Tensile Properties of a Novel Graphene Pattern Stitched Carbon/Epoxy 3D Composite

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Abstract The focus of this study is on the tensile properties of a novel graphene pattern stitched carbon/epoxy 3D composite which could be considered in aerospace industry applications. Tensile modulus and strength are reduced by 18-25% due to filament breakage during stitching; this reduction varies with stitching density. Fabrication induced voids and resin rich regions also contribute. However, extensive delamination observed in the tension loaded unstitched laminate is constrained by the graphene pattern stitching and the proposed 3D reinforced composite is expected to be more damage resistant under lateral impact loading.

Keywords — damage tolerance, stitching, tensile modulus, tensile strength, 3D composites.

I. INTRODUCTION

Carbon fiber-reinforced composites have been utilized extensively by the aerospace industry. Their high specific stiffness and strength, resistance to cracking by fatigue loading, corrosion resistance and ability to shape and tailor make them produce more aerodynamically efficient structural configurations [1, 2]. 2D laminated composites, offering high in-plane stiffness and strength, have been widely used while these structures are generally weak in the thickness direction (z-axis) [3].

Some detailed studies were performed by researchers about the tensile properties of stitched 3D composites [4-13]. Yudhanto et. al. (2015) investigated the effect of various stitch parameters on damage characteristics of stitched composites under tensile load. It was stated that the certain combination of stitch density and diameter resulted as an increased tensile strength by 7% compared to unstitched composites. This was due to the effectiveness of stitching on impeding the growth of edge-delamination [4]. Pingkarawat and Mouritz (2015) studied on the healing performance of stitched composites. Thermoplastic stitching improved the interlaminar fracture toughness and healing efficiency while the mechanical properties reduced due to the microstructural damages as ply crimping and fiber waviness [5]. Pappas et. al. (2018) stated that the delamination mechanisms in tufted composites are significantly affected by tuft geometry [6]. Ravandi et. al. (2016) investigated the effects of through-the-thickness stitching using natural fibers on the interlaminar fracture toughness and tensile properties of flax fiber/epoxy composites. It was concluded that the reduction in tensile properties of the composites due to imperfections caused by stitching was irrelevant from the material types of stitching yarn [7]. Effect of stitch density on tensile properties and damage mechanisms of Vectran® stitched carbon/epoxy composites was experimentally studied by Yudhanto et. al. (2013). It was found that low stitching density improved the tensile strength by 10.4%. The responsible of reduction in tensile stiffness was fiber misalignment in in-plane and out-of-plane directions. And also, the orthogonal binding effect of stitching architecture caused to decrease Poisson’s ratio of composites [8]. Barile et. al. (2017) stated that the through-the-thickness reinforcement together with polar fiber arrangement improved tensile strength by 15.6% and tensile modulus by 16.5% due to a better distribution of the load on the laminas [9]. Goktas et. al. (2017) stated that the manual-type stitching and high stitch densities offer a significant improvement of 74.5% on Mode-I interlaminar delamination [10]. Joshi et. al. (2017) stated that the reinforcement density is one of the key factors affecting strength, stiffness and crack propagation in
composite laminates. By suppressing the damage initiation, densely stitched laminates showed 15.2% higher in-plane stiffness than moderately stitched laminates [11]. Bilisik and Yolacan (2014) investigated the warp–weft directional and off-axis tensile properties of multistitched biaxial woven E-glass/polyester composites. It was stated that the stitching yarn type, the number of stitching directions, and the stitching density generally influenced the tensile properties of multistitched E-glass/polyester woven composites. Multistitching restricted the delamination in cross-sections of composites locally and the delamination did not propagate to the large areas [12, 13].

A stitched composite is among the impact tolerant design of stiffened composites and suitable for aircraft applications. Most investigators report a substantial deterioration of in-plane properties due to damage incurred as a result of stitching process depending on the type of laminate, the lamination technique, the stitching condition and the type of loading. However, any improvement in damage resistance and tolerance arising from stitching is more than compensate for a loss of in-plane strength [14, 15].

It can be seen from the literature that a lot of studies have been performed about the tensile properties of stitched 3D composites. In most of these studies, the influences of stitching parameters as stitching type (manual or machine), stitching density (dense or moderate), stitching direction (one, two or four directions) and stitching fiber types have been investigated. Stitching patterns are merely considered as using different stitching densities. However, the uniform distribution of z-fibers by stitching is critical for the performance of composites and constituted by stitching pattern. In addition, stitching is not only reinforcement to the out-of-direction of the composite but also the in-plane of the composite is reinforced by stitch loops which affect the damage formation under load. The novelty of this study is the used stitching pattern which is inspired by the unique molecular structure of graphene and called as ‘graphene pattern stitching’. Tensile properties of unstitched (US) and graphene pattern stitched (GPS) composites are compared in this study. The array of the pattern is arranged in parallel series with unit-cell walls meeting at exactly 120°. By using this stitching pattern, not only the uniform distribution of z-fibers is provided through-the-thickness, but also the in-plane of the composite is reinforced by stitch loops directed at 0° and ±60°.

II. METHODS AND PROCEDURES

A. Fabric and Stitching Yarn

Carbon woven fabric (Sigmatex, UK) sequenced as [(0°/90°)/(90°/0°)]3 was used to produce US and GPS composites. Twisted carbon yarn (175 turns/m) was used for stitching. The linear density of stitching yarn is 430 tex. The weave of carbon fabric is 5 harness satin. Tow sizes of carbon fabric are 12K on both warp and weft directions. The warp/weft fabric densities are 18 ends/10 cm and warp/weft crimp ratios are 1.0%. The fabric weight is 285 g/m² and the fabric thickness is 0.42 mm.

B. Graphene Pattern Stitching (GPS)

The unit-cell view of GPS is shown in Fig. 1. The schematic views of the followed path of stitching on both front and back faces of the structure and the distribution of z-fibers in through-the-thickness are given in Fig. 2. The position of ±60° stitch-loops is the front-face and the position of 0° stitch-loops is the back-face while the z-fibers are uniformly distributed through-the-thickness.

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**Fig. 1.** The unit-cell of GPS.
The preforms were stitched manually using a darning-needle (size: 14, diameter: 1.0 mm, length: 90 mm). GPS was implemented as loosely and densely by two different dimensions of unit-cell walls as 10 mm (L-GPS) and 5 mm (D-GPS).

C. Composite Manufacturing

The US and GPS composites produced with a VARTM (vacuum assisted resin transfer molding) method. The meshes and peel plies were used on both sides to prevent dry regions as shown in Fig. 3. Epoxy resin system (Araldite LY 564 resin/Aradur 2954 hardener) was used as the matrix. The infused composite samples were kept in an oven at 2h 80°C for curing and 8h 140°C for post-curing of epoxy. Manufacturing and testing of composites are performed at Northwest Composites Centre, University of Manchester, UK.

D. Measurement of Density and Fiber Fraction

The density of the samples was calculated by measuring the weight of the specimens in the air and in the distilled water at room temperature according to ASTM D792-13. The composite fiber fraction (volume based, %) was determined by ASTM D3171-15 using matrix burn off method. Specimens were firstly weighted and placed in a furnace at a temperature of 600°C for 1 hour to burn epoxy portion. Then, the samples were re-weighted to measure the mass and volume percentages of the remained carbon fiber content.

E. Tensile Test

Tensile tests were conducted according to ASTM D3039/D3039M-12 using an INSTRON 5982 test machine fitted with 100 kN load cell and hydraulic grips at 2 mm/min constant displacement. A video extensometer was used to measure the strain of the samples. Table I shows the tensile test parameters. The dimensions of the specimens were 25 mm (width) × 250 mm (length) with a 50 mm gauge length for both the bottom/top of the specimen without tabbing. The test machine and fixed test specimen between grips are shown in Fig. 4. The ultimate tensile strength values were calculated automatically by the Bluehill® software.

| TABLE I |
| TENSILE TEST PARAMETERS |

![Fig. 4. The test machine and fixed test specimen between grips.](image)
Sample width 25 mm  
Sample length 250 mm  
Load cell 100 kN  
Testing length 150 mm  
Testing speed 2.00 mm/min

III. RESULTS

A. Density and Fiber Fraction Results

Table II shows the density, fiber fraction, thickness, and void content results of US and GPS composites. The composite density decreased by GPS. This is due to the local gaps caused by the needle and resulted as resin rich regions. This was also the reason for the increased void contents from US to D-GPS. The positions of stitching loops on front/back faces were the responsible for increase in composite thickness.

|                  | Density (g/cm³) | Thickness (mm) | Fiber fraction (Vf) | Void content (%) |
|------------------|-----------------|----------------|---------------------|-----------------|
| US               | 1.522 (±0.004)  | 1.893 (±0.059) | 53.76 (±0.16)      | 0.53 (±0.10)   |
| L-GPS            | 1.493 (±0.004)  | 2.288 (±0.081) | 49.30 (±0.65)      | 0.71 (±0.00)   |
| D-GPS            | 1.470 (±0.006)  | 2.475 (±0.044) | 47.86 (±0.69)      | 1.48 (±0.19)   |

B. Tensile Test Results

The tensile test results US and GPS composites are presented in Table III. Fig. 5 shows the stress-strain curves of US and GPS composites during tensile loading.

|                  | Tensile strength (MPa) | Tensile modulus (GPa) | Tensile strain (%) |
|------------------|-------------------------|-----------------------|--------------------|
| US               | 703.50 (±91.32)         | 36.76 (±1.00)         | 1.29 (±0.11)       |
| L-GPS            | 526.50 (±82.03)         | 30.10 (±1.60)         | 1.20 (±0.18)       |
| D-GPS            | 498.50 (±49.51)         | 28.40 (±0.98)         | 1.38 (±0.16)       |

As seen in Fig. 5, GPS slightly increased the ductility of composites. The damaged region of US was higher than those of L-GPS and D-GPS composites. A large fiber/matrix delamination was observed in US composites. The delamination of the GPS composite structures after the tensile load locally occurred.
As seen in Table III and Fig. 6, the tensile strength of US was 703.50 MPa while the tensile strengths of L-GPS and D-GPS composites were 526.50 and 498.50 MPa, respectively. These results indicated that tensile strengths of the L-GPS and D-GPS composites decreased by 25% and 29% compared to US composites since dense stitching caused more filament breakages. The tensile modulus of US was 36.76 GPa while the tensile strengths of L-GPS and D-GPS composites were 30.10 and 28.40 MPa, respectively. These results indicated that tensile modulus of the L-GPS and D-GPS composites decreased 18% and 23% compared to US composites as dense stitching caused more filament breakages. Tensile strains of US and L-GPS composites were similar while the tensile strain of D-GPS composite was the highest due to denser out-of-plane and in-plane fiber reinforcement by stitching.

C. Tensile Failure Results

Fig. 7 shows the views of composite samples before and after tensile load. As seen in Fig. 7, the tensile failure of US composite was due to delamination across the sample width, multiple matrix cracks and fiber breakages. D-GPS well confined the tensile failure in a small region, while matrix cracks were not observed as in US. Subjecting the D-GPS composite to uniaxial tension caused multiple fiber breakages, fiber/matrix delamination and stitching yarn breakages.

Tensile failures of US and GPS composites were also examined by an optical microscope as seen in Fig. 8. The cross-sectional view of US after tensile load was observed as a complete delamination of fabric layers. The delamination of L-GPS and D-GPS was locally occurred. Dense stitching confined the delamination more effectively than loose stitching. More severe multiple fiber breakages were occurred in GPS composites since the stitching inhibited the spread of delamination along the sample.
Fig. 8. Optical microscope views of composites.

IV. CONCLUSION

Tensile strength and modulus of the D-GPS laminate decreased by 29% and 23% when compared to US composite, respectively. These results indicated that dense stitching caused more filament breakages, fiber distortions, and resin-rich-regions created by the piercing of stitching needle. The tensile strengths of US and L-GPS were similar. However, D-GPPS showed higher tensile strain than those of the US and L-GPS. Dense stitching increased the ductility and hence toughness of the composite. In addition, dense stitching well confined the tensile failure in a small region. Multiple matrix cracks were not observed as in the US plate. More severe multiple fiber breakages were occurred in GPS composites since the stitching inhibited the spread of delamination along the sample.

It is expected GPS to provide more damage resistance under drop weight impact conditions and since delamination is constrained, the compression after impact strength will be affected less and lead to a more damage tolerant structural configuration. However, further work is required to support these arguments.

ACKNOWLEDGMENT

The Scientific and Technological Research Council of Turkey (TUBITAK) is acknowledged for granting Gaye Kaya postdoctoral study in the framework of TUBITAK BIDEB-2219 International Postdoctoral Research Scholarship Program.

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