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Processing Technique and Performance Evaluation of High-Modulus Organic/Inorganic Puncture-Resisting Composites

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Abstract
The development of low-cost organic/inorganic puncture-resisting composites is suggested to improve their puncture resistance and thermal insulation properties. In this paper, recycled high modulus Kevlar fiber and glass fabric were used together with polyester/low-melting polyester nonwovens. The result shows that the static puncture resistance improves proportionally with Kevlar fibers; but the dynamic puncture resistance remains the same at first and then rises up to 20 wt % for Kevlar fibers. The increasing low-melting polyester fibers on the surface result in an upward and then downward trend for both static and dynamic puncture resistances. The additional polyester/low-melting polyester nonwovens are beneficial for the improvement of puncture resistances. Moreover the thermal conductivity of all the composites is in range of 0.015-0.025 W/m·K. According to the results, Kevlar fibers are also shown to be advisable reinforcement to achieve better tensile and bursting strengths.

Key words: *composite, puncture resistance, thermal insulation, mechanical properties, recycle.*

Introduction

P-aramid fibres have superior properties including high modulus, high tenacity, cut resistance, impact resistance, thermal resistance and abrasion resistance. Recently they have been applied as friction materials, protective clothing, rubber reinforcements, and optical fibres, etc. It is estimated by Teijin Company (Japan) that the global demand forp-aramid fibres would increase to 130,000 tons in 2015 and up to 200,000 tons by 2020. Accordingly a large amount of p-aramid fabric selvedges, which can occur in processing textiles or at the end of the useful life of garments, needs to be disposed of. Because p-aramid fibre is a non-biodegradable material, the traditional approaches have concentrated on incineration, as mentioned in the study conducted by Buggy et al. [1]. Such a method would not only consume more energy during combustion but also release toxic gases such as CO, NO, NO_2 etc. Consequently a favorable method, such as recycling, is attractive to be adopted.

One method of using recycled aramid fibre is to regenerate pulps or yarns. Tsukamoto and Tsunoda chopped aramid fibres to form cotton-like short mats or as reinforcing materials for fibre-reinforced reins and fibre-reinforced elastomer [2]. Tsukamoto and Kosuge invented a recycling process method for spun yarn with recycled high-performance staple fibres [3]. Flambard et al. determined that recycled p-aramid fibres exhibited excellent cutting resistance, almost as high as pure Kevlar. Besides this double knitted fabric woven from regenerated p-aramid yarns made of recycled fibres had good fire properties but bad abrasion resistance [4]. Concerning the comprehensive properties of recycled p-aramid fibres, they may be properly used as the interlayer for puncture-resisting body armour, which would not be subjected to friction forces. Especially recycled aramid fibres are associated with low cost, as compared to pure Kevlar fibres.

Termonia moulded the different stages during the needle puncture of a single ply of plain weave Kevlar fabric, and indicated that the maximum puncture resistance depended on the contact pressure of the tip needle against fibre strands [5]. However, multiple layers of Kevlar fabrics had the ability to resist against the puncture forces, but greatly increased the production cost. Thus many studies have done about the cost reduction of puncture-resisting armour. Mayo Jr et al. impregnated thermoplastic (TP) films into woven aramid fabric and showed that TP-laminated fabrics improve the puncture resistance of fabrics [6]. Kim et al. comparatively studied the stab resistance of thermoplastic and epoxy reinforced fabrics [7]. The epoxy resin reinforced p-aramid fabrics showed both excellent quasi-static and dynamic stab resistances due to the higher toughness of epoxy resin. And rubber latex coated on the unidirectional polyethylene fabrics to improve puncture resistance was studied by Hassim et al. [8]. This study prepared more elastic and flexible but lower puncture resistance fabrics than TP-laminated and resin-reinforced fabrics.

In addition, Decker et al. investigated the stab resistance of shear thickening fluid (STF) - treated Kevlar fabrics [9]. The STF addition dramatically improves the puncture resistance both under high and low speed loading conditions by reducing the mobility of filaments and yarns in the impact zone. Furthermore Kalman et al. discussed the effect of hardness on the puncture property of fabrics intercalated with dry particle and STF. The particle intercalation significantly increases the puncture property, primarily through decreasing mobility in the yarn and fibre. Comparatively the STF inclusion improves the puncture resistance more dramatically [10]. Kang et al. developed an advanced stab proof material composed of STF and Kevlar fabrics.

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***Table 1.** Fineness, length and manufacturer of high-tenacity PA6, PET and low-melting PET.*

Fibre type	Fineness	Length, mm	Manufacturer
High-tenacity PA6	6 D	64	Taiwan Chemical Fibre Co. Ltd., Taiwan.
PET	2 D	51	Far Eastern New Century Corp., Taiwan.
Low-melting PET	4 D	51	Far Eastern New Century Corp., Taiwan.

***Table 2.** Fabric specifications of woven glass fabric.*

Structure	Fineness	Surface mass	Density	Thickness
plain	1 K	328 g/m^2	26 × 34/inch ()	0.31 mm

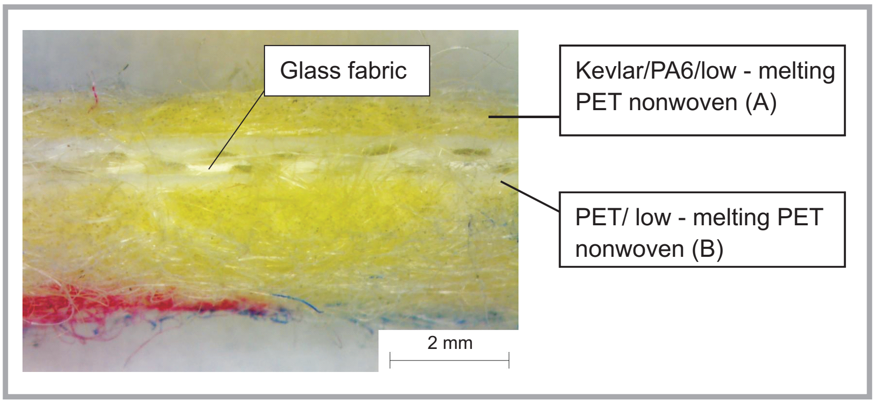

***Figure 1.** Cross section of 3 D laminated composites.*

The STF showed a reversible liquid-solid transition at a certain shear rate, thereby improving the stab resistance of Kevlar fabric [11].

Other than the puncture resistance property, thermal insulation and mechanical strength are considered to be necessary in the application of a body armour interlayer in a high-temperature environment. In this study, a puncture-resisting composite with thermal insulation and appropriate strength was prepared. Therein low-melting polyester fibres were applied to restrict the mobility of fibres and yarns after thermal melting. Moreover low-cost recycled Kevlar fibres were on the surface nonwovens. After insertion of a glass fabric, a three-dimensional structure composite was formed by needle-punching and thermal-bonding techniques to acquire an improved puncture-resisting composite. In the process, the proportions of low-melting and Kevlar fibres were changed, and their tensile strength, bursting strength and static and dynamic puncture resistances were evaluated. In order to satisfy the requirement of a high-temperature environment, their thermal insulation was also characterised.

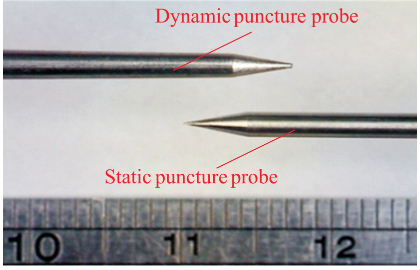

***Figure 2.** Probes for static and dynamic punctures.*

Experimental

Experimental materials

Kevlar fibres were derived from recycled K129 Kevlar unidirectional selvages (50 - 60 mm wide) provided by DuPont Corp., America. The physical parameters of high-tenacity polyamide 6 (PA6), polyester (PET) and low-melting PET are listed in ***Table 1***. Glass fabric (supplied by Jinsor-Tech Industrial Corp., Taiwan) was also used as reinforcement in our study, whose specification is displayed in ***Table 2***.

3D laminated composites preparation

A 3D laminated composite was made from double layers of Kevlar/PA6/low-melting PET nonwovens (A) and the same layer of PET/low-melting PET nonwovens (B), as well as with one layer of glass fabric after needle-punching and thermo-bonding techniques. The composite was needled at both sides with a density of 122 needles/cm^2 and then hot-pressed at 150 °C for 5 minutes under a pressure of 40 kg/cm^2 by a Flat Hot-presser (Relter, Taiwan). The thickness of the composite was set as 3 mm, whose structure is shown in ***Figure 1***. Both nonwovens were made in our laboratory via processes including opening, blending, carding and lapping and then needle-punching at 100 needles/cm^2. The blending ratio of Kevlar fibre, PA6 fibre and low-melting PET fibres in nonwoven A was changed in the processing. Nonwoven A had a weight per unit area of 200 g/m^2, , whereas nonwoven B consistently contained 30 wt% of PET fibres and 70 wt% of low-melting PET fibres, whose basis weight was constant at 150 g/m^2. For glass fabric, its weft yarn was arranged along the machine-direction.

Mechanical testing

The tensile strength of composites was tested according to ASTM D5035-11. An Instron 5566 universal testing machine (Instron, America), operated at a cross-head speed of 300 mm/min, was used to measure the rectangular specimens (180 mm length × 25.4 mm width). The strain-stress curves of the composites were recorded by the testing machine itself. Six specimens each along the machine-direction (MD) and cross-direction (CD) were tested for each parameter.

The bursting strength was also measured by the Instron 5566 Tester (Instron, US) based on ASTM D1883. A semicircular-ended head of 25 mm diameter, attached to a 10 kN load cell, was driven onto the composites at a speed of 100 mm/min. At the same parameters (the same low-melting PET and Kevlar staple fibres), tests of the six square specimens (150 mm length × 150 mm width) were completed for the average result.

Puncture resistance testing

The static and dynamic puncture resistances were both determined to simulate different punch behaviors. The static punch property was established at 508 mm/min using the Instron 5566 universal testing machine (Instron, USA) in accordance with ASTM F1342-05. The dynamic puncture resistance was measured by a drop-tower machine with data acquisitions (type PCD 300A) according to NIJ Standard 0115.00-

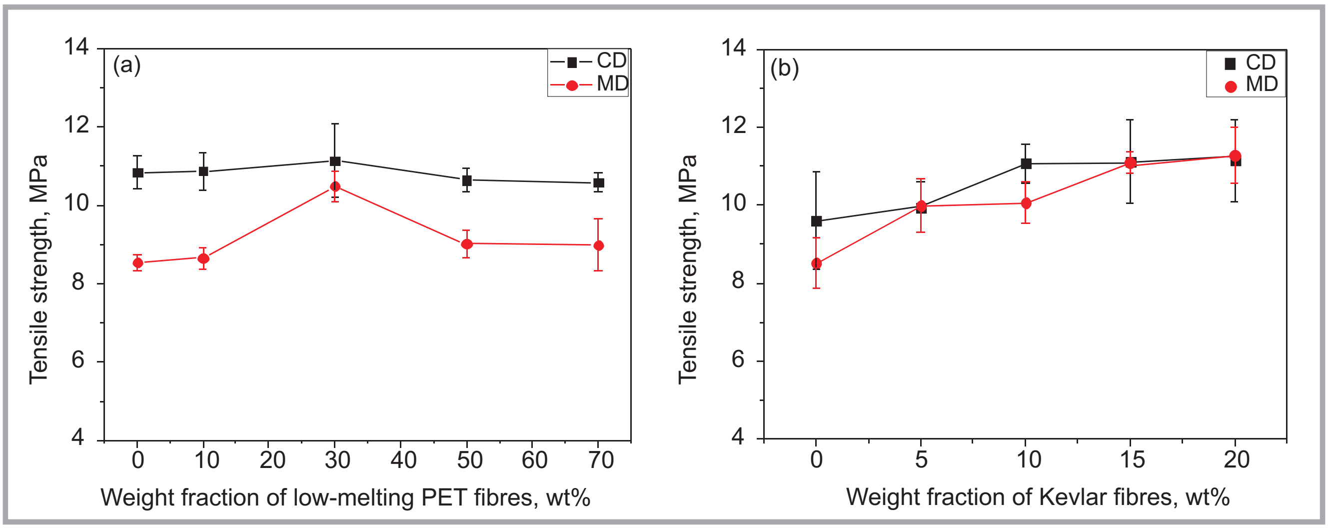

***Figure 3.** Tensile strengths of three-dimensional laminated composites which have (a) varying low-melting PET fibres and (b) Kevlar fibres on the surface nonwovens of the composite.*

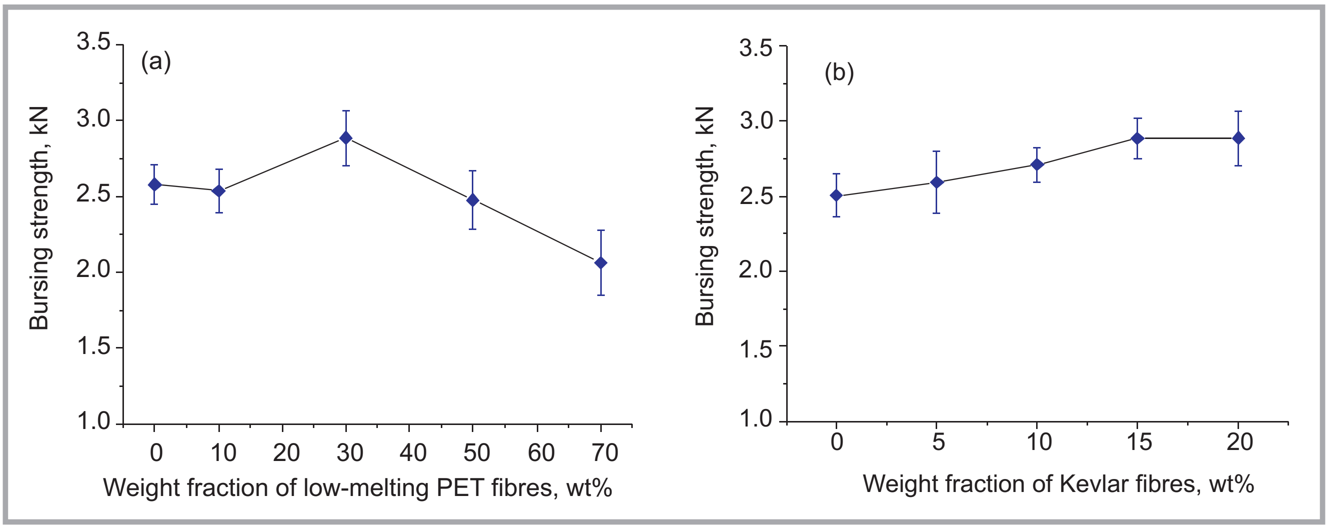

***Figure 4.** Bursting strength of three-dimensional laminated composites with (a) varying low-melting PET fibres and (b) Kevlar fibres on the surface of nonwovens.*

2000. The dynamic probe installed on the 2.8 kg load free fell from a height of 284 mm onto the specimen, which was clamped in the middle of square plates with a 40 mm diameter hollow. The specimens used for study of puncture properties were both sized 100 × 100 mm. The static and dynamic probes are displayed in ***Figure 2***. In this testing, ten specimens were assessed for each parameter of the composites.

Thermal-insulating testing

The thermal-insulating property was characterised by thermal conductivity(W/m·K) employing DXR-I-SPB Guarded-hot-plate apparatus (Xiangtan Huafeng Equipment Manufacture Co. Ltd, China) in relation to ASTM C177. Each test lasted 6 hrs, conducted at a temperature of 100 °C, and three specimens were simultaneously placed into the testing machine, in which cold water flowing at a velocity of 0.4 ml/s passes by the central calorimeter.

Stereoscopic microscopic scans

The transverse surfaces that originated from the 3D-laminated composites and fracture planes which emerged after mechanical testing were both examined using an optical microscope (SZ-CTV Olympus, Japan) furnished with Motic Images Multi-Focus Software (Micro-Optic Industrial Group Co. Ltd, Taiwan).

Results and discussion

Tensile property

The tensile results of composites with various low-melting PET fibres and Kevlar fibres on the surface nonwoven are presented in ***Figure 3***. The tensile strength is defined as the ratio of the maximum force in the force-displacement curve and the cross-section area. It is observed in ***Figure 3.a*** that the tensile strengths, whether it be the cross-machine (CD) or machine-direction (MD), are found to increase and then decrease with an increase in the weight fraction of the low-melting PET fibres, indicating the two-side effect of thermo-bonding fibres on the tensile strength. On the one hand, the growing thermo-bonding points enable the stress to transfer throughout the network of composites, hence improving the tensile property, but on the other, when low-melting PET fibre increases up to a certain region, its aggregations form a large thermo-bonding area in the composites; thus the stress concentration produced on

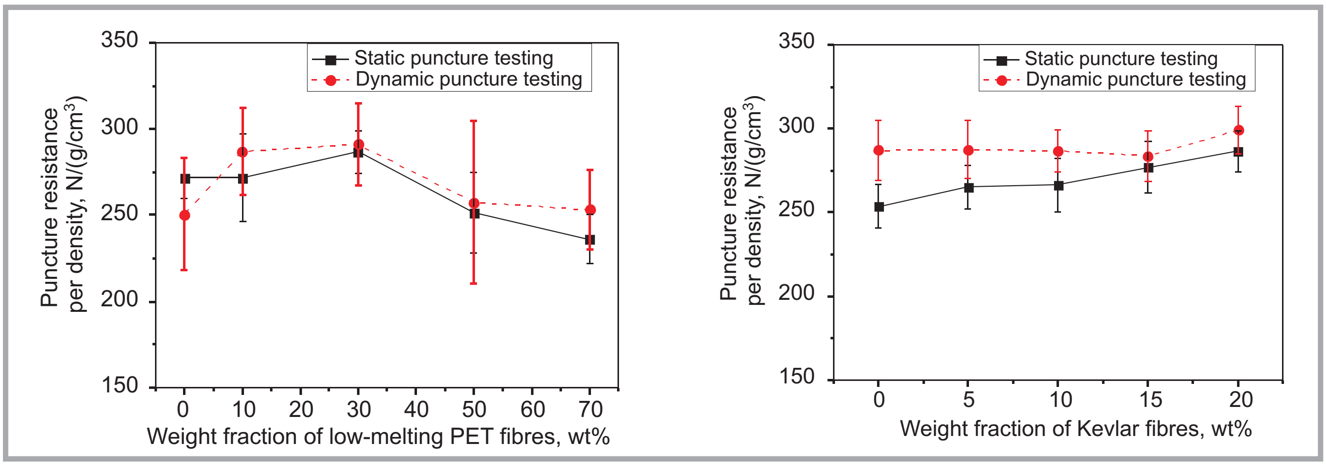

***Figure 5.** Static and dynamic puncture resistances of three-dimensional laminated composites with (a) varying low-melting PET and (b) Kevlar fibres contained on the surface of nonwovens.*

the periphery of the thermal bonding area results in diminished tensile strength. As a result, the highest tensile strength was reached when the composite had 30 wt% of low-melting PET fibres on the surface. In ***Figure 3.b***, low-melting PET fibres remain constant at 30 wt% when the mass fraction of Kevlar fibres varies on the surface of the composites. It is visible that the tensile strength increases dramatically when Kevlar fibres were added to the composites, from 0 to 20 wt%, which is attributed to the superior strength of Kevlar fibres, with their high molecular orientation. Furthermore the tensile strength in CD is not far from that in MD. However, the CD's tensile strength is more than the MD's, about 2.5 MPa with low-melting PET of 0 wt%, 10 wt%, 50 wt% and 70 wt%, shown in ***Figure 3.a***. It is presumed that the composite structure becomes more isotropic when the Kevlar fibres add up to 20 wt%; and the Kevlar fibres' mass-fraction determines the isotropic characteristic more significantly than low-melting PET fibres, as is seen by comparing ***Figures 3.a*** and ***3.b***.

Bursting property

Figure 4 shows the effects of low-melting PET fibres and Kevlar fibres on the bursting strength of three-dimensional laminated composites. The tendency of the bursting strength as the low-melting PET fibres-mass fraction increases is similar to that of the tensile strength. Likewise the bursting strength firstly increases and then decreases with an increase in low-melting PET fibres, as displayed in ***Figure 4.a***. The maximum busting strength reaches 2.89 kN when 30 wt% low-melting PET is contained on the surface of the composites, which also reflects the double-faced effect of low-melting PET fibres. Initially with an advisable increase in low-melting PET fibre added in, the higher tensile strength makes the composites resist against higher bursting strength. Nevertheless the addition of unnecessary extra low-melting PET fibres causes a decrease in the bursting strength, which is mainly governed by the inadequate elongation at break. This is due to the more and more brittle structure with the increase in low-melting PET fibres.

It is found from ***Figure 4.b*** that the bursting strength approaches the maximum at 15 wt% of low-melting PET. Even though the bursting strength of Kevlar/Nylon/low-melting nonwovens increases proportionally with the addition of Kevlar fibres in our study, the whole bursting property of multi-layer laminated composites depends on the agreement of the tensile strength and elongation at break for PET/low-melting PET nonwovens, glass fabric and Kevlar/Nylon/low-melting PET nonwovens, and hence the bursting strength no longer goes up when increasing above 15 wt% (see 20 wt%).

Puncture resistances properties

For the 3D laminated composite, nonwoven A containing Kevlar fibres passivated the probe tip, and nonwoven B was used to reduce the windowing of the fabrics. This structure composed was expected to resist against more puncture energy.

The proportions of thermo-bonding fibres and Kevlar fibres, as well as volume density of the composites are closely related to the puncture resistance property. In this part, the puncture property is expressed as the puncture resistance per density, in N/(g/cm^3). ***Figure 5.a*** indicates the effect of the weight fraction of low-melting PET fibres on the static and dynamic puncture resistances per density of limited composites. It is clear that the puncture resistance varies with weight fraction of thermo-bonding fibres, regardless of static and dynamic puncture properties. The tendency reveals that both the static and dynamic puncture resistances firstly climb up and then drop down, verifying that the static and dynamic puncture properties determine the flexibility and toughness of the composites. Comparing with its respective puncture resistance, the dynamic puncture resistance is inferior to the static puncture when the composite contains only Nylon and Kevlar fibres on its superficial nonwoven. When consisting of 10 wt% low-melting PET fibres, the dynamic puncture resistance seems a little higher than the static puncture resistance.

Of course, the Kevlar fibres act as a reinforcement of static and dynamic puncture-resisting properties, as shown in ***Figure 5.b***. Increasing the Kevlar fibres results in increments of the puncture force at the volume density. This trend is significant for static puncture property. Comparatively the dynamic puncture forces in constant low-melting PET fibres on both sides of the composites perform better than the static dynamic puncture property. However, the dynamic puncture force almost remains at 280 N at a unit volume of the composites from 0 ~ 15 wt% of Kevlar fibres. Until 20 wt%, the dynamic puncture property reveals an obvious augment, which is due to the non-uniform characteristic of Kevlar fibres dispersed in the nonwovens, as well as the needle-punching effect that penetrates the Kevlar fibres into the PET layers. For the static puncture property, the increasing addition of Kevlar fibres brings about an improvement in the static puncture property.

The maximum static and dynamic puncture resistances reach 286.6 and 299.3 N/(g/cm^3), respectively, as shown in ***Figure** 5*. These values are much higher than those of composites with double layers of nonwovens A and glass fabric, reaching 259.5 (static) and 240.5 (dynamic) N/(g/cm^3), as shown by Li et al. [12]. This implies that the addition of nonwoven B is remarkably beneficial for the improvement of both the static and dynamic puncture resistances per unit volume density.

As observed in ***Figure 6.a***, the puncture probe penetrates through the inter-fibre of the glass fabric after static puncture damage. But in ***Figure 6.b***, a relatively neatly damaged edge is found after the dynamic puncture test, showing the brittle fracture of the composites. This indicates the different mechanisms for static and dynamic puncture tests: interfacial friction for the static test and fibre damage for the dynamic test.

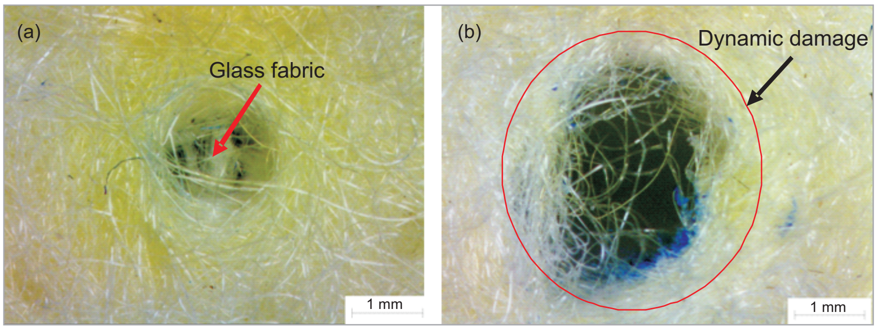

***Figure 6.** Fracture surfaces of composites where 30 wt% low-melting PET is contained after (a)static and (b)dynamic puncture testing.*

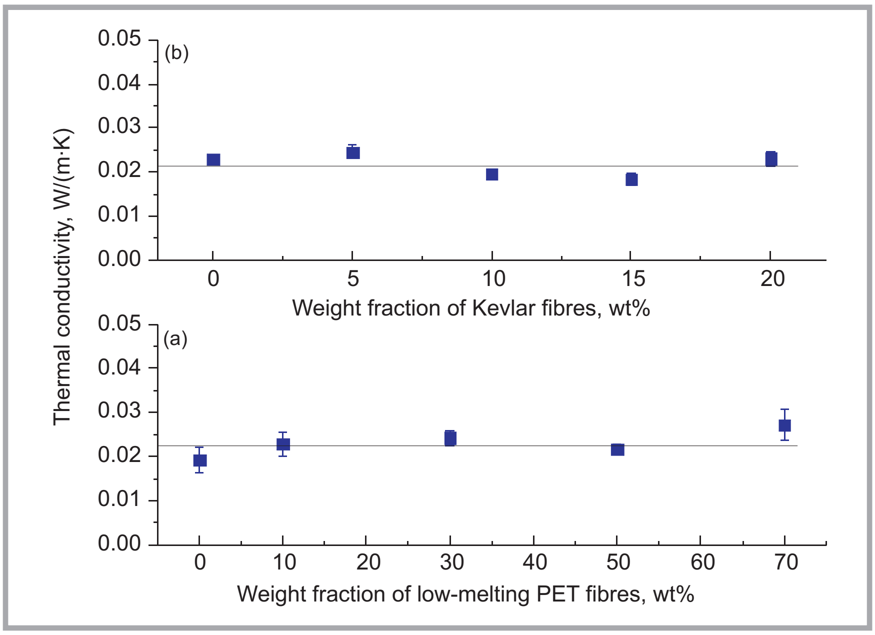

***Figure 7.** Thermal insulating properties of three-dimensional laminated composites with varying mass fractions of low-melting PET fibres (a)and Kevlar fibres (b) contained on surface nonwovens of the composites.*

Thermal insulation property

The thermal conductivity reflects the thermal insulation when heat flux transfers along the vertical direction of the composites. Owing to the higher amount of low-melting PET fibres contained in nonwoven B, molten thermo-bonding fibres fill the inter-space among fibres, forming a thermoplastic layer, which insulates from thermal transmission in the composites. In addition, the needle-punched effect makes the structure compact, which also benefits the thermal insulation. Therefore when superficial low-melting PET fibres or Kevlar fibres change, the thermal conductivity is in range of 0.015 - 0.025 W/m·K (see ***Figure** 7*), which reaches the levels of thermal-insulating materials.

Conclusion

This study prepared organic/inorganic composites composed of two different kinds of nonwovens and a glass fabric via multiple needle-punching and thermal bonding techniques. The superficial nonwoven consisted of recycled Kevlar fibres designed to meet environmental protection requirements and reduce costs in application. The other nonwoven comprised a majority of low-melting PET fibres aimed at acting as thermal bonding layers.

With an increase in low-melting PET fibres in the superficial nonwovens, the mechanical properties and puncture resistances show an upward and then a downward trend, with the optimum at 30 wt% of low-melting PET fibres. However, as the amount of recycled Kevlar fibres is increased, only the tensile strength and static puncture resistance proportionally improve. The bursting strength increases up to 15 wt% of Kevlar fibres. The dynamic puncture resistance is almost maintained, increasing from 0 - 15 wt%, and is remarkably boosted at 20 wt%. In addition, composites with varying low-melting PET and Kevlar fibres have a thermal conductivity of 0.015 - 0.025 W/m·K, reaching the requirements of thermal insulation materials. Comparatively the additional PET/low-melting PET nonwoven layer effectively contributes to the improvement of both the static and dynamic puncture resistances, as well as the thermal insulation. The composites prepared are expected to be used as body armour interlayers under high-temperature circumstances in the future. In the following study, we will focus on the thermal stability of these composites to determine their operating temperature.

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