly viscous thermoplastic matrix. Good impregnation in fiber reinforced thermoplastic composites can be achieved by lower the viscosity at elevated processing temperature. However, the major problem with the processing methods for manufacturing self-

reinforced polymeric composite material is damaging the reinforcement and matrix, while melting the matrix polymer during the forming process. Excessive heating results in fiber relaxation and ultimately causes the fiber to lose molecular orientation, whereas, insufficient impregnation leads to a poor interfacial bonding between the fiber and the matrix. The matrix degrades and embrittles with increasing of the consolidation temperature and time. According to our previous study [6], the optimal conditions for srPET composites are a low consolidation temperature and a short holding time for preventing the degradation of the polyester matrix.

However, a low consolidation temperature makes the resin more viscous, which results in a poor impregnation. On the contrary, higher consolidation temperature degrades the mechanical properties of the composites. Therefore, a balance between impregnation and degradation must be achieved. Meanwhile, it is of important that the processing window must be enlarged.

In this study, we proposed a modified film-stacking method, in which two-step consolidation for lamina and composite preparation was considered. The consolidation conditions for the lamina and the laminated composite can be independent. Because the lamina is rather thin, the problem of temperature uniformity is thus can be neglected. The time for lamina consolidation can be shortened which provide a room for increasing the consolidation temperature. Therefore, it is reasonable to suggest a better consolidation conditions: high temperature and short time, for srPET lamina preparation. It has

also been found that it is quite easy to control the temperature and pressure required for consolidating the lamina. Taking this advantage, we could optimize the process conditions for srPET composites with good impregnation and without degradation.

Effects of the consolidation temperature for lamina and laminated srPET composites on the mechanical properties were examined. The mechanical properties were tensile, flexural and Izod impact tests. The damaged specimens were inspected by microscopy to understand their failure modes.

2 Experiment

2.1 Materials

In this study, biodegradable polyester (Apexa® 4024, Dupont, Japan) in the form of pellets, were used as the matrix. The measured melting point of the biodegradable polyester determined by DSC is 198°C [6]. The recommended processing temperature in the guideline of the biodegradable polyester resin is approximately 215°C.

This study used a PET plain-woven construction fabric as reinforcement. The PET yarn for high denier industrial (HDI) purpose (grade: dope dyed color yarn) was provided by Far Eastern New Century Corporation, Taiwan (www.fenc.com), consisting of 2000 denier multifilament bundles with tenacity of 7.8 ± 0.3 g/denier and elongation of $14 \pm 2\%$. Every multifilament bundle consists of 182 filaments with filament diameter of 32 μm . The multifilament bundles were dope-dyed in black to imitate the appearance of carbon fabric. The PET plain-woven fabric has a fabric weight of 280 g/m². The warp and weft direction of the fabric (5 bundles/cm) balance reasonably well.

2.2 Sample preparation

This study presents a modified film stacking technique to produce high quality impregnated and void free srPET composites. At first, the matrix was prepared into a thin film form (thickness of 400 μm) using compression molding at 200°C for 3 min, at a pressure of 5 MPa. This was followed by quenching the matrix in water by covering it with Teflon films. The srPET composite lamina with average thickness of ~ 480 μm was prepared using the procedure: laid-up one layer of thermal treated PET fabric on PET

thin film at various temperature (200, 210, 220, and 230°C) for 1 min under a pressure of 5 MPa followed by quenching in water with the covering of Teflon films. The srPET laminates were prepared by stacking five layers of lamina at various temperature (200, 205, 210, 215, and 225°C) for 3 min at a pressure of 10 MPa followed by slow cooling to room temperature and demolded. In this experiment, the fiber volume fractions of the srPET composites were approximately 46%.

2.3 Mechanical Test

In this study, a universal testing machine (AG-100kNX, Shimadzu, Japan) was used to perform the tensile (lamina and composite) and three-point bending flexural tests at room temperature according to ASTM D638 (type I), D3039 and D790, respectively. The Izod impact test was performed at room temperature according to ASTM D256 on a pendulum impact tester (CPI, Atlas electric devices, USA) at impact energy of 5.4 J. The impact velocity used was 3.4 m/sec. The dimensions for the Izod impact specimen were $63.5 \times 12.7 \times 2$ mm³, and were provided with a 2.7 ± 0.2 mm deep notch. All the mechanical properties reported represent the average value of five readings at least.

3 Results and discussion

3.1 Effects of consolidation temperature of lamina

The mechanical properties of laminated composite which is composed of lamina are strongly influenced by lamina quality. For viscid thermoplastic matrix, the lamina quality depends on the number of voids, extent of impregnation, and extent of degradation of the matrix. As depicted in introduction, the flow distance and pressure distribution of thin lamina are smaller and more uniform than those of thick laminated composites. The time for lamina consolidation can be shortened which provide a room for increasing the consolidation temperature. Therefore, it is interesting to determine the optimal consolidation condition.

Fig. 1 shows the typical tensile stress-strain curves of the srPET lamina at different consolidation temperatures. The curves for all the srPET lamina show significant yielding and post-yield strain hardening, which are indicative of the reinforcing

effect and structural homogeneity of the srPET lamina. Good tensile properties (steep curve) for the samples prepared at higher consolidation temperature indicated that the srPET lamina structure was more compact at higher consolidation temperatures with lower viscosity and resulted in better impregnation. Impregnation is an important factor governing the mechanical properties of the srPET sample. The slope between the yielding point to the failure point (called the post-yield modulus) represents the reinforcing efficiency of the srPET composites. The post-yield modulus and thus the tensile strength increased with increasing consolidation temperature from 200 to 220°C. This increase was due to adequate impregnation at higher consolidation temperature. The yielding elongation for the srPET lamina is approximately 1.5%, which is near the failure elongation for pure polyester resin. This is clear evidence demonstrating the reinforcing effect of the srPET lamina. This result agrees with our previous study results on the srPET composites [6]. The tensile elongation of the srPET samples was approximately 14% for samples prepared at 200 to 220°C. However, premature fiber breakage was found in the srPET samples at 230°C. Such breakage was believed due to poor interfacial adhesion caused by the degradation and embrittlement of the polyester matrix [6]. Further, the embrittlement of the resin decreases the tensile strain, which in turn significantly decreases the tensile strength. However, for the srPET lamina reinforced by high tenacity PET fibers, the modulus of the srPET lamina depends on the fiber, and less on the matrix. Therefore, the tensile modulus and post-yield modulus for sample consolidated at 230°C were slightly decreased compared to the sample of 220°C. The tensile results show that the polyester matrix is sensitive to temperature above 220°C despite a limited consolidation time. Table 1 summarizes the Young's modulus, tensile strength, yield strength, and post-yield modulus of srPET lamina prepared at different consolidation temperatures. The srPET lamina consolidated at 220°C exhibits the highest modulus of 3.23 GPa, and a strength of 105.8 MPa, which are 21% and 24% greater than the values obtained at 200°C.

Two kinds of tensile damage were observed on the srPET lamina samples, namely shear breakage (Fig. 2a) at 200°C and breaking apart (Fig. 2b) at 220°C. The failure mode of the samples at 200°C was yarn

breakage and matrix fracture with widespread debonding. The failure was due to the poor impregnation between the matrix and the PET yarn at this low temperature. However, the srPET samples at 220°C were found to break apart, which confirms the adequate impregnation and the reinforcing structural integrity at this consolidation temperature. Therefore, the optimal temperature for manufacturing lamina was found to be 220°C and was utilized to prepare samples for srPET composite evaluation.

3.2 Effects of consolidation temperature of composite

The purposes for consolidation of laminated composites include bond together laminae, impregnation of the fiber bundle, removal of voids, and formation of a good interface. This section discusses the effect of consolidation temperature on the mechanical properties of srPET composites prepared using adequately impregnated lamina consolidated at 220°C.

Fig. 3 shows the typical tensile stress-strain curves of the srPET composites at different consolidation temperatures with a constant holding time (3 min). It was observed that the curves were similar in a temperature range of 200 to 215°C. Owing to the fact that good impregnation without void were accomplished by consolidating the lamina at 220°C, thus, the consolidation temperature of the srPET composites was less impact on its functionality. The results indicate that the purpose of consolidation process for srPET composites with good impregnation is to bond together every laminae. The impregnation and interfacial bonding between fiber and matrix were achieved during lamina process. However, high temperature (225°C) consolidated srPET composites caused premature breakage and weak tensile properties. This premature breakage was due to a poor interface between the fiber and matrix caused by the degradation and embrittlement of the polyester matrix. This is also the reason why the post-yield modulus is smallest at 225°C. The experimental values of the Young's modulus, tensile strength, yield strength, and post-yield modulus at different consolidation temperatures are summarized in Table 2. The values of the tensile modulus and strength did not vary significantly in the temperature range of

200 to 215°C. These findings suggest that the processing temperature of the composites could be reduced to 200°C. Therefore, wide processing window with 15°C consolidation temperature (200~215°C) for srPET composites was achieved. The srPET laminated composite sample consolidated at 215°C exhibited the best tensile properties, the tensile modulus and tensile strength is 3.71 GPa and 121.4 MPa, respectively. When the temperature was increased from 215 to 225°C, the tensile modulus and post-yield modulus values were slight decreased (Table 2). This decrease was due to the loss of orientation of the reinforced fiber and the degradation of the polyester matrix [7]. The tensile strength decreased from 121.4 to 95.5 MPa owing to the thermal degradation of the polyester resin and poor interfacial adhesion. This means that an increase in the composite's consolidation temperature and a longer holding time did not contribute to impregnation enhancement but caused matrix thermal degradation and poor interfacial adhesion. The tensile failure for the srPET composites consolidated from 200 to 215°C is breaking apart and that for srPET composites consolidated at 225°C is delamination; these failures are similar to those observed in our previous study [6].

In this study, higher consolidation temperature of 220°C were successful used to prepared the srPET lamina with good impregnation and interfacial bond. However, degradation occurs when consolidation temperature of 225°C were used to consolidate the srPET laminated composites which similar to that consolidated at 230°C for srPET laminae. Remember that the time required to process the composites (3 min) is longer than that required to process the lamina (1 min), owing to the presence of temperature and pressure gradients between laminates when processing composites. That is, the temperature and pressure are distributed evenly over the lamina owing to thinner thickness. A decrease in the time required for consolidation can also prevent the thermal degradation and embrittlement of polyester resins. When complete impregnation occurs during lamina processing (in which case the laminates just need to be adhered to each other during the processing of the composite), a large processing window (200~215°C) is obtained. However, when incomplete impregnation occurs, a

small processing window is obtained. On the basis of results above, it can find that the key procedure for srPET composites is lamina. However, good mechanical properties of srPET composites can be also obtained at consolidation temperature for lamina and composite of 200°C and 215°C, respectively [6]. This result indicates that good impregnation can be achieved at consolidation temperature above 215°C caused by lowering the viscosity. However, thermal degradation occurs when the consolidation temperature for srPET composites elevate to 225°C. Thus, the processing window for srPET composites laminated with poor impregnated laminae is very small (215~220°C) and unstable. Therefore, it is highly suggested to complete effective impregnation while preparing the lamina for a stable and wide processing window.

Table 3 provides a summary of the flexural and Izod impact values of the srPET composites at different consolidation temperatures and a constant processing temperature of lamina (220°C). Similar to the tensile results, the flexural modulus and impact energy of the srPET composites did not change significantly in the temperature range of 200 to 215°C. The srPET composites did not collapse within the crosshead limit (Fig. 4). This indicates the resistance of the reinforced woven fabric to crack propagation. This study did not find evidence for any visible failure in the bent srPET samples, which demonstrates its high toughness. In the temperature range from 200 to 215°C, the flexural properties of the srPET composite were similar. The flexural modulus and strength values of the srPET composite were highest in the case of consolidation at 215°C: 4.80 GPa and 83.5 MPa, respectively. Upon increasing the consolidation temperature from 215°C to 225°C, the flexural modulus and strength values decreased by 24 and 27%, respectively.

Table 3 also lists the resulting notched Izod impact energy of the srPET composites. The impact energy ranged from 840 to 891 J/m for the srPET samples prepared in the temperature range of 200 to 215°C which is similar the best srPET sample obtained in our previous study (854 J/m) [6]. Even though thermal degradation occurred below 225°C, absorbed impact energy as high as 811 J/m could be maintained. The impacted srPET composites did not break apart and showed tensile and compressive failures in two sides of the impacted specimen (Fig. 5). This failure exhibits the resistance of the

reinforced woven fabric to crack propagation. When an impactor encountered the srPET specimen, fiber-bundle breakage occurred around the notched side first, whereas the compressive force increased on the other side of the specimen. Owing to the laminated nature and integrity of the woven fabric architecture, fiber-bundle breakage occurred near the notch area and subsequent extensive delamination occurred along the interlaminar interface.

4 Conclusion

In summary, the processing temperature of srPET influenced the mechanical properties of the srPET composites significantly. Samples of srPET underwent thermal degradation at high temperatures owing to longer holding time during consolidation of composites than that for the lamina. When the srPET composites were processed in the temperature range of 200 to 215°C, no significant difference was observed in their mechanical properties, with adequate impregnation of lamina consolidated at 220°C. The modified film stack technique by 2-step consolidation can be used to achieve a balance between thermal degradation and impregnation. The optimal process conditions included a consolidation temperature of 220°C for the lamina and a subsequent wide consolidation temperature range between 200 to 215°C for the srPET composites.

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Table 1. Tensile properties for the srPET lamina produced at different consolidation temperatures

Temperature [°C]	Tensile strength [MPa]	Tensile modulus [GPa]	Yield strength [MPa]	Post-yield modulus [GPa]
200	85.0±1.9	2.67±0.06	27.8±1.91	0.50±0.30
210	93.3±6.6	3.27±0.06	32.3±0.62	0.56±0.01
220	105.8±3.0	3.23±0.13	33.8±0.45	0.54±0.01
230	95.3±4.1	3.07±0.14	30.3±1.52	0.50±0.01

Table 2. Tensile properties for the srPET composites produced at different consolidation temperatures

Temperature [°C]	Tensile strength [MPa]	Tensile modulus [GPa]	Yield strength [MPa]	Post-yield modulus [GPa]
220/200	117.1±4.1	3.75±0.02	27.8±0.2	0.59±0.04
220/205	112.6±6.3	3.78±0.02	26.4±1.8	0.51±0.02
220/210	108.5±3.3	3.44±0.13	26.3±1.0	0.53±0.04
220/215	121.4±3.9	3.71±0.02	28.5±1.3	0.53±0.05
220/225	95.5±2.7	3.30±0.08	24.8±1.5	0.42±0.04

Table 3. Flexural and impact properties for the srPET composites produced at different consolidation temperatures

Temperature [°C]	Flexural strength [MPa]	Flexural modulus [GPa]	Impact energy [J/m]
220/200	82.4±1.0	4.62±0.05	890.6±13.6
220/205	78.9±1.7	4.71±0.15	840.2±28.5
220/210	77.3±0.8	4.46±0.24	850.9±77.8
220/215	83.5±3.4	4.80±0.17	841.1±83.4
220/225	61.0±0.4	3.65±0.19	811.3±59.6

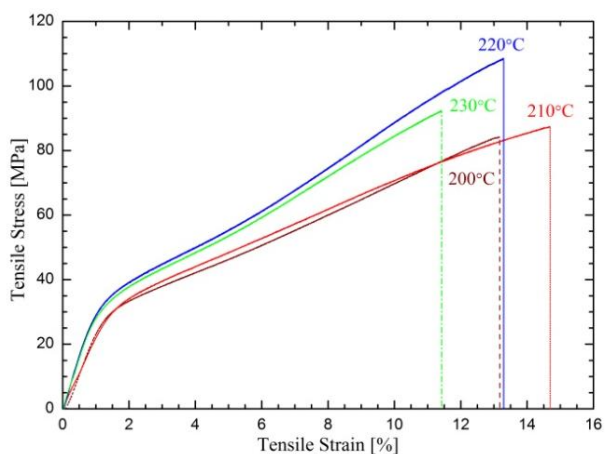


Fig.1. Typical tensile stress-strain curves of the srPET lamina at different consolidation temperatures

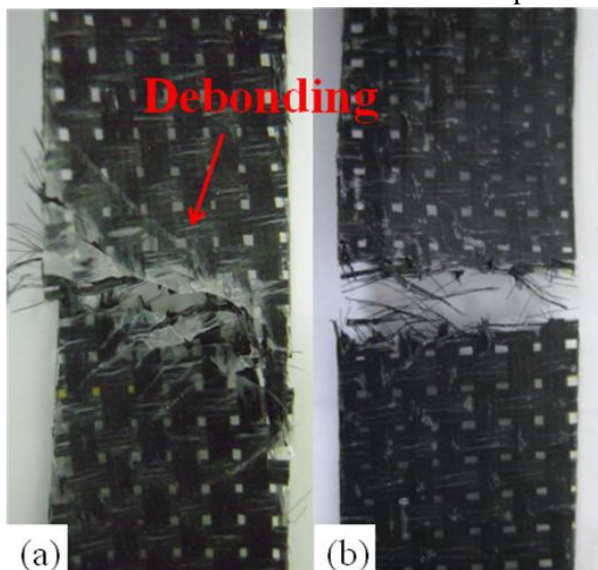


Fig.2. Typical tensile failure images for srPET lamina. (a) shear breakage damage for consolidation temperature at 200°C srPET sample, (b) break-apart damage for 220°C srPET sample

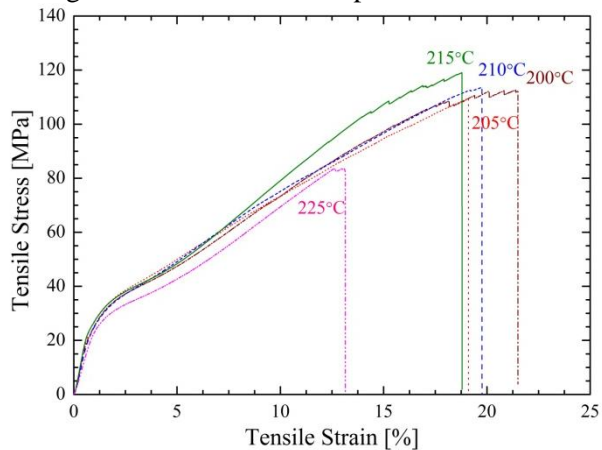


Fig.3. Typical tensile stress-strain curves of the srPET composites at different consolidation temperatures

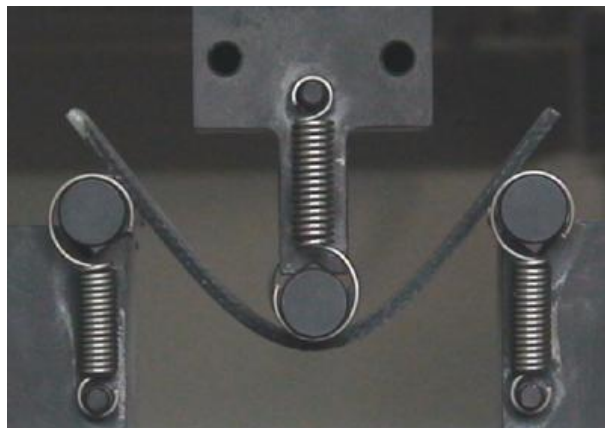


Fig.4. The image for srPET composite during flexural test.

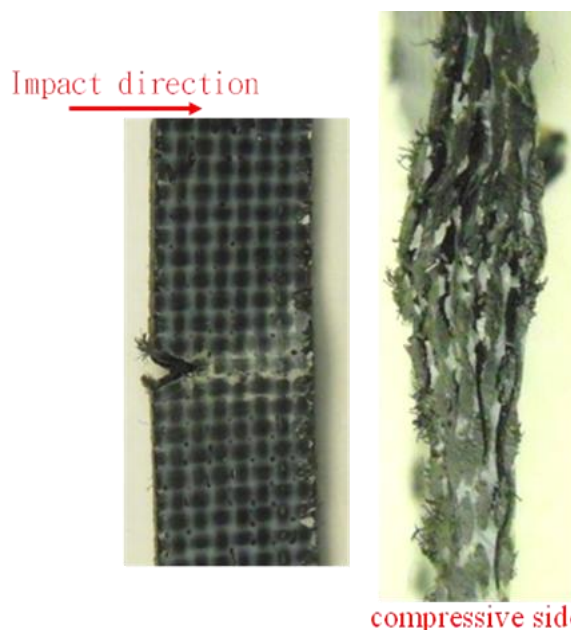


Fig.5. The impact failure images for srPET composites.