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# **Reusing Textile Waste as Reinforcements in Composites**

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**ABSTRACT:** Polyester (PET) has wide applications in textile industries as textile fiber and its share continues to grow. Substantial quantities of cotton/polyester blend fabrics are disposed every year due to technical challenges, which pose a big environmental and waste-dumping problem. The aim of this study is to evaluate the potential of discarded cotton/PET fabrics as raw materials for composites. If their inherent reinforcement properties can be used in composites, an ecological footprint issue can be solved. In this study, we investigate three concepts for reuse of cotton/PET fabrics for composites: compression molding above the  $T_m$  of PETs, use of a matrix derived from renewable soybean oil, use of thermoplastic copolyester/polyester bi-component fibers as matrix. All three concepts have been explored to make them available for wider applications. The effects of processing parameters such as compression temperature, time and pressure are considered in all three cases. The third concept gives the most appealing properties, which combine good tensile properties with toughness; more than four times better tensile strength than the first concept; and 2.2 times better than the second concept. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40687.

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#### INTRODUCTION

Reusing valuable resources in more feasible way contributes to sustainable living. There are several ongoing lines of research for reuse of products made of nonrenewable resources.<sup>1</sup> Polyethylene terephthalate (PET) is a widely used thermoplastic polymer made from nonrenewable petroleum-based raw material. PET is used extensively for beverage bottles and textile fibers. While PET bottles are recycled to a large extent, recycling of PET fabrics is much more challenging, particularly because they are generally mixed with other fibers.<sup>1</sup> A large portion of PET ( $\sim$ 39 million tons of PET out of 49 million tons used in 2008) was used in the textile industry. About 74% of the PET used in the textile industry is used to make staple fibers. These staple fibers are mostly mixed together with cotton fibers to produce cotton/PET blend fabrics.<sup>2,3</sup> These blended fabrics must be disposed of after use and they are often incinerated to regain energy, but the most part ends up landfills.<sup>2</sup> The degradation of PET is slow, and there are environmental problems. In addition, the valuable nonrenewable petroleum-based resource is wasted.

There are ways of extracting PET from cotton/PET blend fabrics, but these methods are still not economically feasible.<sup>4,5</sup> PET from sources such as bottles and fibers is recycled in different ways; melting of PET,<sup>6–8</sup> glycolysis,<sup>9</sup> hydrolysis,<sup>10</sup> and methanolysis.<sup>11</sup> However, these techniques cannot be used to recycle cotton/PET blends in fabrics, which are most often present at a 1:1 ratio. Mixtures of cotton and PET in fabrics cannot be separated mechanically. Another method is to dissolve PET from cotton/PET blend fabrics; this is possible in practical sense, but it is not economically feasible because PET has a limited number of solvents-which are both expensive and industrially inappropriate.<sup>2</sup> There is also the possibility to extracting cellulose from the cotton/PET blend fabrics and recycling the cellulose to viscose (or by similar process) and also recycling the PET residue. However, making this route industrially feasible also appears to be quite challenging, both from a technical and economic point of view. It is necessary to find alternative methods that are less expensive, to recycle/reuse, and handle cotton/PET blend fabrics. The possible recycling options that are available have the effect that the material would lose its value in some or several respects, i.e., be downgraded. Thermodynamically, the entropy inevitably increases and a substantial part of the energy put into the fabric is lost. Efforts to make a woven material of high regularity are wasted if a woven fabric is shredded. If the discarded fabric can be used as is, and be further refined to make a new construct, downgrading is avoided.

This study investigates ways of reusing discarded cotton/PET blend fabrics as reinforcement in composites. Three compression molding concepts were evaluated. In the first method, the

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Composite	Abbreviation	Matrix	Fiber: Matrix ratio	Mold temperature (°C)	Mold pressure (kPa)	Molding time (min)
Type 1		PET from fabric itself				
(i) Without plasticizer	1		50 : 50	270, 280, 290	20	0 : 20, 0 : 30, 0 : 40
(ii) With plasticizer	1P		50 : 50	270, 280, 290	20	0:20,0:30,0:40
Type 2	2	Soybean oil based thermoset resin	50 : 50	170, 185, 200	160, 180, 200	5, 7, 10
			60 : 40	170, 185, 200	160, 180, 200	5, 7, 10
			70 : 30	170, 185, 200	160, 180, 200	5, 7, 10
			80 : 20	170, 185, 200	160, 180, 200	5, 7, 10
Туре З	3	Thermoplastic bi- component fiber	40 : 60	130, 140, 150	90, 110, 130	2, 5, 7
			50 : 50	130, 140, 150	90, 110, 130	2, 5, 7
			60 : 40	130, 140, 150	90, 110, 130	2, 5, 7
			70 : 30	130, 140, 150	90, 110, 130	2, 5, 7

Table I. Composites and Their Processing Parameters

fabric was simply compression molded above the melting temperature of the polyester fabric, either as received or with addition of plasticizer. This route has already been investigated for cotton/PET (with 65% PET) by Zou et al.<sup>2</sup> In the second method, a bio-based resin from soybean oil was used as matrix. The third method used a thermoplastic core sheath type bicomponent fiber carded into a nonwoven fabric. This was placed between layers of the discarded cotton/PET fabrics, where it acted as a matrix. The core component in the bicomponent fiber does not melt at the compression molding temperature, while the sheath component melts and forms the matrix. These composite preparation methods were studied in order to find a feasible way to produce composites from textile waste. The first method is a rather straightforward method to recycle textile waste, and it has also been demonstrated. A draw back with compression molding is the thermal degradation of the cotton fibers, and therefore we also studied the two other concepts. In the second method we use a liquid thermoset resin as the matrix, and in the third method we used a bicomponent fiber as the matrix. Both methods enable compression molding at a temperature at which cotton does not degrade thermally. This should give composites with improved mechanical properties.<sup>2</sup> Laminates were manufactured according to three different concepts, and then they were evaluated regarding mechanical performance, as quantified by tensile and impact tests. Dynamic mechanical thermal analysis (DMTA), differential scanning calorimetry (DSC), and thermogravimetric analysis (TGA) were used to describe the process-related effects of the composites. Water absorption tests were performed to determine the amount of water uptake by the specimens. Morphological analysis was done by scanning electron microscopy (SEM) images.

#### **EXPERIMENTAL**

#### Materials

Discarded cotton/PET (50 : 50) plain-weave bed linen fabrics were obtained from Textilia, Sweden. The 210 g/m<sup>2</sup> fabrics had

been used in hospitals, care centers, or other institutions. Plasticizers, glycerol (ACS reagent grade,  $\geq$  99.5%), and 2-phenylphenol (reagent grade, 99%), were supplied by Sigma-Aldrich Sweden AB. Ethanol (reagent grade, 99.8%) was supplied by Fisher Scientific. Free radical initiator, tertbutylperoxybenzoate, for crosslinking of the thermoset was supplied by Aldrich Chemical Company, WY. Bi-component fibers (CoPET/PET, core sheath construction) were obtained from Fiberpartner ApS, Vejle, Denmark. The length of the bi-component fiber was 51 mm, denier 4 and tenacity 3.4 g/d. Copolyester sheath melts at 110–120°C. Methacrylated soybean oil (MSO) was synthesized according to the method described by Adekunle et al.<sup>12</sup>

# **Composite Preparation**

**Carding and Needle Punching of Bi-Component Fibers.** Bicomponent fibers were carded using a machine supplied by Cormatex, Italy. The frequency of the trolley during carding was 40 Hz, which corresponded to 7.5 m/min at the outlet. The carded web was needle-punched to make a uniform nonwoven mat using a needling machine supplied by Certec, France. The frequency during needle punching was 200 cp/min and the feed rate was 1.5 m/mm.

Laminate Preparation. Fabrics were cut to 20 cm  $\times$  20 cm swatches. The swatches were dried in an oven at 105°C for 2 h. The composites were produced by compression molding of fabric pieces with or without plasticizers, and with or without matrix, between two hotplates of a compression molding machine supplied by Rondol hot press, UK. Matrix and processing parameters are specified in Table I. The compression factors for all three types of composite production cannot be generalized. The thickness of the composite laminates varied from 0.5 mm to 1.4 mm depending on type of composite and processing parameters. DSC melting endotherm curves were used to decide the compression temperature.



*Type 1—Compression molding of neat fabrics (C1).* The concept 1 composite molding parameters followed those of Zou et al.<sup>2</sup> Besides compression of the neat fabrics, the fabrics were also compressed after spraying with plasticizers; glycerol or 2-phenylphenol (5 wt % of fabric). The composition and molding parameters are given in Table I.

Type 2—Compression molding of soybean based thermoset resin reinforced with blended fabric (C2). Thermosetting resin made from soybean oil, MSO, was used as matrix. Initiator, 2 wt % tert-butyl peroxybenzoate, was mixed with the MSO and used to impregnate the fabric manually. The composition and molding parameters are given in Table I.

*Type 3—Compression molding of bi-component reinforced with blended fabric (C3).* Fabrics were molded into composites with bi-component fibers (CoPET/PET), where bi-component fiber sheaths were melted to form the matrix. Carded and needled bi-component fibers were placed in-between the layers of blended fabric and subjected to compression molding to form composites. The composition and molding parameters are given in Table I.

# CHARACTERIZATION

Mechanical, thermal, and viscoelastic analyses were done to determine the performance of the composites. Specimens were cut according to the usual standards from the laminates using laser cutting technique (GCC, Laserpro Spirit, the Netherlands). Specimens were stored under ambient conditions, and conditioned for 24 h at 50% relative humidity and 23°C before testing according to DIN EN ISO. Type 3 composites were tested in machine direction and in transverse direction, as the inner core of the bicomponent gives extra strength in the machine direction.

## Mechanical Testing

Tensile testing was carried out using a Tinius Olsen H10KT tensile tester according to ISO 527 standard. An extensometer was used to measure the strain. The gauge length was 50 mm and the test speed was 10 mm/min. Load cell capacity was 5 kN. Ten dumbbell-shaped specimens were analyzed for each sample. Tensile strength, Young's modulus, and strain at break were noted.

Charpy impacts tests were carried out using a Zwick impact tester according to ISO 179 standard. At least 10 specimens were tested per sample to see the distribution. The test was carried out flatwise direction for unnotched specimens.

# Water Absorption Test

Water absorption tests were conducted to indirectly quantify the porosity of the composites. After drying in a desiccator at  $60^{\circ}$ C for 24 h, four specimens per sample were immersed in 100 mL of RT tap water in a 500-mL beaker. Masses of the  $36 \times 10$  mm-sized specimens were determined daily for ten days. For each reading, the specimens were padded with paper tissue to avoid surface water. After weighing, they were returned to the water.

# Thermal Analysis

Differential scanning calorimetry was carried out using a DSC Q2000 from TA Instruments, New Castle, DE. The temperature range was  $-30^{\circ}$ C to  $300^{\circ}$ C with a heating rate of  $10^{\circ}$ C/min. There were two heating ramps and one cooling ramp. The

experiment was done under nitrogen atmosphere. Three specimens were tested for each type comprising 1 : 1 fiber-matrix ratio (Type 1 where fabric had a cotton-PET ratio f 1 : 1; Type 2 where 50% was fabric and the rest was MSO resin; Type 3 where 50% was fabric and the rest was bi-component matrix). The sample size was ~10 mg. Glass transition temperature ( $T_g$ ) and peak melting temperature ( $T_m$ ) were noted.

Thermogravimetric analysis was performed using TA instruments, TGA Q 500, supplied by Waters LLC, USA. Approximately 15 mg of the samples were heated from  $30^{\circ}$ C to  $600^{\circ}$ C at  $10^{\circ}$ C/min in a nitrogen purge stream. The flow of nitrogen was 50 mL/min. As for DSC (see above), three specimens were tested for each composite type with a fiber-matrix ratio of 1 : 1.

# **Viscoelastic Analysis**

Dynamic mechanical thermal analysis was carried out using a DMA Q800 from TA Instruments (New Castle, DE). A dualcantilever clamp was used to perform the tests. The frequency was 1 Hz and the amplitude was 15  $\mu$ m. The temperature range was from 30°C to 150°C at a heating rate of 3°C/min. The dimensions of the specimens were 35  $\times$  10  $\times$  1 mm<sup>3</sup>. Three specimens were tested for each sample. Storage modulus and loss modulus were noted, to assess the elastic response and viscous response of the specimen.

# Morphological Analysis

Morphological analysis was done by studying the cross sections of the tensile-fractured composite specimens by scanning electron microscope, using a JEOL JSM SEM at 10 kV accelerated voltage. The specimens were selected as described above for DSC.

# RESULTS

# Mechanical Testing

**Type 1 Composites.** *Tensile test.* Tensile properties were analyzed for the specimens made by melting the polyester of the fabric at different compression temperatures and times. Tensile properties were also analyzed for specimens with plasticizers, glycerol (G), and 2-phenylphenol (2P).

Figure 1(a) shows the tensile strengths of the specimens. G and 2P indicate the specimens with glycerol and 2-phenylphenol plasticizers, respectively. Change in processing temperature and time have an influence on tensile strength. Specimens without plasticizers had tensile strength of 18 MPa when the compression temperature and time were  $270^{\circ}$ C and 20 s, respectively. On increase of the compression temperature to  $280^{\circ}$ C, the tensile strength increased to 23 MPa. Increase in tensile strength was also seen for the specimens made at  $270^{\circ}$ C for 30 s. With further increase of the compression temperature to  $290^{\circ}$ C, the tensile strength decreased. Longer compression time reduced the tensile strength; it was reduced to 16 MPa when the compression time was increased to 40 s at  $270^{\circ}$ C.

When glycerol was used as plasticizer to produce specimens at  $270^{\circ}$ C for 20 s, the tensile strength of 8 MPa was considerably lower than that of the samples without the plasticizer. On increasing the temperature and time to  $280^{\circ}$ C and 30 s, tensile





**Figure 1.** (a) Tensile strength of the Type 1 composites with and without plasticizers at different processing temperature and time (G, glycerol; 2P, 2-phenylphenol). (b) Tensile modulus of the Type 1 composites with and without plasticizers at different processing temperature and time (G, glycerol; 2P, phenylphenol). (c) Percentage elongation of the type 1 composites with and without plasticizers at different processing temperature and time (G, glycerol; 2P, erol; 2P, 2-phenylphenol).

strength was increased. The tensile strength was reduced when compression temperature and time were increased to 290°C and 40 s. These composites followed a trend that was similar to that of the specimens without plasticizers.

The highest tensile strength was obtained when specimens were processed with 2-phenylphenol plasticizers at  $270^{\circ}$ C for 20 s. The tensile strength was decreased when the compression temperature was raised to  $280^{\circ}$ C and  $290^{\circ}$ C. A similar trend was seen when compression time was increased to 30 and 40 s. Statistical ANOVA showed that the tensile strengths of the composites with 2P plasticizer were not significantly different from each other, except the composites made at  $270^{\circ}$ C and 20 s.

The tensile modulus of the specimens is shown in Figure 1(b). Similar trends of increase and decrease in the property by increasing compression time and temperature were noted. But the moduli of specimens with plasticizers were lower than the specimens without plasticizers. The highest modulus of 3 GPa was obtained when specimens were prepared at 280°C for 30 s without plasticizers. Statistical analysis showed that the tensile moduli of composites without plasticizer were significantly different from the composites with plasticizers (G and 2P), but the

tensile moduli of the composites with G and 2P were not significantly different.

Figure 1(c) shows the percentage elongation of composites. The highest elongation was obtained from specimens with 2-phenylphenol plasticizer. Specimens with plasticizers showed good elongation that was comparable with that of specimens without plasticizers. Statistical analysis showed that there was no significant difference in elongation between the composites with and without plasticizers.

The high processing temperature can cause thermal degradation of the cotton fiber, which will affect its mechanical properties and also the interfacial adhesion between the cotton fiber and the polyester matrix. The obtained results could be interpreted as a result of thermal degradation of the cotton fiber.

*Impact test.* Impact strength was studied for the composites without plasticizer as shown in Figure 2(a). The impact strength was dependent on temperature and time of compression of the composite laminate. Impact strength of 25 kJ/m<sup>2</sup> was obtained for the specimen processed at  $270^{\circ}$ C for 20 s. The strength increased to 27 kJ/m<sup>2</sup> on increasing the compression time, but





Figure 2. (a) Impact strength of the Type 1 composites without plasticizer at different processing temperature and time. (b) Impact strength of the Type 1 composites with glycerol plasticizer at different processing temperature and time. (c) Impact strength of the Type 1 composites with 2-phenylphenol plasticizer at different processing temperature and time.

on further increase of compression time, the impact strength fell to 22 kJ/m<sup>2</sup>. This trend was apparent when the temperature increased to  $280^{\circ}$ C and  $290^{\circ}$ C. There was a slight increase in impact strength in most cases when the temperature was raised to  $280^{\circ}$ C, but the impact strength fell when the temperature was raised further to  $290^{\circ}$ C.

The impact strength of composites with plasticizers is shown in Figure 2(b,c). This follows a trend that is similar to that of composites without plasticizers. The impact strength of composites with plasticizers was slightly better than that of composites without plasticizers. Composites with glycerol showed better results than those with 2-phenylphenol. Composites processed at 270°C for 20 s had an impact strength of 28 kJ/m<sup>2</sup> with glycerol and 26 kJ/m<sup>2</sup> with 2-phenylphenol. The decreased impact strength for the composites processed at 290°C can indicate cotton degradation.<sup>2</sup> Change in processing time did not influence the results significantly, except for the Type 1 composite when going from 270 to 290°C.

**Type 2 Composites.** *Tensile test.* The tensile properties of the composites made from waste textile reinforcement and biobased matrix were analyzed. Several factors such as fiber-matrix ratio, compression temperature, pressure, and time were taken into consideration for optimization of tensile properties.

Variation of tensile strength with fiber-matrix ratio is shown in Figure 3(a). The tensile strength of composites increased on changing the fiber-matrix ratio from 50 to 80 wt % and keeping other factors constant. Tensile strength was 52 MPa when the composite consisted of 50 wt % fiber. When the fiber wt % increased to 60 and 70 wt %, the tensile strength increased to 54 and 58 MPa, respectively. With further increase in wt % of fiber to 80, the tensile strength dropped significantly to 42 MPa.

We studied tensile strength in relation to processing temperature, time, and pressure. The tensile strength increased on increasing the temperature from  $170^{\circ}$ C to  $185^{\circ}$ C and  $200^{\circ}$ C. Composite had a tensile strength of 45 MPa when the compression temperature was  $170^{\circ}$ C, and it increased to 52 MPa at higher temperatures. A similar trend was seen when processing time was increased. The tensile strength increased from 45 MPa to 50 MPa and 51 MPa when the processing time increased from 5 min to 7 min and 10 min, respectively. The tensile strength increased when the pressure was increased from 160 kPa to 180 kPa, but there was a slight decrease in tensile strength when the pressure was raised further to 200 kPa.

Tensile modulus followed a trend similar to that of tensile strength in all cases. Tensile modulus increased on increasing





Figure 3. (a) Tensile strength of Type 2 composites w.r.t fiber-matrix ratio at constant processing parameters (time, temperature, and pressure). (b) Tensile modulus of Type 2 composites w.r.t fiber-matrix ratio at constant processing parameters (time, temperature, and pressure). (c) Elongation at break of Type 2 composites w.r.t different temperature, time, and pressure.

the fiber wt % from 50 to 60 and 70, but it decreased significantly on increasing the fiber to 80 wt % [Figure 3(b)]. We also analyzed changes in tensile modulus relative to temperature, time, and pressure, and the results followed the trend seen with tensile strength.



Figure 4. Impact strength of Type 2 composites with different weight % reinforcement, compression pressure, compression temperature, and compression time.

The elongation of the composites decreased on increasing the processing temperature and time [Figure 3(c)]. Specimens had 1.7% elongation when the laminate was processed at 170°C. The elongation fell to 0.8% and 0.7% when the processing temperature increased to 185°C and 200°C, respectively. A similar trend observed when compression time was increased. Elongation decreased when the compression pressure increased from 160 kPa to 180 kPa, but it increased when the pressure was raised to 200 kPa. The percentage elongation of composites prepared at 170°C was found to be significantly different from that of composites prepared at 185°C and 200°C, but the latter two temperatures gave results that were not significantly different. Similar statistical results were obtained when we analyzed three different processing times. The decrease in tensile elongation indicates the formation of a more rigid and brittle crosslinked network, which can be the result of a higher degree of crosslinking when the processing temperature and time is increased.

*Impact test.* The impact strength of the composites is shown in Figure 4. Impact strength increased on increasing the fiber loading in the composite from 50 wt % to 60 wt % and 70 wt %, but it decreased when the amount of fiber in the composite was increased to 80 wt %. This indicates poor stress transfer in the



Figure 5. (a) Interaction plot for tensile strength of Type 3 composites with different fiber-matrix ratio, compression temperature, compression time, and compression pressure. (b) Interaction plot for tensile modulus of Type 3 composites with different fiber-matrix ratio, compression temperature, compression time, and compression pressure. (c) Interaction plot for % elongation with different fiber-matrix ratio, compression temperature, compression time, and compression pressure. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

composite with high fiber loading which can be due to poor fiber-matrix impregnation. Toughness increased when the processing pressure was increased from 160 to 180 and 200 kPa. A similar trend was seen when the processing temperature was increased from 170°C to 185°C and 200°C. The impact strength of the composite was 64 kJ/m<sup>2</sup> when it was processed for 5 min, but it increased to 70 kJ/m<sup>2</sup> and 71 kJ/m<sup>2</sup> when processing times were 7 and 10 min. These results can be explained by the



Figure 5. (Continued).

better fiber-matrix impregnation at higher processing temperature and pressure, and longer processing time. Even though there were slight changes in all the results with change in processing parameters, except for the composites with 80 wt % fiber, all the other impact strengths were statistically insignificant.

Type 3 Composites. Tensile test-tested in machine direction (longitudinal). The tensile strength in longitudinal direction of the composites is shown in Figure 5(a). An interaction plot was drawn, considering several factors such as fiber loading, temperature, pressure, and time of molding. Bi-component was melted to form laminates; it had a tensile strength of 58 MPa. When fabric was added as reinforcement, the tensile strength improved. A tensile strength of 88 MPa was achieved on introducing the fabric as reinforcement. This was expected, as reinforcement gives extra strength to composites. Tensile strength was also dependent on temperature, pressure, and time of compression. When the temperature was raised from 130 to 150°C and other factors kept constant, the tensile strength increased from 62 to 73 MPa when composite had 40% fabric. The tensile strength of composite consisting of 60 wt% reinforcement was 54 MPa when 90 kPa of pressure was applied during compression. The tensile strength rose to 64 MPa when the pressure was increased to 110 kPa, and it rose to 94 MPa when the pressure was increased to 130 kPa, with all other factors kept constant. The change in compression time of the composite laminates affected the tensile strength. Tensile strength of 61 MPa increased to 65 MPa when the time of compression was changed from 2 to 5 min and keeping other factors constant for composites with 40% reinforcement.

In general, the tensile strength increases when the proportion of reinforcement is increased. But changing the compression factors could change the trend that is seen in the interaction plot. All four factors have an influence on tensile strength, and each factor affects the results of the other three factors.

An interaction plot of E-modulus is shown in Figure 5(b). The E-modulus of neat bi-component laminate was high, and was around 5981 MPa. Introduction of fabric reduced the modulus of the composites. Keeping the compression factors constant and changing the wt % of reinforcement affected the modulus; when 40 wt % reinforcement was used, the modulus decreased to 3557 MPa. It was reduced further to 2854 MPa when composites had 60 wt % reinforcement. The modulus increased when compression time was changed and other factors kept constant: it was 2584 MPa when compression time was 2 min and it increased to 3414 MPa and 4580 MPa when the time was changed to 5 and 7 min, respectively. The effect of pressure was seen when all the factors were keep constant except pressure: the tensile modulus increased from 3707 MPa to 5170 MPa when the pressure of compression was increased from 110 kPa to 130 kPa.

Similarly, compression temperature also affects the tensile modulus. A modulus of 3557 MPa was obtained when the temperature was  $130^{\circ}$ C, but it increased by increasing the temperature and keeping other factors constant. It rose to 4336 MPa and it was increased further to 5814 MPa when the compression temperature was  $140^{\circ}$ C and  $150^{\circ}$ C, respectively.

The E-modulus of neat bi-component laminate was high, while the waste fabric used as reinforcement had lower modulus. This



could be due to orientation of bi-component in one direction. Thus, the introduction of reinforcement decreased the modulus of the composites. The results were affected by the compression factors such as time, pressure, and temperature.

The percentage elongation of the composites was between 4% and 16% and the interaction plot is shown in Figure 5(c). The interaction plot describes the effect of each factor on percentage elongation and the effect of each factor on the others. Bicomponent laminate had a percentage elongation of 12.6%. This was reduced to 11% and 7% on inclusion of reinforcement of 40 and 60 wt %. The percentage elongation decreased on increasing the compression temperature. It fell from 12.7% to 11.8% and further to 10.1% when the temperature was 130, 140, and 150°C. The effect of compression pressure was noticed when there was a change in pressure and all the other factors were kept constant. The percentage elongation increased from 4.3% to 7.5% and further to 11% when the pressure was increased from 90 kPa to 110 kPa, and further to 130 kPa, respectively.

The interaction plots show the importance of considering the four factors together. The effect of unmelted bi-component core was seen, as it increases the mechanical properties of composites. It was also important to study the properties of the composites in transverse direction.

Tensile testing—tested in a direction perpendicular to the fibers (transverse). Good tensile properties were obtained on addition of bi-component fibers, but these properties were obtained along the machine direction. So, it is necessary to look at the properties along the transverse direction. Tests were conducted perpendicular to the bi-component alignment. In general, the tensile properties became less, as expected, as the core of the bicomponent (which is not melted) improved the properties along the machine direction. Similar trends in tensile properties were seen in both directions.

The tensile strength of the melted bi-component laminate was around 38 MPa. It increased on inclusion of reinforcement while other factors were kept constant [Figure 6(a)]. Tensile strength was 43, 51, and 64 MPa when composite consisted of reinforcement of 40, 50, and 60 wt %. The tensile strength changed from 43, 45, and 51 MPa when the compression temperature increased from  $130^{\circ}$ C to  $140^{\circ}$ C, and then to  $150^{\circ}$ C. Similarly, the effects of change in pressure and time were noted in the interaction plot.

Figure 6(b) shows the E-modulus of the composites. E-modulus was lower in the transverse direction than in the machine direction. A more even trend was seen when there was change in percentage of reinforcement in the composites. The E-modulus of the bi-component laminate was around 4763 MPa and it decreased on inclusion on fabric, as the modulus of bi-component was higher than that of the waste fabric. This could be due to intact bonding in neat bi-component laminates, whereas introduction of fabric reduces the bonding. Reduction of the amount of bi-component in the composites reduced their E-modulus of the composites. The tensile modulus was 3936, 3094, and 2329.5 MPa when bi-component was replaced with 40, 50, and 60 wt % reinforcement, respectively.

The effect of change in temperature is shown in the interaction plot, with other factors kept constant. Modulus was 3094, 3230, and 3250 MPa when the compression temperature was 130, 140, and 150°C. There was change in modulus when compression time was changed but other factors were kept constant. E-modulus was 2329, 3561, and 3711 MPa when the compression time was 2, 5, and 7 min, respectively.

Figure 6(c) shows the percentage elongation of the composites, which ranged between 4% and 14%. The elongation of bicomponent was reduced from 12.6% to 1.1% by changing to transverse testing direction. The effect of change in compression factors is shown in the interaction plot. Similarly, there was change in elongation when there was change in the amount of reinforcement in the composites.

The importance of the orientation of the bi-component fibers was noted by testing the laminates for mechanical properties in the longitudinal and transverse directions. The effect of processing parameters on tensile properties was evident in all cases. Every processing parameter also affects the results with the other parameters. The degradation of cotton was negligible and this was due to lower compression temperatures.]

Impact test. Figure 7(a) shows that the toughness improved with increase in proportion of fabric. This observation held both for samples cut along and for samples across the fiber direction of the uni-directional Type 3 composites. Note that the bi-component fiber core does not melt during composite preparation, and thereby maintains a uni-directional orientation, which makes the composite highly anisotropic. Impact strength was measured against change in fiber-matrix ratio while keeping other processing parameters constant. In all cases, impact strength was higher when the laminates were tested in longitudinal direction than in transverse direction. In both directions, the impact strength increased on increasing the amount of reinforcement in the composites. The impact strength of pure bi-component laminate in longitudinal direction was 37 kJ/m<sup>2</sup>, and it was increased to 49, 53, 54, and 59 kJ/m<sup>2</sup> by replacing bi-component of 40, 50, 60, and 70 wt % with the same amount of waste fabric reinforcement, respectively. Similarly, impact strength in transverse direction was increased from 24 kJ/m<sup>2</sup> to 29 kJ/m<sup>2</sup> by replacing 40 wt % bicomponent with 40 wt % reinforcement. It was further increased to 32, 35, and 38 kJ/m<sup>2</sup> by replacing bi-component of 50, 60, and 70 wt %.

Impact strength was slightly increased on increasing the processing temperature and keeping other factors constant [Figure 7(b)]. Impact strength was 49, 54, and 55 kJ/m<sup>2</sup> when the processing temperature was 130, 140, and 150°C. There was a small effect on impact strength when the processing pressure or time was changed.

These results were expected, as impact strength increased on increasing the fiber content, the increasing processing temperature, the processing time, and the processing pressure. Addition of reinforcement improved the strength of the composites. Several factors such as melting of bi-component sheath, flow of matrix, and adhesion of matrix, etc. decide the impact strength





Figure 6. (a) Interaction plot for tensile strength of Type 3 composite where samples were harvested across the uni-directional bi-component fiber axis w.r.t different fiber-matrix ratio, compression temperature, compression time, and compression pressure. (b) Interaction plot for tensile modulus of Type 3 composite where samples were harvested across the uni-directional bi-component fiber axis w.r.t different fiber-matrix ratio, compression temperature, compression time, and compression temperature, compression time, and compression pressure. (c) Interaction plot for % elongation of Type 3 composite where samples were harvested across the uni-directional bi-component fiber axis w.r.t different fiber-matrix ratio, compression temperature, compression time, and compression pressure. (c) Interaction plot for % elongation of Type 3 composite where samples were harvested across the uni-directional bi-component fiber axis w.r.t different fiber-matrix ratio, compression temperature, compression time, and compression pressure. (c) Interaction plot for % elongation of Type 3 composite where samples were harvested across the uni-directional bi-component fiber axis w.r.t different fiber-matrix ratio, compression temperature, compression time, and compression pressure. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



Figure 6. (Continued).

of the composites, and the above-mentioned factors could be responsible for the variation in the impact properties. The processing parameters affect impact strength, and these results could also be affected by other parameters. The importance of the orientation of bi-component fibers was seen in all cases, as the impact strength decreased severely on changing the direction of testing from longitudinal to transverse.

# Water Absorption

**Type 1 Composites.** Water absorption of plasticized Type 1 composites is shown in Figure 8(a). Water absorption was reduced when plasticizers were used. 2-phenylphenol composites absorbed less water than glycerol composites. Composites absorbed 19 wt % water without plasticizers; with glycerol and 2-phenylphenol, the water absorption was reduced to around 16 and 14 wt %, respectively.

Figure 8(b) shows water absorption with different molding temperatures. Water absorption was reduced from 19 to 15 wt % when the temperature was raised from 270 to 280°C, but the water absorption increased when the processing temperature was 290°C. The pores created on degradation of reinforcement may be the reason for increased water absorption.

The effect of processing time on water absorption is shown in Figure 8(c). At lower temperature (270°C), higher processing time gives less water absorption. Water absorption was reduced from 19 to 15 wt % when the processing time was increased from 20 s to 40 s.

# Type 2 Composites

Figure 9(a) shows water absorption according to the fibermatrix ratio. Composites absorbed more water when the fiber content was increased. Water absorption was more evident over the first 3 days, and it reached a point close to saturation. Composite with 50 wt % fiber absorbed around 19 wt % water at the end of 10 days. When the fiber content in the composite was increased to 60, 70, and 80 wt %, the water absorption was around 26, 34, 39 wt %, respectively.

As with Type 1, the water absorption was affected by processing temperature and time. Water absorption became less on increasing the processing temperature: 19, 15, 12 wt % when the processing temperature was 170, 185, and 200°C, respectively.

Water absorption decreased when the compression time was increased. When the compression time was 5, 7, and 10 min, the corresponding water absorption was around 19, 16, and 14 wt %. Proper spreading of matrix, good fiber–matrix adhesion, and reduced pore volume might be the reasons for reduced water absorption at higher temperatures.

**Type 3 Composites.** Figure 9(b) shows the water absorption of the composites on change of fiber–matrix ratio. The water absorption increased on increasing the wt % of reinforcement in the composites. The water absorption was more evident on the first day and increased slightly from Day 2. Bi-component laminate absorbed only around 1 wt % water at the end of tenth day. On inclusion of 40 wt % reinforcement, the water absorption rose to 17 wt %. It rose further to 26 wt % when the composite had 50 wt % reinforcement. It increased even more to 35 and 39 wt % when the composites had 60 and 70 wt % reinforcement. The high water absorption was due to pores present in the composites. Porosity tests and calculations confirmed the presence of pores in the composites.





**Figure 7.** (a) Impact strength of the Type 3 composites tested along the longitudinal and transverse directions with different weight % reinforcement. (b) Impact strength of Type 3 composites tested along the longitudinal and transverse directions with different compression temperatures.

The water absorption of the laminates was affected when the processing temperature changed but when other factors were kept constant. Water absorption decreased on increasing the temperature, and this could be due to proper melting of bicomponent. The water absorption was 23, 19, and 12 wt % when the compression temperature was 130, 140, and 150°C.

Water absorption changed on changing the processing time while keeping other factors constant. When we increased the compression time from 2 to 5 min, the water absorption was reduced to 27 wt % from 34 wt % after 10 days. When the time was further increased to 7 min, there was no significant change.

Similarly, the effect of processing pressure was noted when other parameters were kept constant during manufacture of composites. Water absorption was more on the first day, and there was slight increase from Day 2. Water absorption increased on increasing the processing pressure from 90 kN to 130 kN.

#### Thermal Analysis

**Differential Scanning Calorimetry.** DSC curves showed that the melting temperature was reduced by the addition of plasticizers (Figure 10). Melting temperatures were reduced from 260 to  $255^{\circ}$ C on addition of plasticizer glycerol and from 260 to  $254^{\circ}$ C on addition of plasticizer 2-phenylphenol. The effect of



Figure 8. (a) Water absorption of Type 1 composites with and without plasticizers. (b) Water absorption of Type 1 composites at different processing temperatures. (c) Water absorption of Type 1 composites with different processing time.

2-phenylphenol was better than that of glycerol: 2-phenylphenol reduced the melting temperature more than glycerol.

**Thermogravimetric** Analysis. Thermogravimetric analysis shows the percentage weight loss of the composite specimen when the specimen is heated at uniform rate in a controlled atmosphere. The processing temperature of Type 2 composite was well below the melting temperature of polyester in the





**Figure 9.** (a) Water absorption of Type 2 composites with different fibermatrix ratio. (b) Water absorption of Type 3 composites with different fiber-matrix ratio.

reinforcement fabric, and the degradation of cotton was negligible. Thus, the onset of degradation at elevated temperature in TGA is delayed.

The effect of plasticizers was interpreted as delayed the onset of decomposition. This could be due to lower processing temperature as the plasticizer reduced the melting temperature. The effect of plasticizers was evident at lower temperatures, as it delayed the start.

TGA curves of all three types of composites are shown in Figure 11. The decomposition of type 1 composites starts early, while the Type 3 is delayed.

# Viscoelastic Analysis

**Dynamic Mechanical Thermal Analysis.** Storage modulus, loss modulus, and tan  $\delta$  of the composites were analyzed, and the results are given in Table II. Types 1 and 3 composites had better storage modulus than Type 2; this could be due to lower porosity and better fiber-matrix adhesion in Types 1 and 3 composites. Bi-component-based composites had the highest values of storage modulus. Similar trend was seen in loss modulus, as bi-component-based composites had the highest loss modulus.



Figure 10. Differential scanning calorimetry curves of Type 1 composites with and without plasticizers.

The highest point in the tan  $\delta$  curve was reduced from 87.5°C to 80.2°C by glycerol and from 87.5°C to 79.3°C by 2-phenylphenol.

#### Morphological Analysis

Figure 12 shows microscopic images from the scanning electron microscopy (SEM) of all three types of composites. Micrographs a and b show the SEM images of Type 1 specimens. The polyester of the fabric melts and acts as the matrix, which can be seen in Figure 12(a), while the imprints of the fibers can be seen in Figure 12(b). The images confirmed the melting of polyester which binds firmly to cotton fibers before tensile testing, and the fiber pull-out was evident on tensile testing. This confirms the low tensile strength of Type 1 composites.

Figure 12(c,d) are SEM images of Type 2 specimens. The fabric was reinforced with MSO resin at fiber-matrix ratio of 1 : 1. Figure 12(c) shows the good interaction between fiber and matrix, which restricts the fiber pull-out. Figure 12(d) shows the good penetration and crosslinking of MSO matrix in the fabric. The above results confirm the good tensile strength of these composites.

Figure 12(e,f) are SEM images of Type 3 specimens. The bicomponent fiber sheath acts as a matrix while fabric acts as reinforcement. Figure 12(e) shows the good melt of bi-



**Figure 11.** Thermogravimetric curves for three types of composites with a fiber–matrix ratio of 1 : 1.



 Table II. Dynamic Mechanical Thermal Analysis Results for Type 1 Composites with and Without Plasticizers

Composite	Highest point in tan $\delta$ curve (°C)	Storage modulus at 30°C (MPa)	Loss modulus at 30°C (MPa)
No plasticizer	87.5	2209	33.3
Glycerol	80.2	2094	31.8
2-phenylphenol	79.3	2132	32.4

component sheath which binds with both fiber and bicomponent core. Figure 12(f) shows that fiber break and the fiber pull-out was negligible. These results match the tensile properties of the specimens.

# DISCUSSION

Several natural fiber composites have already been developed from different virgin fibers such as jute, hemp, flax, sisal, etc.<sup>13–</sup><sup>16</sup> In these composites, the fibers are combined with petroleumbased matrix (PP, PE, etc.) or renewable resource-based (soybean-based, linseed-based resin, etc.) These composites have superior properties, and can be used in many applications



Figure 12. Scanning electron microscopic images (Panels a and b, Type 1; Panels c and d, Type 2; and Panels e and f, Type 3).

including automobiles and construction. Type 1 composites may have properties that are inferior to those of these composites. They cannot be used in technical applications, but they could be used in less stress structures. Types 2 and 3 composites have given results that are comparable with those of other natural fiber composites, so they can be used in more sophisticated structures.<sup>17</sup>

In a previous research, Zou et al. prepared composites with properties similar to those of Type 1 where the fabric was pressed at temperatures higher than the melting point of polyester.<sup>2</sup> Composites were also made with inclusion of glycerol and 2-phenylphenol plasticizers in the fabric.<sup>2</sup> The highest tensile strength was obtained when the composites were compressed at 280°C for 35 s without plasticizers. The effect of plasticizers on the melting point of PET was shown in results from DSC curves; they lower the melting point. The effects of these plasticizers on composites were demonstrated in the tensile, flexural and impact properties; composites with 2-phenylphenol gave better results than the ones with glycerol–with the exception of impact strength.

Type 1 composites showed a similar trend to that in the results by Zou et al. regarding tensile strength of the composites with or without plasticizers: a decrease in tensile strength on addition of glycerol and an increase in tensile strength from 2phenylphenol plasticizer.<sup>2</sup> But it was possible to achieve more than two and four times better tensile strength with Types 2 and 3 composites, respectively, which is needed for structural applications. This was expected as the processing temperature in Type 1 composite is higher, which can cause degradation of the cotton fibers. The processing temperatures used for Types 2 and 3 were lower, thus giving little or no degradation of fibers. Soybean-based resin and bi-component fiber properties play a crucial role in Types 2 and 3 composites. In Type 3 composites, the sheath of the bi-component fiber melts while the core remains unmelted, which gives additional tensile strength to the composites. The fiber-matrix ratio influenced the properties, and these could be tailored by changing the ratio. Several other processing parameters such as temperature, pressure, and time affected the properties. Orientation effect was seen in Type 3 composites where the nonwoven bi-component fabric was used; the core that remains after melting the sheath gives additional tensile strength in one direction.

The influence of each processing parameter and the indirect effects of one parameter on the others were seen in all three tensile properties. High tensile strength was obtained at lower temperature when the plasticizer 2-phenylphenol was used in Type 1 composites, and this could be due to reduction in melting temperature. The tensile strength of the Type 2 composites increased on increasing the fiber content of the composites, until the fiber content reached to 80 wt %. The tensile strength dropped to 42 MPa when the fiber content of the composites increased to 80 wt % of the composites. Inadequate matrix to spread throughout the laminate, unevenness in matrix spreading, and poor fiber–matrix adhesion may be reasons for decrease in tensile strength. Several parameters such as degree of melting of bi-component sheath, impregnation of matrix in

the fiber, role of the unmelted bi-component core, heat distribution in the laminates, time, and pressure needed for even distribution of matrix etc. can influence the properties for Type 3 composites.

Degradation of cotton in the reinforcement fabric may be the reason for the trend of decreasing the tensile elongation on increasing the temperature and processing time in Type 1 composites, while good adhesion between fiber and matrix and even spreading of thermoset matrix could be the reason for reduction in tensile elongation in Type 2 composites. The cotton fibers could be expected to degrade at the higher compression temperatures. To minimize the degradation, compression time was kept to a minimum. On introducing the thermoset resin matrix in Type 2 and the bicomponent fiber in Type 3 composites, it was expected to give additional mechanical strength and also to reduce the compression temperature. By reducing the compression temperature, there is no or minimal degradation of cotton. In Type 3, the chosen copolyester sheath melts at 110-120°C; as a result, the degradation of cotton is minimized during composite production. A negligible degradation effect on processing would lead to delayed onset of degradation of Types 2 and 3 in TGA.

In Type 1 composites, longer exposure of cotton at higher temperature reduced the impact strength for the composites processed at 290°C for 40 s. In Type 2 composites, impact strength decreased when the amount of fiber in the composite was increased to 80 wt %. This is a similar observation to that with tensile strength, and the reason could be improper adhesion between fiber and matrix and uneven spreading of matrix.

In Type 1 composites, higher processing time gives less absorption of water due to proper impregnation through good melting of matrix; but at higher temperature (290°C), longer processing times might increase water absorption because of degradation of reinforcement. Type 2 composites absorbed more water when the fiber content increased; this is because the thermoset matrix used absorbs negligible amounts of water and reduction of matrix by replacing with reinforcement contributes to absorption of water. Even distribution of heat over the laminate, giving enough time for the matrix to melt well, and giving time for matrix to bind with fibers could be reasons for reduced water absorption when the processing time of the composites is increased. In Type 3 composites, water absorption increased on increasing the processing pressure. This may be due to restriction of spreading of matrix by pressure, which gives improper impregnation and wetting.

In Type 1 composites, increase in the compression time and temperature to 30 s and 280°C led to melting of polyester fibers and gave better adhesion between matrix and fiber, which was apparent from the improved mechanical properties. However, at higher compression temperature and longer compression time, the cotton will degrade, which reduces the mechanical properties. Thus, it is necessary to reduce the compression temperature and time by reducing the melting temperature.

Scanning electron microscopic images of the cross sections of the specimens confirmed the results from tensile strength. These images showed the spreading of matrix, the fiber-matrix interface, fiber imprints, and fiber pull-out.



# CONCLUSIONS

Three possible routes for use of discarded 50/50 cotton/polyester bed linen as raw material for heat compression molded composites were evaluated. The results can be related to the previous work of Zou et al.<sup>2</sup> where the separation of two fibers (cotton from PET) is omitted because cotton acts as a reinforcement and improves the strength of the composites. It was possible to obtain better mechanical properties than in Zou's work, with different types of composite processing by reusing the waste textile as reinforcement. The fabric (cotton along with PET) acted as reinforcement in Types 2 and 3 composites, which gave better mechanical strength to the composites than reported in Zou's work. Processing temperature was lower in Types 2 and 3 composites than in Zou's work, as external matrix was used, which reduced the degradation of cotton and thus gave better tensile strength. The highest tensile strength of 88 MPa was obtained with Type 3 composites, which are comparable with several natural fiber composites. Melting and recrystallization erases the orientation that PET fibers got during manufacture by melt spinning. Thus, woven fabric reinforcement potential still remains if an external matrix is added. The results obtained in this project will help to produce composites from waste textiles with good properties for use in wider applications.

Further investigation of textile-based composites with enhanced properties may lead to the use of these recycled textiles into the composite field. Recycled cotton/polyester fabrics show good promise for use in the composite field. The characteristics of end-of-life textile materials (fiber/yarn dimensions, content of dyes, cross-linked resins, and functional additives) should be studied in future investigations as these parameters may influence the performance of these recycled fabrics in composite applications.

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