NCF/BMI COMPOSITE MATERIALS: EFFECT OF STITCHING THREADS

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1. Introduction

In recent years, the use of stitching technology is currently regarded as one approach for the cost-effective assembly of textile fabrics, including those with multi-layered non-crimp fabrics (NCFs)[1–2]. Carbon fiber non-crimp fabric, with many advantages such as less fiber crimp and higher mechanical properties, better drapability, better preforming and so on, has great potential for its application in the field of advanced resin matrix composites. Meanwhile, stitching threads used for NCFs are commonly made of polymer fibres such as polyester(PET) and nylon, etc. However, the stitching threads, being dissimilar to the reinforcing fibers and the matrix, may affect the overall performance of fiber/resin composites[3-4]. Therefore, it is important to assess the interfacial bonding between the stitching threads and the matrix.

Meanwhile, the use of NCF/bismaleimide(BMI) composites is more and more popular, especially in liquid composite moulding (LCM) process. Thus, BMI resins have played an important role as matrix materials in high-temperature aerospace structural applications due to their superior thermal stability and fatigue resistance at high humidity[5]. In such applications, the composites are exposed to harsh and changing environments featuring a wide range of temperatures and “hot-wet” exposures, which accelerate decline in their mechanical and other properties[6]. A variety of correlative experimental and theoretical researches have been done to discuss the impact of aging on the material performance. Some work focused on moisture absorption mechanism in BMI resin and its composites. A two-stage diffusion model[7] has been established: the initial fast diffusion can be predicted by Fick’s law, but no equilibrium uptake was observed after nearly a year, while the second stage considered the structural relaxation induced by absorbed moisture. The similar result came from the analysis of Li[5], and his FTIR results showed that the non-Fick diffusion behavior was due to the hydrogen bonding between water and the matrix.

During the moisture absorption of composites, physical changes such as micro-cracks propagation and swelling, as well as chemical changes such as hydrolysis and chemical scission increase, can degrade properties of the materials. Also, the fiber/matrix interface can be influenced by moisture absorption[8] because resin is extremely easy to absorb water which leads to volume expansion, while carbon fiber is almost non-absorbent, so that the resin’s swelling generates stress and can cause interface debonding[9]. So the study on the interfacial bonding strength of NCF/BMI composites is also important.

In this study, to assess the effect of stitching threads in NCF, the differential scanning calorimetry(DSC) and thermogravimetry (TG) were measured. A approach was employed to assess the fibre/matrix adhesion between polymer threads and BMI resin by transverse fibre bundle (TFB) tests, which could measure interfacial bonding strength of the fibre/matrix interface in tensile specimens by applying normal stress until failure. In order to assess the effect of stitching threads on hygrothermal property, the interlaminar shear strength of NCF/bismaleimide(BMI) composites subjected to hygrothermal aging was tested under wet and dry condition. Electron microscopy was used to observe the microstructure of materials.

2. Materials and Experiments

2.1 Materials

Five types of stitching threads were studied in this paper, including 33 dtex PET, 83 dtex PET, 83 dtex meldable PET, 110dtex poly-p-phenylene terephthamide(PPTA) and 33 dtex polypropylene (PP). The term dtex is the mass in grams per 10000 meters.

The BMI resin 6421 used in this study was supplied by Beijing Aeronautical Manufacturing Technology Research Institute. The curing process is 150°C
/1h+160°C/1h+180°C/2h+220°C/2h, then cooling to room temperature.

2.2 Properties of Stitching threads

The heat resistance of these stitching threads were defined by means of DSC and TG (STA409 C/3/F), with the heating rate of 10°C/min, from 25°C to 800°C.

Based on GB/T 14344-2003, tensile properties of stitching threads were carried out using Instron IX4465 testing machine, with the loading rate of 20mm/min. The gage length was 250mm.

The fiber/matrix adhesion between stitching threads and bismaleimide resin (BMI6421) were estimated by transverse fiber bundle (TFB) tests[10-11]. Based on ASTM D638, tensile specimens of BMI resin 6421 containing stitching threads embedded in the middle were manufactured. The stitching threads were arranged in the transverse direction. Tensile properties of the samples were measured on Instron IX4465 testing machine. Electron microscopy was used to observe the microstructure of fracture surface.

2.3 Moisture resistance of NCF composites

Two kinds of unidirectional laminates and three kinds of quasi-isotropic laminates were manufactured by resin transfer molding(RTM) with BMI 6421 resin. And the reinforced fabrics were NCF-U1, NCF-B1 and unidirectional fibres. Unidirectional NCF laminates are made of uni-axial NCF(NCF-U1) and quadri-axial laminates are made of uni-axial NCF and biaxial NCF(NCF-B1), respectively. The layer sequence of unidirectional laminates were [0°]₁₁. The layer sequence of quasi-isotropic laminates were [(+45°/-45°)/(0°/90°)]₂₈.

The hydrothermal properties of unidirectional laminates and quasi-isotropic laminates were investigated. Moisture absorption and desorption curves of these laminates were obtained by weighting the specimens until their weight reached constant[12]. The percent moisture content \( M_i \) defined as

\[
M_i = \frac{m_i - m_0}{m_0} \times 100\%
\]

where \( m_i \) is the weight of moist material and \( m_0 \) is the weight of dry material.

In order to examine the relationship between test temperatures and the interlaminar shear strength of composites subjected to hygrothermal aging, several groups of specimens were put in the same hygrothermal environment for 7 days and 14days.

Based on JC/T 773-2010, interlaminar shear strength (ILSS) of the specimens above were carried out using Instron 5565 testing machine. The geometry of the samples is 20*10*2mm³. The failure mechanisms were discussed, and the fracture morphology of the composites were observed using CS3400 scanning electron microscopy (SEM).

3. Results and discussion

3.1 Properties of Stitching threads

To assess the effect of stitching threads, the DSC and TG were measured. Fig.1 shows the result. Table.1 shows the \( T_m \), \( T_d \), crystallinity of stitching threads. The three types of PET stitching threads have the same melting temperature(\( T_m \)), but were separated by the area of melting peaks, which were attributed to the differences of crystallinity.

The tensile strength of stitching threads can be improved, when increasing the crystallinity. Table.2 shows the tensile properties of stitching threads. It was found that the number of polymer tows embedded in the TFB test specimens does not show significant effects on their tensile strength. For example, the 33 dtex and 83 dtex PET have almost the same tensile strength, while the tensile strength of 83 meldable dtex PET is 439.14MPa. This indicates that, as long as there are threads that can provide sufficient interfacial debonding locations to cause the formation of the critical cracks, the failure stress of the TFB specimen is almost independent of the number of polymer threads incorporated. It also reveals that the elongation of PET stitching threads are 21.30%, 20.60% and 43.70%. In real stitching process, it requests that the elongation of stitching threads should be suitable with the resin, between 10% and 40%. Although the tensile strength of PPTA stitching threads is much higher than other threads, its elongation is too small, only 2.43%, which may induce fiber pick and break in real manufacturing process. As to PP stitching threads, which have good compatibility with BMI resin, were dissolved in it.

In order to further assess the adhesion between stitching threads and BMI resin, the tensile strength of TFB specimens was determined. Fig.2 shows the result. For the TFB specimens successfully tested in this study, nearly all of them failed in the middle where the stitching threads were located. Fig.3
shows the fracture surface of TFB specimens. It is seemed that the tensile strengths of specimens containing stitching threads is lower than the pure BMI resin specimen(65.6MPa), being about 73% of those of the pure resin specimen. The strength of matrix is much higher than the interfacial bonding strength between two dissimilar materials and fiber-matrix interfacial cracking always occurs first and results in failure of the specimen. Due to the higher strength than other specimens containing stitching threads, PP shows the best compatibility with BMI resin, which is in keep with the result of heat resistance studies[10]. Fig.4 shows the micro-structure of fracture surface of TFB specimens. For the TFB specimens with PPTA threads, the threads-matrix interfaces were relatively weak, as demonstrated by the SEM photographs in Fig. 4(d), where the smooth and clean surfaces of PPTA threads can be observed.

It is noted that, in the tensile testing of the TFB specimens, premature failures occur prior to reaching the failure load of the matrix, accompanied by a sudden drop in tensile stress. It can therefore be postulated that once the applied stress reaches the normal bonding strength of the stitching threads/BMI interface, tiny cracks form as a result of threads-matrix debonding at multiple locations and these cracks spontaneously coalesce into larger cracks due to large stress concentrations at crack fronts, triggering the tensile failure of the specimen. Consequently, the stress at the specimen failure would be equivalent to the bonding strength of the threads-matrix interface.

Above all, the 33 dtex and 83 dtex PET are more suitable to be used in stitching process.

3.2 Moisture resistance of NCF composites

3.2.1 Moisture absorption and desorption

Moisture absorption and desorption curves of unidirectional laminates and quasi-isotropic laminates were shown in Fig.5, Fig.6. The moisture absorption of NCF/BMI composites is a function of time. In Fig.5 and Fig.6, the solid lines were the Fick’s diffusion curves obtained by fitting the moisture absorption,

\[ M_s = M_o \left(1 - \exp \left[-7.3 \left(\frac{D\tau}{b^2}\right)^{0.75}\right]\right) \] (2)

where \(M_o\) is the saturation level of water absorption, \(D\) is the diffusion coefficient, \(b\) is the specimen thickness.

The water uptake is influenced by several parameters[14-15]: (1) the hydrophilic character of the matrix and fibers, (2) the adhesion between the fibers and the matrix, (3) the micro-cracks and voids in the material, (4) the “channels” in the fibrous plies. The resin polymer network controls the absorption of water, and in turn the absorption of water influences the network. The formation of more voids and micro-cracks, which is advantageous to water uptake, is induced by the absorption of water, accordingly the water absorption saturation level increases.

As shown in fig.5 and fig.6, no matter in unidirectional laminates or quasi-isotropic laminates, NCF/BMI composites have a higher rate of moisture absorption and moisture desorption than unidirectional fabric/BMI composites.

ILSS of unidirectional laminates and quasi-isotropic laminates in different hygrothermal processes were shown in Fig.7, Fig.8. “\(W\)” means moisture absorption, “\(D\)” means moisture desorption, “\(A_0\)” means blank sample. Such as, “7W” means 7days of moisture absorption. The short beam method is one of the simplest tests and is widely used for measuring the macroscopic interlaminar shear strength of composites (ILSS). The composites shear properties are mainly dominated by matrix and fiber/matrix interface[16]. So we can obtain information about changes of composites shear properties under wet conditions. In addition, the “channels” in the fibrous plies cause resin rich regions in the NCF/BMI composites, decreasing the fiber volume fractions, which will affect the mechanical performance and moisture resistance.

It is clear that ILSS of two kinds of unidirectional laminates in different hygrothermal processes are almost the same. But ILSS of quasi-isotropic composites with NCF reinforced is lower than unidirectional fabric reinforced, due to the existence of stitching threads. Combined with microstructures of the fracture in composites, it seems that stitching threads has a significant influence on ILSS of NCF/BMI composites in different hygrothermal processes, since it absorb water easier than carbon fiber, which may reduce the strength of materials[13].

The micro-structure of fracture surfaces of NCF/BMI laminates were shown in Fig.9, Fig.10. It is found that the surface of fibers is smooth with slight amount of resin adhered on the surface for
the wet specimens, and the dominating deformation mechanism is debonding. It suggests that the interfacial bond strengths between the fiber and the matrix are poor due to hygrothermal aging.

In addition, there is an obvious difference between dry specimens and wet specimens: matrix in wet specimens is in forms of granular or powdered due to matrix plasticization, while dry specimens exhibit shear-band. This result just proves that dry and wet specimens have different failure patterns and different ILSS values (as shown in Fig. 7, Fig. 8).

Conclusion

In real stitching process, it requests that the elongation of stitching threads should be suitable with the resin, between 10% and 40%. The 33 dtex and 83 dtex PET are more suitable to be used in stitching process.

No matter in unidirectional laminates or quasi-isotropic laminates, NCF/BMI composites have a higher rate of moisture absorption and moisture desorption than unidirectional fabric/BMI composites. The “channels” in the fibrous plies cause resin rich regions in the NCF/BMI composites, decreasing the fiber volume fractions, which will affect the mechanical performance and moisture resistance.

References

Table 1. $T_m$, $T_d$, crystallinity of five types of stitching threads

<table>
<thead>
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<tr>
<td></td>
<td>33 dtex PET</td>
<td>83 dtex PET</td>
<td>83 dtex meldable PET</td>
<td>33 dtex PP</td>
<td>110 dtex PPTA</td>
</tr>
<tr>
<td>$T_m$(℃)</td>
<td>256</td>
<td>256</td>
<td>255</td>
<td>115/255</td>
<td>436</td>
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<tr>
<td>$\Delta H_m$(J/g)</td>
<td>68.76</td>
<td>39.12</td>
<td>20.58</td>
<td>28.5</td>
<td>128.50</td>
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<tr>
<td>Crystallinity (%)</td>
<td>65.3</td>
<td>37.2</td>
<td>19.5</td>
<td>13.7</td>
<td>—</td>
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<tr>
<td>$T_d$(℃)</td>
<td>435</td>
<td>433</td>
<td>437</td>
<td>429</td>
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Table 2. Tensile properties of five types of stitching threads

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<td>33 dtex PP</td>
<td>110 dtex PPTA</td>
</tr>
<tr>
<td>Tensile Strength/MPa</td>
<td>571.12</td>
<td>586.22</td>
<td>439.14</td>
<td>427.56</td>
<td>2620.20</td>
</tr>
<tr>
<td>CV/%</td>
<td>6.78</td>
<td>6.51</td>
<td>7.35</td>
<td>6.35</td>
<td>6.45</td>
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<tr>
<td>Tensile Modulus/GPa</td>
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<td>3.48</td>
<td>0.73</td>
<td>1.02</td>
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<tr>
<td>CV/%</td>
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<td>3.34</td>
<td>9.28</td>
<td>3.61</td>
<td>0.95</td>
</tr>
<tr>
<td>Elongation /%</td>
<td>21.30</td>
<td>20.60</td>
<td>43.70</td>
<td>41.80</td>
<td>2.43</td>
</tr>
<tr>
<td>CV/%</td>
<td>6.87</td>
<td>5.37</td>
<td>7.56</td>
<td>2.17</td>
<td>6.24</td>
</tr>
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</table>
Fig.1. The DSC-TG curve of five types of stitching threads

(a) 33 dtex PET
(b) 83 dtex PET
(c) 83 dtex meldable PET
(d) 33 dtex PP
(e) 110 dtex PPTA
Fig. 2. The tensile properties of TFB specimens

Fig. 3. The fracture surface of TFB specimens

(a) Tensile Strength
(b) Tensile Modulus

(a) 33 dtex PET
(b) 83 dtex PET
Fig. 4. The micro-structure of fracture surface

(a) 83 dtex meldable PET

(b) 110 dtex PPTA

Fig. 5. Moisture absorption and desorption curves for two kinds of unidirectional laminates
Fig. 6. Moisture absorption and desorption curves for three kinds of quasi-isotropic laminates

Fig. 7. ILSS of two kinds of unidirectional composites in different hygrothermal processes

Fig. 8. ILSS of three kinds of quasi-isotropic composites in different hygrothermal processes
Fig. 9. The micro-structure of fracture surface of unidirectional laminates

(a) NCF-U1

(b) Fabric-UD1

Fig. 10. The micro-structure of fracture surface of quasi-isotropic laminates

(a) NCF-U1

(b) Fabric-UD1