Effect of Knitted and Woven Textile Structures on the Mechanical Performance of Poly(lactic acid) Textile Insert Injection-Compression Moldings

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ABSTRACT: The main issue concerning poly(lactic acid) (PLA) has been its brittleness, thus limiting its potential to only a minute number of applications. Some attempts have been made to modify or blend PLA with tougher materials to improve its deformability, with varying degrees of success. This study primarily concentrates on enhancing the toughness of PLA without the incorporation of any additives whatsoever, thus the end product would consist of 100% biodegradable PLA. It is shown here that the toughness of PLA can be significantly enhanced by merely improvising processing techniques. The textile-insert molding technique was applied in this case, whereby PLA fabrics were attached to the surface of PLA resin by injection-compression molding. It was found that the molding conditions have an adverse effect on the interfacial adhesion between the fabrics and resin, which subsequently affect the impact performance of the moldings. The fabric was able to distribute impact loads toward a larger area thus enhancing impact resistance of the substrate resin. However, knitted and woven fabrics would impart different impact load dispersion characteristics due to their inherent variation in stretchability. © 2012 Wiley Periodicals, Inc. J Appl Polym Sci 000: 000–000, 2012

Key words: poly(lactic acid); toughness; textile insert molding

INTRODUCTION

Increasing awareness of the adverse environmental impact as well as the ever dwindling of available landfill space for disposing nondegradable polymeric waste has prompted many researchers and industrialists to turn to totally or semi biodegradable polymers for solutions. Typical fully biodegradable polymers that have been popular among researchers include polycaprolactone, poly(butylene succinate), and poly(L-lactic acid) (PLA).[1–5] PLA has been studied extensively for biomedical applications such as sutures and drug-releasing systems due to its good biodegradability and mechanical properties. Despite its high stiffness and strength, PLA tends to be brittle. This has led many researchers to either refrain from incorporating reinforcements, or would blend PLA with other tougher resins such as polycaprolactone and/or other plasticizers to obtain a more balanced property distribution.[6–9] However, this might affect the degradability of PLA as well as biocompatibility especially if the product were to be used for medical applications. Moreover, most of these toughening agents would significantly undermine the strength, stiffness, and clarity of PLA.

In this study, an attempt has been made to reinforce the PLA system by using a different processing approach without the addition of modifiers or plasticizers. Layers of PLA woven and or knitted fabric attached to a PLA substrate by means of injection-compression molding. This technique of layering a tougher material (in this case, the fabric) on brittle substrates (in this case, the injected resin) to provide high-impact resistance is not new, as it has been used in the manufacturing of bullet proof vest and laminated glass panes. However, the usage of the injection-compression molding technology to produce these laminates is a novel idea that can also be adopted for mass production since it has very short cycle times akin to conventional injection molding processes. This technology has been commonly used to produce thin substrates for optical media such as compact discs or DVDs[10,11] and optical devices such as lenses.[12] The purpose of this investigation is to gauge the extent of toughening that can be attained from the insertion of the fabric and also to determine the processing parameters during injection-compression molding that exert the largest influence on the toughness of the composite.

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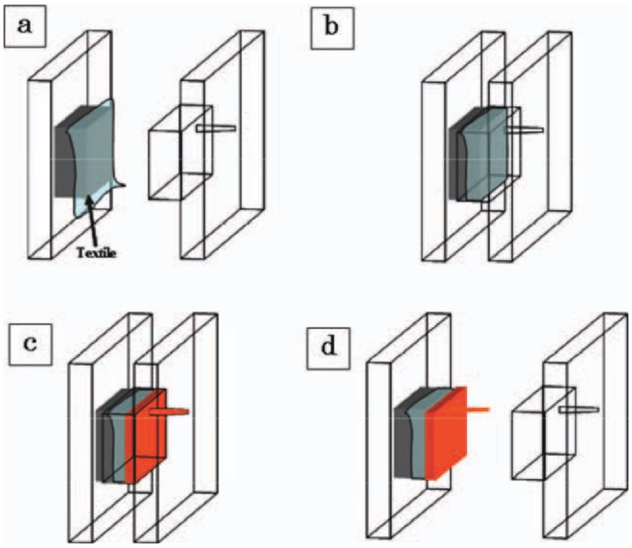

Figure 1 Textile insert injection molding sequence: (a) insertion of textile into mold; (b) mold closure leaving an opening; (c) injection and compression; and (d) mold opening end sample ejection. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

EXPERIMENTAL

Materials

PLA pellets (Unitika: TE 2000; M_w = 120,000) were used as the injected resin while PLA woven fabric (LACEA H100J: Mitsui Chemicals, Japan) with a thickness of 0.1 mm and density of 1.29 g/cm^3 was cut into 120 mm × 110 mm sheets and used as the inserts. PLA knitted fabric were fabricated by using a knitting machine with fibers spun from the same PLA material. Since the knitted fabrics would curl upon fabrication, they were compressed and flattened at 120°C for 3 h to ease handling. The thickness and density of the knitted fabrics were 0.5 mm and 1.31 g/cm^3, respectively.

Specimen preparation by injection-compression molding

Figure 1 illustrates the sequence of textile insert injection-compression molding. The textile insert was attached onto the square-plate (100 mm × 100 mm) SKD-11 hardened steel mold with double-sided tapes prior to the closure of the mold. The distance between the moving and stationary part of the mold was controlled so that there is a 5-mm gap prior to the injection of the resin. The resin would then be injected and the mold would close fully during the holding stage whereby compression of the resin would commence. The molded specimens would bear a final thickness of 3.0 ± 0.2 mm, with or without the presence of textile inserts. During the compression stage, the nozzle-valve was shut to prevent back flow of the resin into the barrel. The variables during injection-compression molding are described in Table I.

Textile inserts consist of either a single- or double-layered fabrics. Single-layered textile inserts comprise of either knitted or woven fabric while double layered inserts consist of a combination of both knitted and woven fabrics. The double layer fabrics were inserted in such a way that the knitted fabric would face the injected resin so that the resin could penetrate through the openings of the knitting and onto the surface of the woven fabric to cause adherence. A comparison between a woven and knitted structure can be found in Figure 2. In this circumstance, the woven fabric would form the outermost surface of the molding.

Mechanical testing

The molded plates were subjected to drop weight impact test using an Instron Dynatup 9250 HV impact tester. The height of the striking tup was adjusted to yield impact energy of 10 J. Upon impact, a load-deflection curve was obtained, with the area under the curve representing the impact energy absorbed by the specimens. Since the textile inserts were only attached to one surface of the specimens, impact tests were conducted either on the textile side or the substrate side. Textile insert specimens impacted on side of the knitted, woven or a combination of both fabrics are designated as knitted-up, woven-up, and double-up, respectively, whereas specimens impacted on the substrate side are known as knitted-down, woven-down, and double-down, respectively. Specimen configurations during impact tests are schematically represented in Figure 3. A load cell attached to the striking tup would record the impact loads throughout the period of specimen deformation. At least five specimens were tested at each condition to ensure repeatability of the results.

Three-point-bending tests were also performed on the textile insert moldings by using an Instron 4466 Universal Testing Machine equipped with a 5 kN load cell. The span length was set at 48 mm while testing speed was 3 mm/min. The loading configuration during the bending test was similar to that of the drop weight impact test, as previously indicated in Figure 3.

TABLE I
Molding Conditions

Barrel temperature (°C)	170, 190, 210
Compression distance (mm)	5
Compression time (s)	0.1, 1.0, 3.0
Injection speed (mm/s)	100
Mold temperature (°C)	30

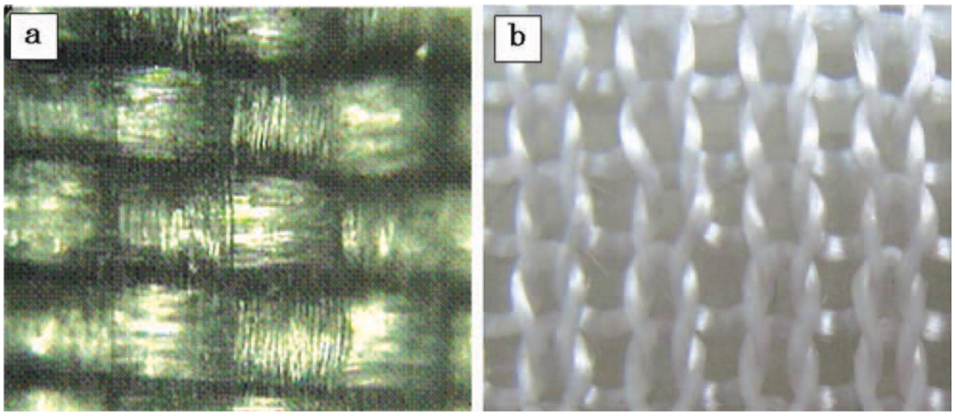

Figure 2 Structure of (a) woven and (b) knitted textile inserts. Magnification = ×100. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

RESULTS AND DISCUSSION

Effect of molding conditions on appearance of textile insert specimens

The effects of the compression time and barrel temperature on the appearance and quality of specimens can be seen from Figure 4. Significant tearing of the textile inserts could be observed due to the presence of high-shear stresses from resin flow when low barrel temperatures or short compression times (rapid compression) were used. However, significant improvements can be seen when the compression time was prolonged to 3.0 s and good specimens could be obtained irrespective of barrel temperature. Therefore, all specimens were prepared at a compression time of 3.0 s while various barrel temperatures were applied to vary the adhesion between the resin and textile inserts.

Flexural properties of textile insert moldings

The effect of textile inserts on the flexural stiffness and strength of the moldings are clearly presented in Table II. It can be seen that the attachment of the textile inserts did not significantly affect the flexural properties of the moldings, even though the fiber volume fraction especially in the knitted and double textile insert moldings were quite high. This is because the strength and stiffness of the textiles were quite low since they were only attached to the surface of the moldings. Moreover, the textiles would easily stretch (as in knitted fabrics) or tear (as

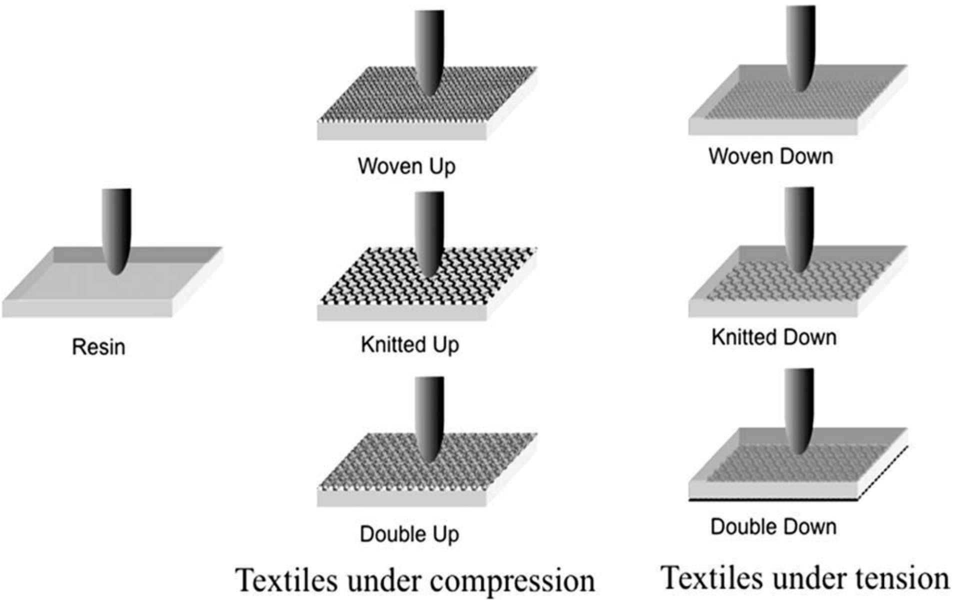

Figure 3 Specimen configuration and the corresponding impact test direction.

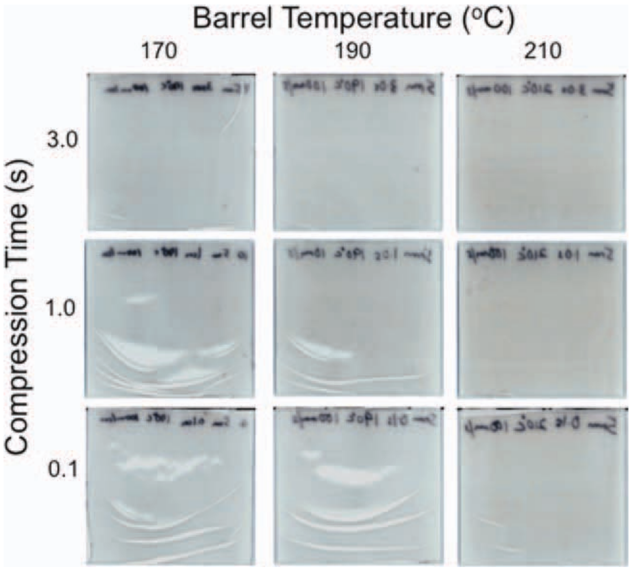

Figure 4 Effect of barrel temperature and mold compression time on the quality of textile insert moldings. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

in woven fabrics) upon tension; hence, they were unable to act as effective reinforcements for the bulk during static loading.

Impact performance and fracture characteristics of woven textile insert moldings

To elucidate the fracture mechanism of textile insert moldings during impact loading, the characteristic load-deflection curves obtained during impact tests were separated into two parts, i.e., energy to first maximum load (E_L) and progressive energy (E_P). E_L was determined from the area under the load-deflection curve until the first maximum load, which represents the energy required to initiate fracture in the specimen. Meanwhile, E_P was determined from the remaining area under the load-displacement curve and is defined to be the energy absorbed during bending, interfacial delamination and fiber fracture after the initiation of fracture. The sum of E_L and E_P would indicate the total impact energy absorbed by the specimen, as shown in Figure 5.

TABLE 2
Fiber volume fraction in the textile insert moldings and their corresponding flexural properties

Specimen	Fiber Volume Fraction (%)	Flexural Modulus (GPa)	Flexural Strength (MPa)
Resin	0	4.6	120.5
Woven-Up	15.7	5.5	114.7
Woven-Down	15.7	4.7	119.3
Knitted-Up	53.4	5.1	111.7
Knitted-Down	53.4	4.7	93.2
Double-Up	69.1	4.4	101.8
Double-Down	69.1	4.3	102.1

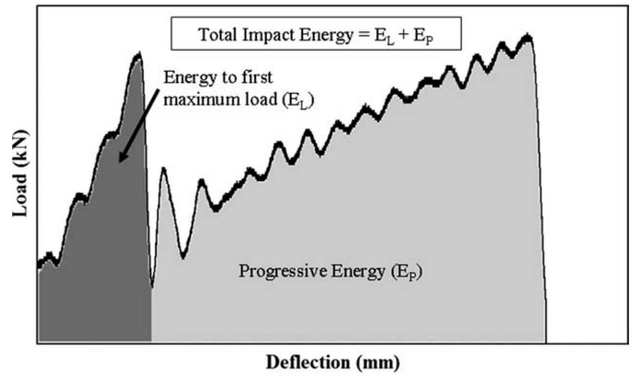

Figure 5 Typical load-deflection curve during impact test and the relative energies defined by the fracture characteristics of the specimens.

Figures 6 and 7 show the respective E_L and E_P values when impact tests were performed on the woven textile insert moldings prepared at various barrel temperatures. The specimens exhibit very low E_L values regardless of barrel temperature and impact configuration, which indicates that the woven textiles were unable to improve the fracture resistance of the substrate even when the specimens were impacted in the woven-down direction. This could be due to the rigid woven structure of the textile insert that did not provide sufficient free space for stretching of the fiber bundles during impact loading. However, the woven textile contributed significantly toward enhancing the E_P, which was primarily due to textile-substrate delamination in the case of woven-up configuration or fiber fracture in the case of woven-down configuration. The higher E_P recorded for woven-up specimens suggest that delamination plays a more dominant role in impact energy absorption than fiber-fracture. With increasing barrel temperature, the resin was able to further

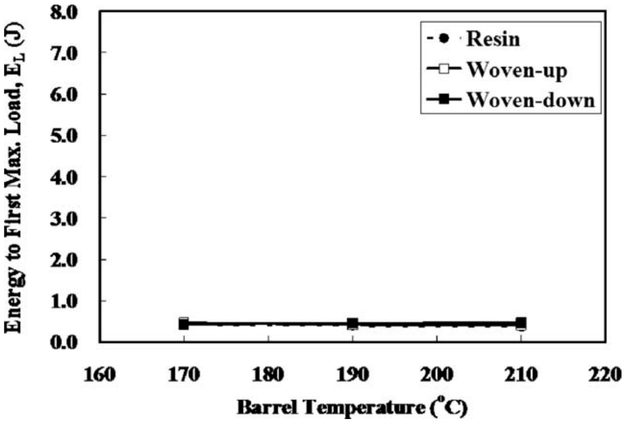

Figure 6 Effect of barrel temperature and impact configuration on the energy to first maximum load for woven textile-insert specimens and PLA resin substrate.

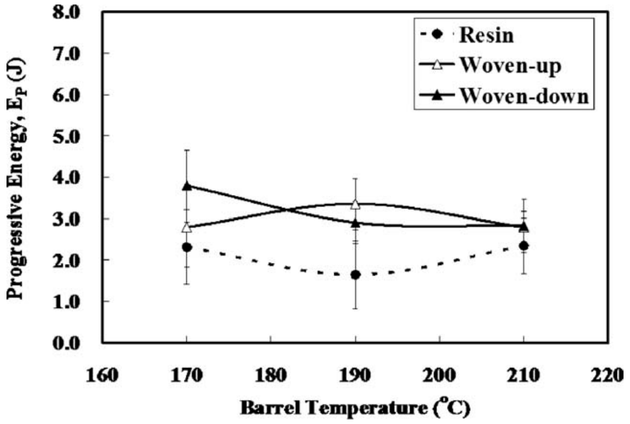

Figure 7 Effect of barrel temperature and impact configuration on the progressive energy for woven textile-insert specimens and PLA resin substrate.

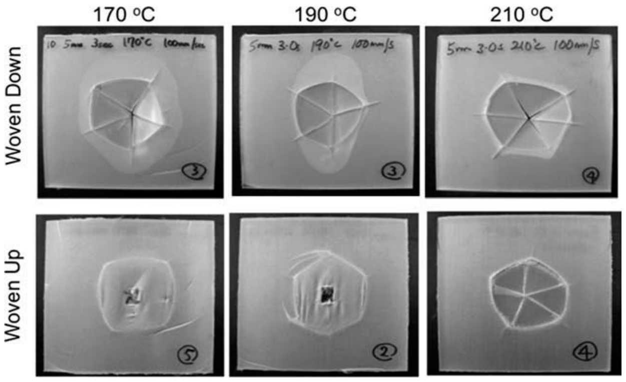

Figure 9 Fracture appearance of woven textile insert moldings prepared at various barrel temperatures and impacted at various configurations.

melt the textile surface and improve interfacial adhesion although this would also result in the weakening of fiber bundles in the textile insert. This contributed to the reduction in E_P values for both the woven-up and woven-down specimens due to less delamination and lower resistance to textile tearing as barrel temperature was increased.

The total impact energy absorbed during impact loading of woven textile insert specimens is depicted in Figure 8. An improvement in impact energy absorption of up to 40% could be seen in woven textile insert moldings as compared with the resin substrate. The woven textile insert moldings were generally more susceptible to impact loadings in the woven-down configuration especially when they were prepared at high barrel temperatures. The tup would initially puncture the woven fabric and proceed to cause extensive delamination at the textile-substrate interface prior to the fracture of the substrate, as can be seen from Figure 9. Although the occurrence of delamination is an indication of weak textile-substrate interface due to insufficient melting of the textile surface by the injected resin, it was also an effective impact energy absorption mechanism. Hence, higher impact energies were recorded whenever large delaminated areas were observed in the specimens upon impact. The textile inserts were also effective in dispersing loads if the specimens were impacted in the woven-up configuration.

When higher barrel temperatures were used, such delamination was absent thus indicating an improvement in textile-substrate adhesion although a reduction in impact energy absorption was also imminent. Moreover, tearing of the woven textile along the crack propagation path in the substrate was observed. Since the woven textile was only about 200-μm thick, the tear strength was considerably low and did not contribute significantly toward impact energy absorption.

Impact performance and fracture characteristics of knitted textile insert moldings

Figures 10 and 11 show vastly different E_L and E_P values for knitted textile insert moldings as

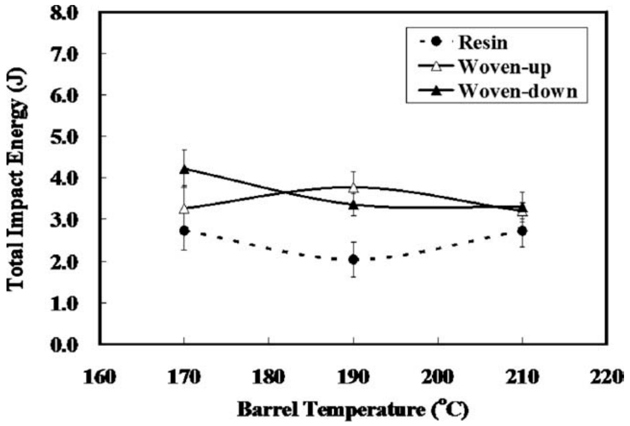

Figure 8 Effect of barrel temperature and impact configuration on the total impact energy for woven textile-insert specimens and PLA resin substrate.

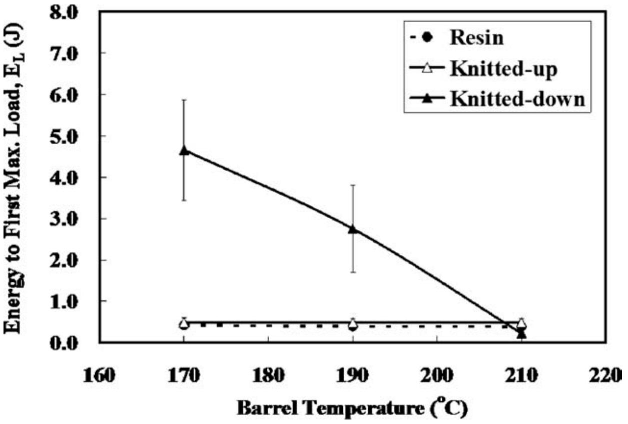

Figure 10 Effect of barrel temperature and impact configuration on the energy to first maximum load for knitted textile-insert specimens and PLA resin substrate.

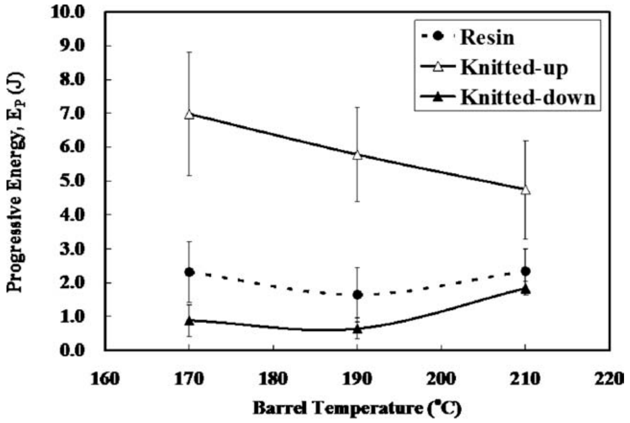

Figure 11 Effect of barrel temperature and impact configuration on the progressive energy for knitted textile-insert specimens and PLA resin substrate.

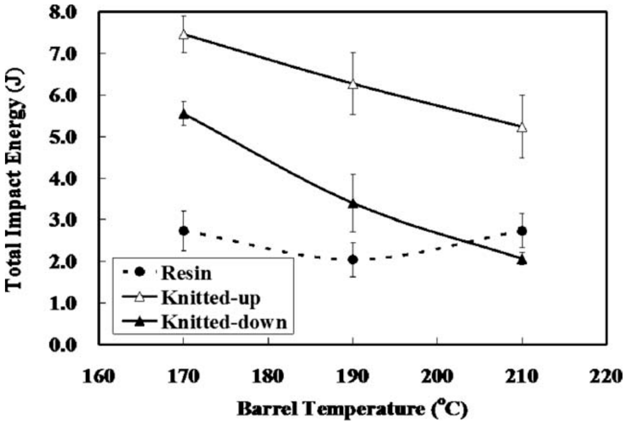

Figure 12 Effect of barrel temperature and impact configuration on the total impact energy for knitted textile-insert specimens and PLA resin substrate.

compared with the woven specimens. When the knitted textile insert specimens were impacted in the knitted-down configuration, substantially higher E_L values were recorded as compared with the substrate resin alone, more so when the specimens were prepared at low barrel temperatures. The ability of the knitted textile to stretch under tension provided substantial support to the substrate during impact loading. As noted in Figure 11, the specimens impacted in the knitted-down configuration would exhibit very low E_P values thus suggesting that most of the load-bearing mechanisms were exhausted prior to and during the initiation of fracture Therefore, it is thought that fiber fracture in the knitted textile inserts would occur simultaneously with substrate fracture. This is only possible if the fiber bundles in the textile insert retain sufficient strength and flexibility even after being exposed to the heat from the resin during injection molding. At high-barrel temperatures (above 190°C), however, the injected resin would partially melt the fiber bundles in the textile, thus significantly reducing its strength and flexibility. In this case, the textile would not be able to contribute effectively toward impact energy absorption, which explains the reduction in E_L values in the textile insert specimens as barrel temperature increases.

In contrast, the knitted-up specimens would exhibit low E_L but high E_P. This indicates that the knitted textile was only effective in impact energy absorption after the initial fracture in the substrate has occurred. It is also interesting to note that the E_P in knitted-up specimens was significantly higher than the E_L in knitted-down specimens. The higher E_P in knitted-up specimens is attributed to a few factors; one of which

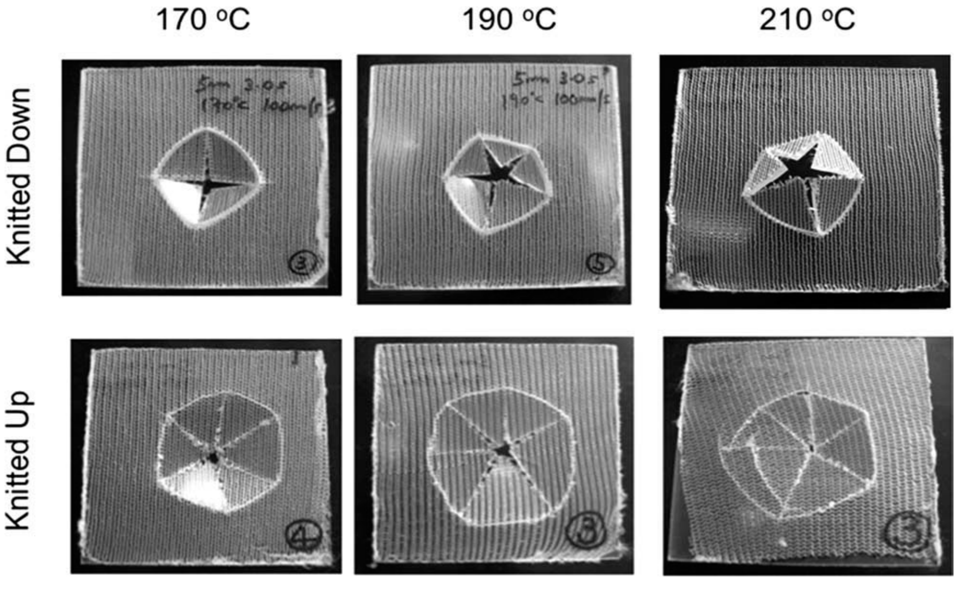

Figure 13 Fracture appearance of knitted textile insert moldings prepared at various barrel temperatures and impacted at various configurations.

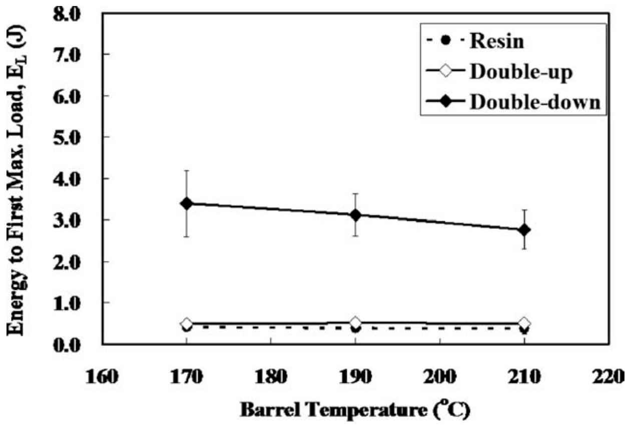

Figure 14 Effect of barrel temperature and impact configuration on the energy to first maximum load for double textile-insert specimens and PLA resin substrate.

is the ability of the textile to stretch further without being damaged by the sharp edges of the fractured substrate such as in the knitted-down configuration. Another reason would be the ability of the knitted textile to disperse impact loads toward a wider area, which improves the efficiency of impact energy absorption by the substrate resin.

The total impact energy absorbed by knitted textile insert moldings is depicted in Figure 12. The knitted textile insert specimens were able to resist much higher impact loadings as compared with the resin substrate especially when they were impacted in the knitted-up configuration. However, similar textile-substrate interfacial delamination found in woven textile insert moldings could not be observed from impact fractured photographs of knitted textile insert moldings depicted in Figure 13. Therefore, in the case of knitted textile insert moldings, the impact energy was largely dissipated by textile stretching, effective dispersion of impact loading, substrate crack propagation, and fiber rupture in the textile insert.

When the specimen was impacted on the substrate side (knitted-down), smaller radial and circumferential cracks were observed (Fig. 13), indicating that the distribution of impact load was not as effective as in the knitted-up configuration. It is thought that the impact energy absorption mechanism in the knitted-down configuration consist of mainly substrate crack propagation and fiber rupture.

Impact performance and fracture characteristics of knitted-woven double-layered textile insert moldings

When both woven and knitted textile inserts were combined and incorporated as double-layered textile inserts, a distinct improvement in impact performance could be observed from Figures 14 and 15. The impact fracture behavior of double textile insert specimens were similar to that of knitted textile insert moldings whereby high E_L and E_P values were achieved when the specimens were impacted in the double-down and double-up configurations, respectively. However, it is interesting to note that these specimens were able to retain high E_L and E_P values even at high barrel temperatures when they were impacted in the double-down and double-up configurations, respectively. This suggests that both knitted and woven textiles were able to contribute their positive characteristics toward improving the impact energy absorption characteristics of the molding. The strength of the woven textile was not affected since it was protected by the knitted textile from direct contact with the hot injected resin. As such, the woven textile was able to effectively disperse impact loads in both the double-up and double-down impact configurations.

The total impact energy absorption characteristics of double textile insert moldings are illustrated in Figure 16. It can be seen that the double textile insert

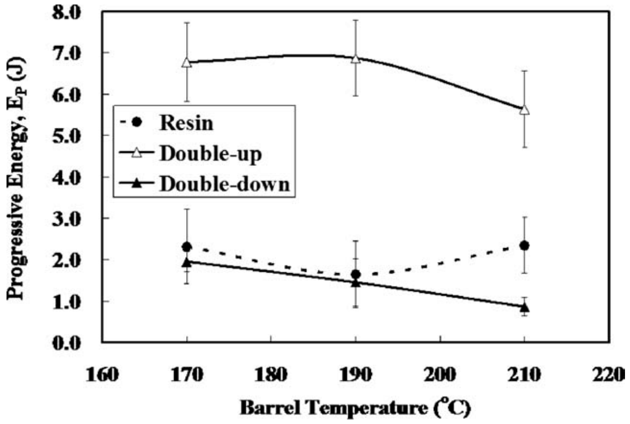

Figure 15 Effect of barrel temperature and impact configuration on the progressive energy for double textile-insert specimens and PLA resin substrate.

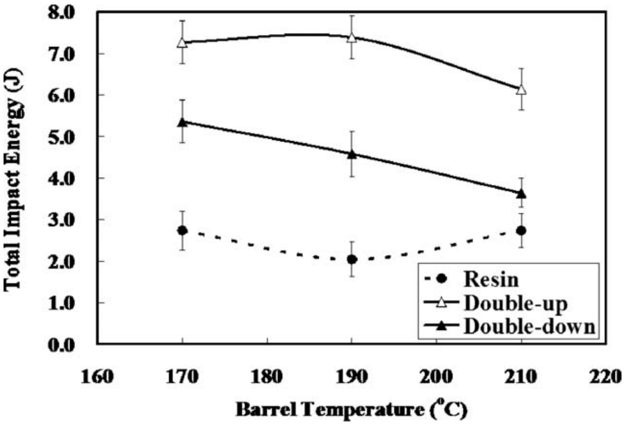

Figure 16 Effect of barrel temperature and impact configuration on the total impact energy for double textile-insert specimens and PLA resin substrate.

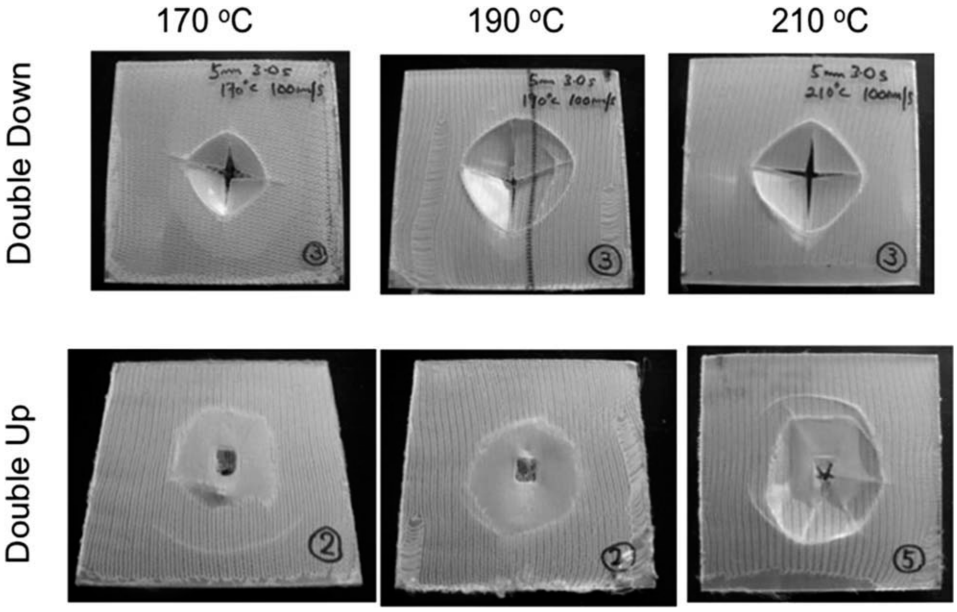

Figure 17 Fracture appearance of double textile insert moldings prepared at various barrel temperatures and impacted at various configurations.

moldings recorded up to 400% improvement in impact resistance as compared with the neat resin. It is noteworthy that the impact resistance was very high irrespective of whether the specimens were impacted in the double-up or double-down configuration when the specimens were prepared at 170°C barrel temperature. This improvement in impact properties is largely attributed to the enhanced heat resistance of the double-layer textile insert, which enabled the textile inserts to effectively act as impact load bearing and dispersion agents. From Figure 17, it can be seen that specimens impacted in the double-down configuration would experience a combination of substrate cracking, delamination at the textile-substrate interface, textile stretching, and fiber rupture. The extent of delamination decreases with increasing barrel temperature. However, in the case of double-up configuration, delamination was observed in all specimens irrespective of barrel temperature. This delamination, combined with other impact energy absorption mechanisms such as substrate cracking, textile stretching and fiber rupture effectively contributed toward the enhancement in impact energy absorption.

CONCLUSIONS

The textile insert injection-compression molding technique by using similar materials for both the substrate and textile insert was proven to be a viable option to enhance the impact resistance of an inherently brittle material. This could be a better option for enhancing the toughness of moldings as opposed to other techniques that involve the incorporation of foreign additives, which could compromise on the biodegradability and thermal properties of the material. Results show the importance of preserving the integrity of the textile insert in order for it to be effective as impact energy absorption and distribution agent, thus low barrel temperatures were preferable. Knitted textile inserts were able to impart higher impact resistance to the substrate due to their ability to stretch upon tension. Meanwhile, delamination between the textile and substrate was also an effective impact absorption mechanism found in woven textile insert moldings. Therefore, with a combination of both woven and knitted textiles in double textile insert moldings, significant improvement in impact resistance was achieved.

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