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Inter-/Intra-Laminar Reinforced Hybrid Fibre Composites by Needle Punching and Thermal Bonding: Evaluation of Mechanical and Static Puncture Properties

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Abstract

This study comparatively presents the static puncture property of different structures of intra-/inter-laminar reinforced hybrid composites via needle punching and thermal bonding techniques. The tensile and bursting properties of two composites with inter-laminar reinforcement by needle-punching and inter-and-intra- laminar reinforcement by both needle punching and using Kevlar fibres were also evaluated comparatively. The significance of process parameters including the low-melting PET fibre content, take-up speed of the punching machine, the plied orientation between the nonwoven and fabric and thermal bonding on the static puncture resistance was firstly investigated to seek out the significant parameters. The effects of significant processing parameters on static puncture and mechanical properties were explored afterwards. The research result shows that the plied orientation, low-melting PET content and thermal bonding affect the static puncture resistance most significantly. The maximum tensile strength and bursting strength occurred when hybrid composites after thermal bonding were composed of parallel-plied nonwovens and 90°-orientated glass fabric, as well as 70 wt% low-melting PET fibres. Recycled Kevlar fibre reinforcement dissipates additional static puncture resistance, and makes the static puncture resistance higher, as well as the tensile and bursting strengths for resultant hybrid fibre composites. Employing recycled Kevlar fibres is economical for the fabrication of hybrid composites. Diversified economical hybrid composites will be applied as a wall interlayer or garment interlining in the future.

Key words: *hybrid composite, needle punching, thermal bonding, puncture, recycle.*

Introduction

A large amount of fibre reinforced composites are applied in the aerospace, automobile, transportation and building industries due to their high specific strength and specific modulus. Fibre reinforced composite has widely substituted conventional materials such as metal, wood and so on because of its low cost and small weight. It is composed of reinforcing fibres including glass fibre, Kevlar fibre and others, as well as resin matrix. Of interest is the resin matrix, which is made by the textiles nonwoven processing technique after thermal bonding, such as polypropylene fibres [1, 2], low-melting PET fibres [3], polyester fibres [4, 5], polyamide fibres [6] and bicomponent fibres [7].

Many studies of nonwoven resin focused on their mechanical properties such as tensile, flexural, burst and impact performance [8 - 12]. In order to reinforce its property, high-strength fibres or fabric was inserted between nonwovens. In such cases, the fibre interface strength becomes a persistent problem for researchers. Therefore some chemical methods or stitching are used to improve this problem [2, 4, 13 - 18]. However, the chemical methods bring environmental pollution because the solvents are poisonous. The stitching periphery of composites is subjected to damage, and bears lower forces due to stress concentration. In this study, the lateral needle punching technique is used for enhancing the interface strength.

Fibre reinforced composites occasionally suffer from sharp-pointed impacts in their long service life. The static puncture resistance of composites is also taken into account. P-aramid fabric and ultra-high molecular weight polyethylene fabric were usually used as the reinforcement, and Polyethylene (PE) and Surlyn (SU) [19], or low-density Polyethylene (LDPE) and epoxy resin [20], or Nylon [21] were regarded as matrix material. Especially for those high-viscosity thermoplastic polymers such as LDPE, it is difficult to completely impregnate them into the inter-fibre space of woven fabrics, as indicated by Kim et al. [20]. If in these woven fabrics there still remained some spaces, it would form a stress concentration region and the whole composite would be damaged by the interspaces of fabrics, which decreases the strength of whole composite as a resultant. In this study, the nonwoven needle punching technique is explored to reinforce the strength of whole composites by forming needle punching points among woven fabrics, thus the stress concentration becomes scattered and the whole composite presents higher mechanical strength. Low-melting PET fibres form thermal-bonding points after thermal treatment, and act as the matrix in composites via a hot press. Kevlar fibres, used as intra-reinforcing fibres in the nonwoven surface, also have ability to further improve mechanical properties, especially the static puncture property. Therefore, the aim of this research work is to discuss the process parameters including take-up speed, plied orientation and thermal bonding on static puncture property of two different structures-inter-laminar reinforced composites (PET/Glass control composites) and intra-and-inter-laminar reinforced hybrid composites (Kevlar/PET/Glass hybrid composites).

Experimental

Experimental materials

18 tex PET fibers and 36 tex low-melting PET fibres (LPET) of 51 mm length were supplied by Far Eastern New Century Corporation, Taiwan. Glass plain fabrics were provided by Jinsor-Tech Industrial Corp., Taiwan, and their physical properties are shown in ***Table 1***. Recycled Kevlar® fibres of 10.8 tex fineness and 50 - 60 mm length were taken from 25.38 tex D Kevlar® unidirectional selvages provided by DuPont Company, USA. 54 tex high-strength Nylon 6 fibres of 64 mm length, 1.09 cN/tex tenacity, and 24.7% elongation were supplied by Taiwan Chemical Fibre Co. Ltd., Taiwan.

Table 1. Physical properties of glass fabric.

Structure	Linear density	Surface weight, g/m^2	Density, picks/cm	Thickness, mm
Plain	1 K	328 ± 1.5	10 × 13	0.310 ± 0.017

Table 2. Mechanical properties of K-nonwoven.

Kevlar, wt%	LPET, wt%	Nylon, wt%	Tensile strength, (kN/m)^a	Bursting strength, kN	Puncture resistance, N
20	30	50	9.7 ± 0.78	1.16 ± 0.18	15.80 ± 2.60

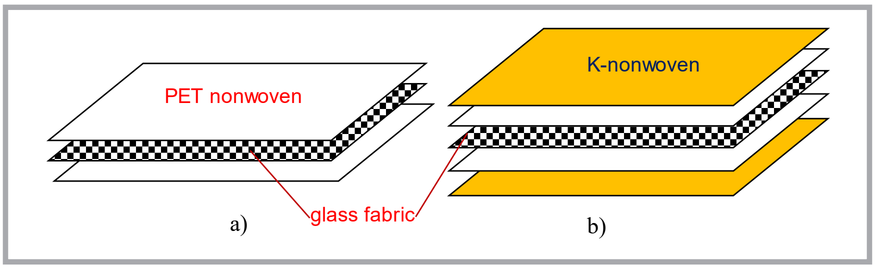

Figure 1. Schematic diagrams of a) control composite and b) hybrid reinforced composite.

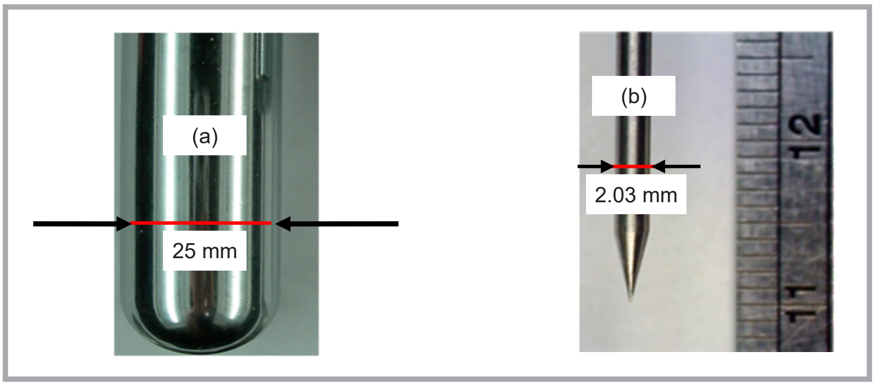

Figure 2. Burst (a) and static puncture (b) probes.

Sample preparations

Varying weight proportions of low-melting PET were blended with PET fibres and then needle-punched into nonwovens. The low-melting PET fibres were varied as 30, 50, 70 & 100 wt%, and the take-up speeds were changed as 2.10, 1.40, 1.07 and 0.85 m/min. The stroke frequency was 150 needles/min. The unit weight of nonwoven was 150 g/m^2. The needle-punched density conversely proportionally transformed with the take-up speed of nonwovens.

After nonwoven preparation, a layer of glass fabric was inserted between PET/low-melting PET nonwovens and then needle-punched, forming control needle-punched composites. The effect of the plied orientation between the nonwoven and glass fabric was discussed in relation to tensile, bursting and static puncture resistances. The arrangement of plied orientation was mentioned in our previous study [22]. Afterwards 200 g/m^2 of Kevlar®/Nylon/Low-melting PET nonwoven (K-nonwoven) was compounded with control composites, resulting in hybrid reinforced composites. The mechanical properties of K-nonwoven are listed in ***Table 2***. Moreover the influence of K-nonwoven was investigated by comparison of the above three properties' behaviour for low-melting PET control composites and low-melting PET/Kevlar hybrid reinforced composites. Both the control composites and hybrid fibre composites (as given in ***Figure 1***) were calendered through 1.5-mm-distance rollers at a temperature of 160 °C and speed of 0.5 m/min. As a result, the addition of low-melting PET fibres and Kevlar fibres were used to form an intra-laminar reinforced structure, and needle-punching bonding formed an inter-laminar reinforced structure. Based on these techniques, intra-/inter-laminar reinforced hybrid composites were successfully fabricated.

Property testing

A tensile test, burst test and static puncture test were performed using an Instron 5566 Universal Tester (Instron, USA) with a 1 kN load cell, according to ASTM D5035, ASTM D1883 & ASTM F1342, respectively. Tensile samples were strips of 180 × 25 mm^2, measured at 300 mm/min. Squared samples of 150 × 150 mm^2 and 100 ×100 mm^2 were used for burst and static puncture tests. The probes travelled at a speed of 100 and 508 mm/min, and then penetrated through the samples. Specifications of the probe for the burst and static puncture tests are displayed in ***Figure 2***.

__Table 3.__ Univariate significance of process parameters on the static puncture property. a) R^2 = 0.750 (Adjusted R^2 = 0.715). __Please note__: Content indicates the low-melting PET fibre content, orientation shows the plied orientation between the nonwovens and woven fabric, and thermal is an abbreviation of thermal bonding.

Source of variation	Sum of squares	df	Mean Square	F-value	Sig
Corrected model	11783.116[a]	29	406.314	21.703	0.000
Intercept	50629.251	1	50629.251	2704.302	0.000
Content	816.525	3	272.175	14.538	0.000
Orientation	2400.651	2	1200.326	64.114	0.000
Speed	481.475	3	160.492	8.572	0.000
Thermal	157.151	1	157.151	8.394	0.004
Content ×orientation	1396.921	6	232.820	12.436	0.000
Orientation × thermal	1724.805	2	862.403	46.064	0.000
Content × thermal	139.191	3	46.397	2.478	0.062
Content × speed	416.645	9	46.294	2.473	0.011
Error	3931.566	210	18.722	-	-
Total	174106.420	240	-	-	-
Corrected Total [a]	15714.682	239	-	-	-

__Table 4.__ Duncan test for plied orientations.

Plied orientation	N	Subset	
		1	2
3.00	40	23.0906	-
1.00	160	24.5057	-
2.00	40	-	33.0253
Sig.		0.093	1.000

Significance analysis

Significance analysis of process parameters including low-melting PET fibres, take-up speed and plied orientation were investigated with respect to the tensile, burst and static puncture properties of control composites using SPSS 19 software. All the analysis was based on One-way ANVON F-test statistic at a significance level of 5%. Four factors: the low-melting PET content, take-up speed (needle-punching machine), plied orientation between the nonwoven and fabric as well as thermal bonding are discussed in the significance analysis. Thereinto, low-melting PET content has four levels including 30 wt%, 50 wt%, 70 wt%, and 100 wt%; with the take-up speed also having four levels including 2.10 m/min, 1.40 m/min, 1.07 m/min, and 0.85 m/min; the plied orientation - three levels including 0°/0°, 90°/90°, & 0°/90° and thermal bonding - two levels (thermal-bonded and non-thermal-bonded). Therefore the between-group degrees of freedom (df) of the three factors are, successively, 3, 3, 2, 1 for each independent variable. The F-value (F) is defined as the ratio of the mean-square between each variable and the total error. Sig shows the F-distribution probability when the F-value (F) is no smaller than the critical value. Since the Sig value is lower than the significance level (0.05), the parameter (variable) affects the static puncture property more significantly. Conversely Sig > 0.05 shows it affects the static puncture resistance insignificantly. The critical value is the F-statistic following the F-distribution with K−1, N−K degrees of freedom under the hypothesis that each variable (process parameter) affects the static puncture property insignificantly as compared to the between-group and within-group variability; where K shows the number of levels for each factor, and N the total number of samples. Six samples were repeatedly measured for calculation within-group variability. Through SPSS analysis, processing parameters that significantly affect the static puncture resistance are researched.

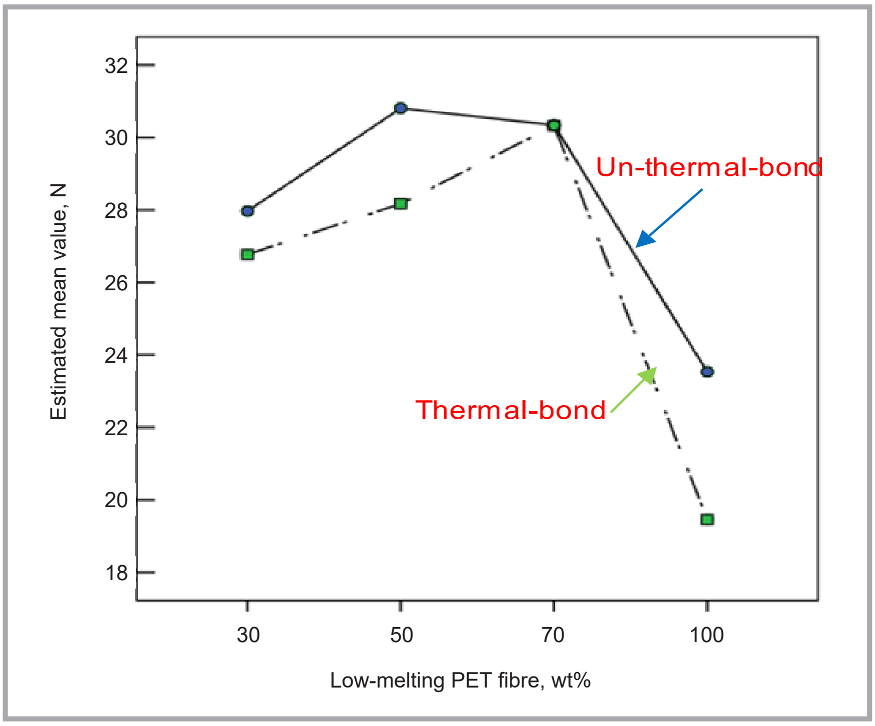

__Figure 3__. Estimated value of the static puncture resistance of a control composite with different low-melting PET fibres.

Results and discussions

Effect of process parameters on the static puncture property of the control composite

In our previous study, the effects of the low-melting content and needle-punched density on thermal-bonded and un-thermal-bonded composites, and the influence of plied orientation on two composites containing a constant low-melting content and processing at a constant needle-punching density were investigated, respectively [22]. In this study, all process parameters as well as the low-melting PET content, take-up speed, plied orientation and thermal bonding effect were all discussed with relation to the static puncture property of control needle-punched composites. As shown in ***Table 3***, the plied orientation, low-melting content and their interaction affect the static puncture resistance most significantly. In addition, interaction between the plied orientation and thermal bonding as well as between the low-melting content and plied orientation obviously transforms the static puncture property. Therefore the effects of the low-melting PET content, plied orientation and thermal bonding on the tensile and bursting property are investigated in the following study. For the main effect, plied orientation reveals a high influence on the static puncture because of the highest F value, which implies that an appropriate structure match between the nonwoven and fabric produces an excellent static puncture-resisting composite.

Table 4 shows the mean puncture resistance of each plied orientation based on Duncan tests. Duncan's multiple range test, shortened as the Duncan test, is used to compare the means between two groups of the control composite with different plied orientations based on one-way ANOVAN. It shows that of all the estimated values, the arrangement with parallel-plied nonwoven and 90°-plied fabric exhibits the highest static puncture resistance property. Interestingly ***Figure 3*** displays that thermal-bonded composites have higher estimated static puncture resistance than non-thermal-bonded ones for the all values estimated. For control composites with parallel-plied nonwoven and 90°-orientated glass fabric, thermal bonding improves the static puncture resistance, as seen in ***Figure 4***. This can be explained by ***Table 3***, showing that the plied orientation and thermal bonding have an interactive effect. ***Figure 3*** also shows that the optimal low-melting PET fibre content varies for non-thermal-bonding and thermal-bonding composites. According to estimated values, when containing 50 wt% low-melting PET fibres, the non-thermal-bonded composite has the highest estimated mean puncture resistance. However, the addition of 70 wt% low-melting PET fibres is best for thermal-bonded composites. This difference is related to fibre volume fractions and plasticity between composites. For the non-thermal-bonding composite, the compression contact and surface deformation between the testing probe and samples resists puncture energy [23, 24]. After the thermal bonding process, the composite becomes stiffer and more plastic. A part of low-melting PET impregnated into the fabric interspace confines the viscoelastic deformation of the whole composite. Therefore the contact pressure becomes the dominant source of resisting puncture [25], which can be explained by the typical puncture behaviour of composites. It is found from ***Figure 5*** that the puncture behaviour of the non-thermal-bonded composite lags behind that of the thermal-bonded composite, with the former having a lower initial slop than the latter. This suggests viscoelastic and compression deformation of the non-thermal-bonded composite.

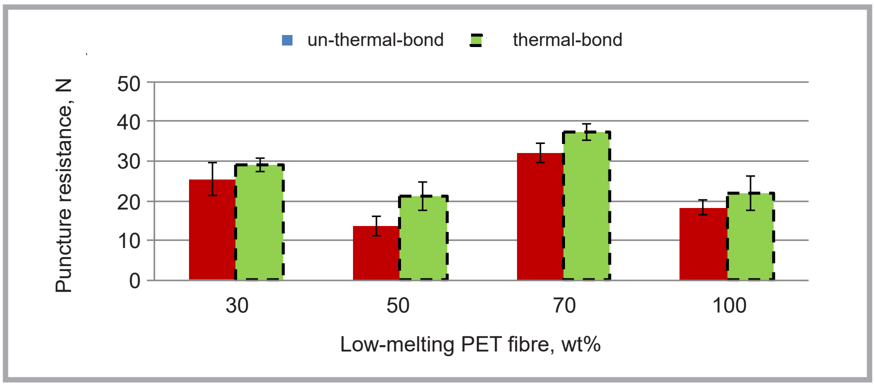

Figure 4*. Static puncture resistances of control composites with different low-melting PET fibres arranged with parallel-plied PET nonwovens and 90° - orientated glass fabric.*

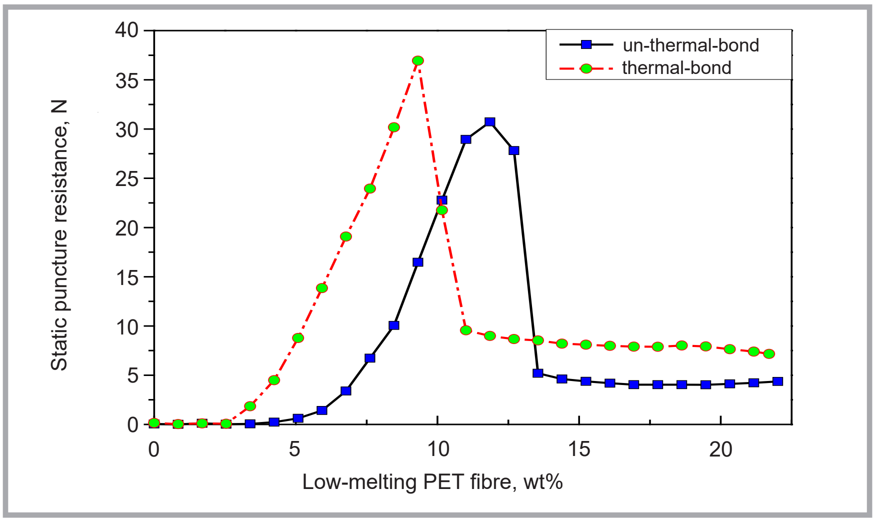

Figure 5*. Static puncture behaviors of non-thermal-bonded and thermal-bonded composites containing 70 wt% low-melting PET fibres.*

Effects of process parameters on the tensile property of control composites

Figure 6 (see page 88) shows the tensile strength of control composites in relation to variations on the low-melting PET fibre content, plied orientation and thermal bonding. Control composites have two-directional tensile strength, including machine-direction (MD) strength and cross-direction (CD) strength. MD indicates the output direction of the needle punching machine, and CD is perpendicular to MD. It is clear that thermal bonding improves the tensile strength for different plied orientation and low-melting PET fibre content, which shows that molten-state low-melting PET further enhances the interface layer of nonwoven and glass fabric other than needle-punch bonding. The tensile strength with different low-melting PET fibres reveals that composites containing 70 wt% low-melting PET fibres had the highest tensile strength, as seen in ***Figure 6.a***. This is due to the amount of total fibres and compactness of interpenetrating fibres. ***Figure 6.b*** shows that the tensile strength firstly increased and then decreased an increase in low-melting PET fibres for non-thermal-bonding and thermal-bonding composites. However, the tensile strength changes slightly even with an increase in low-melting PET fibres, as shown in ***Figure 6.c***. This indirectly confirms the interaction of the low-melting PET content and plied orientation, the reason for which being that nonwovens composed of low-melting PET fibres were cross-plied, and the amount of fibres in each direction was close to each other.

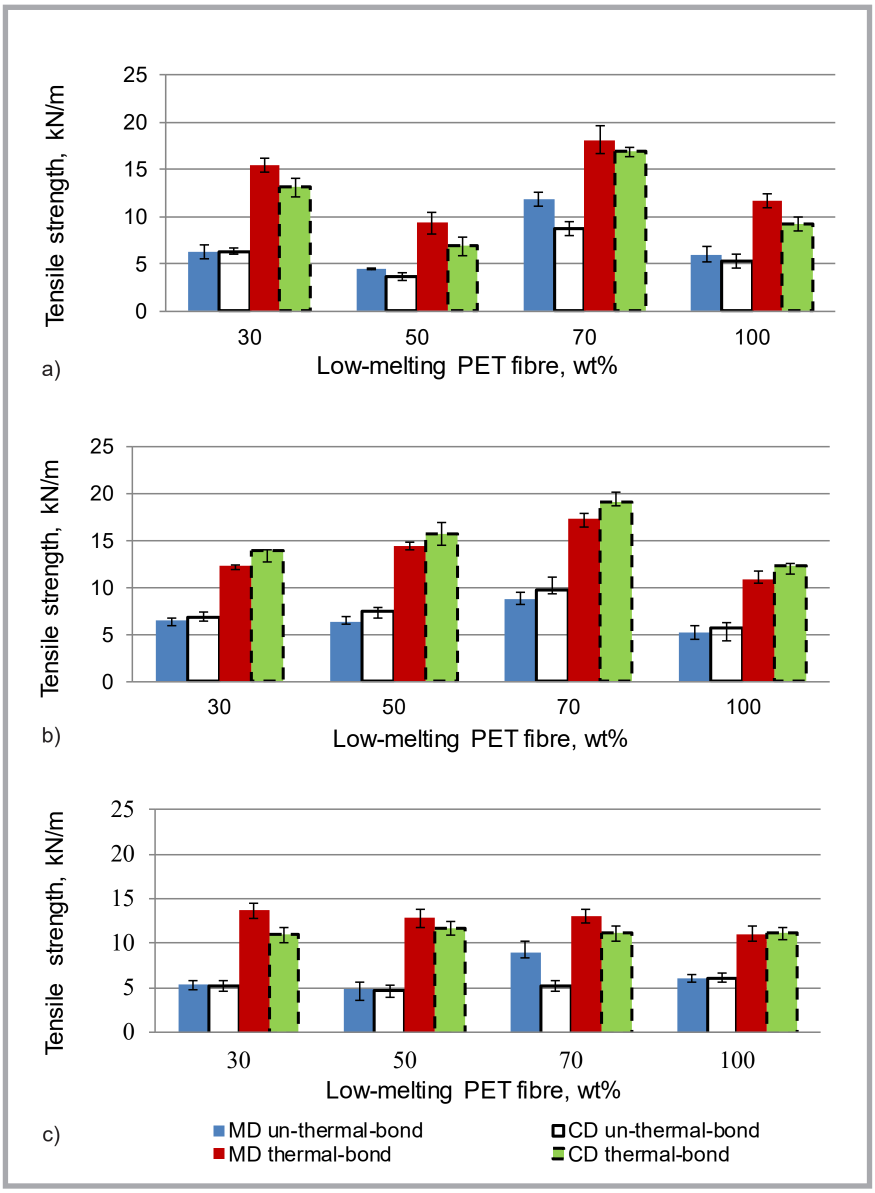

Figure 6. *Tensile strengths of non-thermal-bonded and thermal-bonded control needle-punched composites with parallel-plied PET nonwovens and 0° (a) or 90° (b) - orientated glass fabric, as well as cross-plied PET nonwovens and 0° - orientated glass fabric (c).*

Table 5. *Tensile, burst and static puncture properties of control composites and hybrid reinforced composites.*

Mechanical property		Hybrid reinforced composite		Control composite	
		Non-thermal-bond	Thermal-bond	Non-thermal-bond	Thermal-bond
Tensile strength, kN/m	MD	17.75 ± 0.92	36.16 ± 1.61	8.77 ± 0.75	17.36 ± 0.94
	CD	38.00 ± 2.38	39.96 ± 6.69	9.81 ± 1.31	19.04 ± 0.37
Bursting strength, kN		3.98 ± 0.29	2.50 ± 0.15	0.99 ± 0.017	1.18 ± 0.101
Puncture resistance, N		74.51 ± 11.28	95.71 ± 4.42	31.98 ± 2.36	37.33 ± 1.89

Effects of process parameters on the burst property of control composites

Figure 7 shows the bursting strength of control composites with different low-melting PET fibre content, plied orientation and thermal bonding. Similarly 70 wt% low-melting PET fibres have the highest bursting strength, which agrees with the static puncture and tensile strength results. This is because the bursting strength is related to the strength, elongation and fibre distribution in each direction. Composites were damaged in the most weakened points during the burst test, therefore the bursting strength was higher for a more isotropic structure [26]. For different plied orientation, the arrangement with parallel-plied nonwoven and 90° glass fabric displays a higher bursting strength than others, which also corresponds to the tensile result in ***Figure 6.b***, and confirms that the isotropic nonwoven structure is positive for burst improvement. Analogously thermal bonding improves the bursting strength of the control composites. The difference is that composites with parallel-plied nonwoven have no significant distinction between non-thermal-bonding and thermal bonding. However, thermal-bonded composites with cross-plied nonwoven have a much higher bursting strength than non-thermal-bonded ones, which may reflect a more significant impact of the nonwoven structure on the bursting strength.

Property comparison of control and hybrid reinforced composites

Table 5 shows the static puncture resistance, tensile strength and bursting strength of control and hybrid reinforced composites. Hybrid reinforcement by K-nonwoven, needle punching and thermal bonding significantly increased the static puncture resistance from 31.98 N to 74.51 N, which illustrates the high-performance fibres' remarkable effect on the static puncture property. Through thermal bonding, the static puncture resistance of hybrid reinforced composites increased up to 95.71 N, which is an improvement of 20 N compared to the non-thermal-bonded composite. The difference between them is larger than that of the control composite. Low-melting PET was bonded woth Kevlar® , Nylon and PET fibres, thereby increasing the strength and compactness of the whole hybrid composite. As a result, the contact pressure when the puncture probe penetrates into the composite leads to a much larger deformation region, which improves the static puncture resistance [25]. The more compact structure of thermal-bonded hybrid reinforced composites can be indirectly explained by the bigger initial slope in ***Figure 8***.

The tensile behaviours of non-thermal-bonded and thermal-bonded control composites and hybrid reinforced composites are comparatively given in ***Figure 9***. After K-nonwoven reinforcement, the tensile strength rises up to 17.75 kN/m (MD) and 38.00 kN/m (CD), respectively, improving by 102.39% and 108.29% as compared to the control composites; and simultaneously, the breaking strain is reduced to 90% and 62%, respectively, in MD and CD (see ***Table 4***). This demonstrates the K-nonwoven reinforcement's influence on the tensile property, as high-tenacity Kevlar® fibres and Nylon 6 fibres were contained in the composites and endured a higher tensile load than the control composite. However, the elongation of the hybrid reinforced composite is decreased, especially in CD (***Figure 9.a***). After thermal-bonding, a larger improvement is found for hybrid reinforced composites, as displayed in ***Figure 9.b***. However, the thermal-bonded hybrid reinforced composites show complicated tensile behaviour, which implies the multi-level damage mode of hybrid reinforced composites, that is, tensile damage is firstly generated from the glass fabric, then the PET nonwoven, and finally the K-nonwoven. During the tensile process, the tensile load transferred from the glass fabric to the nonwoven via needle-punching points, and then shifted from the low-melting PET/PET layer to the K-nonwoven layer. After the tensile force reached the maximum breaking elongation, the whole composite was completely ruined. The tensile strength of hybrid reinforced composites ultimately reaches 36.16 kN/m (MD) and 39.96 kN/m (CD) (see ***Table 4***). Furthermore the difference in tensile strength between CD and MD becomes smaller after thermal-bonding, which may be responsible for the higher static puncture resistance of thermal-bonded hybrid composites. On the other hand, the control composite presents a brittle tensile character after thermal-bonding.

Comparatively non-thermal-bonded hybrid composites displayed a four times higher burst strength than control composites, as shown in ***Figure 10*** (see page 90). This explains that high tenacity fibre played a dominant role for the burst property. After thermal bonding, the bursting strength and breaking elongation became lower, the reason being that lower tensile breaking elongation leads to a lower bursting strength, which corresponds well with the results in ***Figure 9***. The bursting strength results from the interactive effect of the tensile strength and breaking elongation for each direction. Therefore thermal bonding reacts to the burst property differently from the tensile property compared with ***Figures 9*** and ***10***.

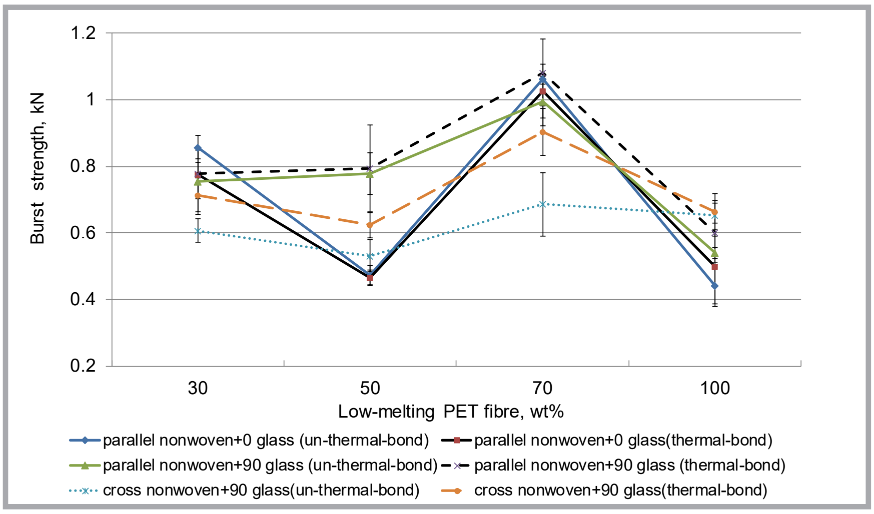

Figure 7. *Bursting strengths of control needle-punched composites with parallel-plied nonwovens and 0° or 90°-orientated glass fabric, and that with cross-plied nonwovens and 90°-orientated glass fabric after being non-thermal-bonded or thermal-bonded.*

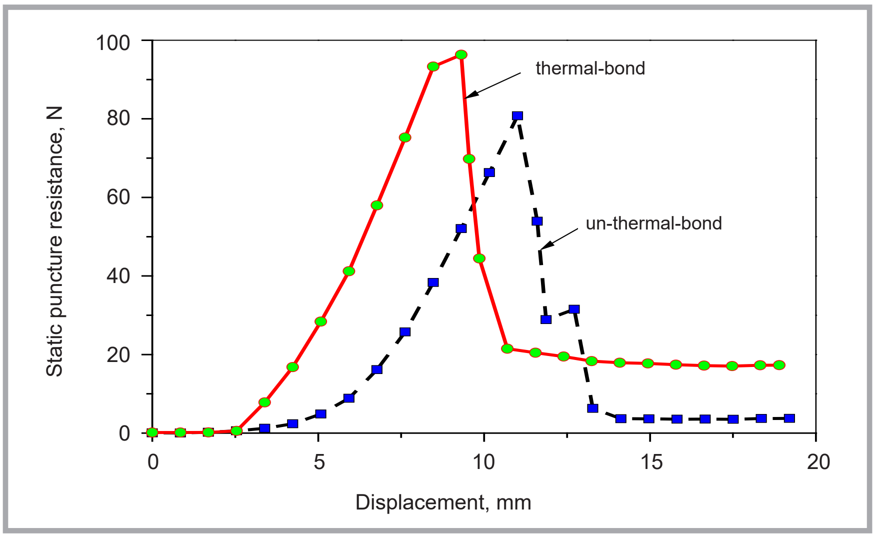

Figure 8. *Static puncture behaviours of hybrid reinforced composites after being non-thermal-bonded or thermal-bonded.*

Conclusions

This study comparatively explores the static puncture and mechanical properties of different structures of inter-/intra- laminar reinforced hybrid composites. The control composite composed of low-melting PET nonwoven and glass fabric is reinforced by inter-laminar using the needle punching technique. The other hybrid fibre composite made with Kevlar nonwoven, low-melting nonwoven and glass fabric has an inter-and-intra- reinforced structure via needle punching and thermal bonding techniques. The effects of process parameters including the low-melting PET fibre content, take-up speed, plied orientation and thermal bonding on the static puncture resistances of low-melting PET reinforced control composites were established using significance analysis. A property comparison of the hybrid reinforced composite and control composite was conducted for static puncture resistance as well as tensile and burst properties.

In a significance analysis, the interaction effects between the plied orientation and thermal bonding as well as the low-melting PET fibre content and thermal bonding were found to be more significant for static puncture resistances of the control composite. After thermal bonding, the static puncture resistance estimated firstly increased and then decreased with an increase in low-melting PET fibres. The critical low-melting PET content was 70 wt%. From the whole estimated value, we can state that thermal bonding was negative for the improvement

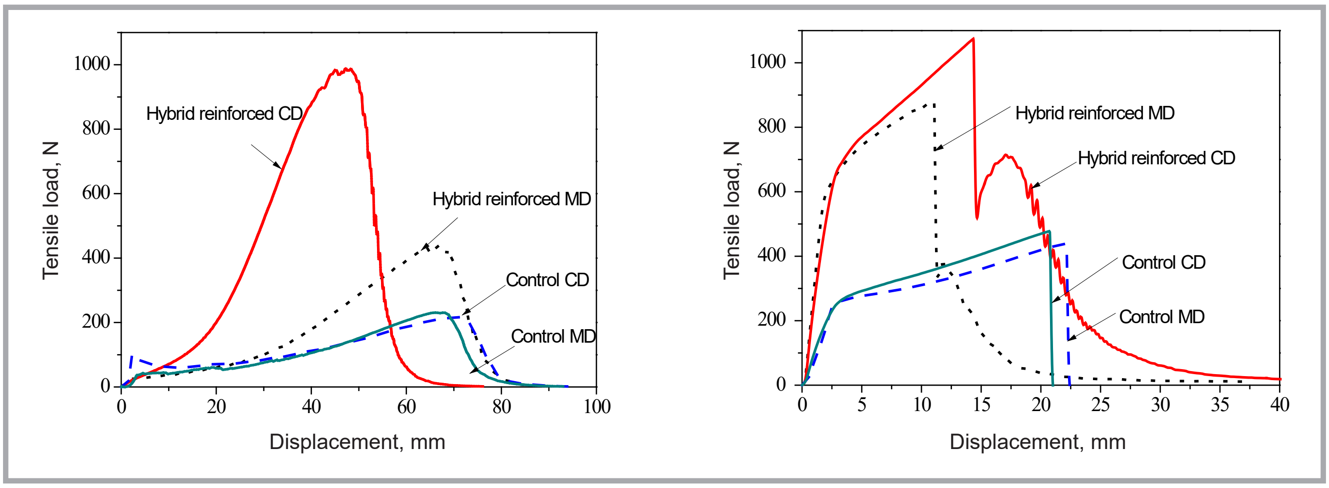

Figure 9. Tensile curves of control composite and hybrid reinforced composite that contained 70 wt% low-melting PET fibres in parallel-plied PET nonwovens and 90°-orientated glass fabric non-thermal-bonded (a) and thermal-bonded (b).

of static puncture resistance. For control composites with parallel-plied nonwoven and 90°-orientated glass fabric, thermal bonding improved static puncture resistance.

The maximum tensile strength and bursting strength occurred when control composites were composed of parallel-plied nonwovens and 90°-orientated glass fabric, as well as 70 wt% low-melting PET fibres. At the optimal process parameters, the tensile strength and bursting strength first increased and then decreased with an increase in low-melting PET fibres.

Inter-and-intra-laminar hybrid composites reinforced by needle punching and Kevlar fibres possessed higher static puncture resistance as well as tensile and bursting strength compared to the inter-laminar reinforced control composite. Contact pressure and planar deformation between the composites and probe was responsible for static puncture resistance. A prediction model of static puncture resistance will be constructed in the following study. The isotropic structure of the nonwovens and fabric affected the tensile strength, but the homogeneity nonwoven structure influenced the bursting strength significantly. Furthermore the static puncture resistance correlates well with the tensile and bursting strength.

The intra-/inter-laminar hybrid fibre reinforced composite has excellent static puncture resistance as well as tensile and bursting strength. Recycled Kevlar fibre improves the static and mechanical properties of hybrid composites due to the additional intra-laminar reinforcement. Therefore the intra-/inter-laminar reinforced structure has prospective applications as a wall inter-layer and garment interlining in the future. In addition, recycled Kevlar fibres are economical for composite fabrication, and also have benefits for environmental protection.

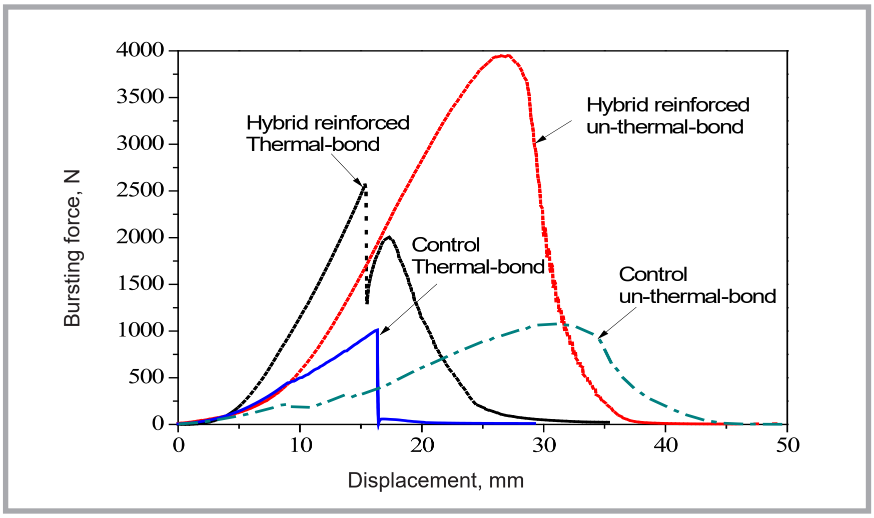

Figure 10. Bursting curves of control composite and hybrid reinforced composite that containing 70 wt% low-melting PET fibres in parallel-plied PET nonwovens and 90°-orientated glass fabric after being non-thermal-bonded (a) and thermal-bonded (b).

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