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Building Life-Cycle Enhancement Multifunctionality into Glass Fiber Reinforced Composite Laminates via Hierarchical Assemblies of Aligned Carbon Nanotubes

Palak B. Patel¹, Carolina Furtado², Jeonyoon Lee³, Megan F. Cooper⁴,
Luiz H. Acauan⁵

Massachusetts Institute of Technology, Cambridge, Massachusetts, 02139, USA

Stepan V. Lomov⁶, Iskander S. Akhatov⁷, Sergey G. Abaimov⁸
Skolkovo Institute of Science and Technology, Moscow, 121205, Russia

Brian L. Wardle⁹

Massachusetts Institute of Technology, Cambridge, Massachusetts, 02139, USA

For aerospace vehicles, where weight reduction is important, studies have been performed on composites to include different functionalities besides their primary structural function. Some of these functionalities include energy savings, self-health monitoring, ice protection system, and self-curing sensing capabilities, and have been demonstrated individually in carbon fiber reinforced polymer composites. Nano-engineering techniques enable integrating these functionalities in composite systems to add multifunctionalities with insignificant changes in dimension or weight of the composite system, while ensuring that the mechanical properties such as strength are maintained or enhanced. Here, glass fiber reinforced polymer composites are nanoengineered to add multiple multifunctionalities concurrently via hierarchical assemblies of vertically aligned carbon nanotubes. In this preliminary study, the nanoengineering of the composite suggests life-cycle enhancements via an increase in interlaminar shear strength.

¹ Graduate Researcher, Department of Mechanical Engineering, Massachusetts Institute of Technology

² Post-Doctoral Researcher, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology

³ Post-Doctoral Researcher, Department of Chemical Engineering, Massachusetts Institute of Technology

⁴ Undergraduate Researcher, Department of Material Science and Engineering, Massachusetts Institute of Technology

⁵ Research Scientist, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology

⁶ Contract Researcher, Center for Design, Manufacturing and Materials, Skolkovo Institute of Science and Technology

⁷ Professor, Center for Design, Manufacturing and Materials, Skolkovo Institute of Science and Technology

⁸ Assistant Professor, Center for Design, Manufacturing and Materials, Skolkovo Institute of Science and Technology

⁹ Professor, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology

I. Introduction

Combining one or more functional capabilities of subsystems within a structure can provide system-level savings, particularly for weight-critical applications such as aerospace vehicles. Therefore, numerous studies have been conducted on multifunctional composite structures that perform additional functions apart from the primary structural functions. For example, composite structures with life cycle enhancements [1-15], manufacturing energy savings [9-14], self-health monitoring, ice protection system [15], and self-cure sensing capability [11-13] have been demonstrated. While individual multifunctionalities have been demonstrated, integration of these multifunctionalities into a single engineering solution has not been undertaken.

Nanoengineering presents a significant opportunity for the integration of functions due to the possibility of embedding the required additional functionalities on the nanoscale without the necessity to modify shape, design, or load carrying capacity of the structure. The properties of the integrated nano-technology, such as significantly small size and insignificant weight, allows for an addition of multifunctionality with little or no increase in the dimensions or weight of composite structures. Here, an integrated-multifunctional nano-engineered system is implemented in glass fiber composite laminate structures. The system is designed to be capable of independent yet synergistic functionalities in life-cycle enhancements, energy savings during manufacturing, in-situ cure (manufacturing) monitoring, and in-service damage sensing. In this paper, various architectures of carbon nanotubes (CNTs) are used to nanoengineer composite laminates manufactured using Hexcel E-glass/913 unidirectional glass fiber prepreg to manufacture an integrated multifunctional composite (IMC) with a focus on demonstrating the life-cycle enhancement multifunctionality by increasing the quality and interlaminar shear strength of the composite.

II. IMC Manufacturing

The fabrication of the IMC has been described below. This includes the synthesis of the various carbon nanotube architectures that provide the integrated multifunctionalities, their integration into the prepreg-based laminate system, and curing of the composite.

A. Synthesis of Multifunctional CNT Architectures

Multiple architectures of carbon nanotubes have been integrated to provide different multifunctionalities to the composite system. Between the plies of the laminate, two different CNT architectures were studied by placing them in the interlaminar region of the composite in separate specimens. The CNT architecture, referred to as nanostitch 1.0 [1-6], consist of vertically aligned carbon nanotubes (VACNTs). The other CNT architecture, nanostitch 2.0 [7-8], is a novel architecture of buckled arrays of VACNTs. To synthesize the CNT architectures, a thermal catalytic chemical vapor deposition (CVD) process was utilized for growth on catalyzed silicon wafers inside a quartz tube furnace. The silicon wafer used to grow VACNTs for nanostitch 1.0 had uniform layers of iron and Al_2O_3 catalyst deposited on the surface, while the silicon wafers used to grow VACNTs for nanostitch 2.0 had a patterned array of catalyst, created by shadow masking, deposited on the surface. While nanostitch 1.0 integrates the 20-micron heighted VACNTs as grown in the interlaminar region, nanostitch 2.0 utilizes buckled 40-micron heighted VACNTs to form denser forests as seen in Figure 1. While nanostitch is integrated into the interlaminar region, randomly oriented commercial CNT film (Tortech, CNTM4) is placed and integrated into the top and bottom of the laminate. The addition of these CNT architectures to the composite system are the key to the integration of multifunctionalities in composites [16].

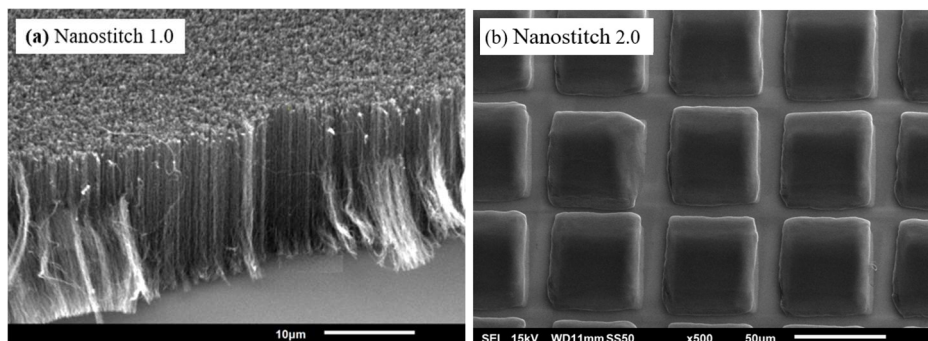


Fig. 1 Scanning electron micrographs of the CNT architectures placed in the interlaminar region of the IMCs: a) nanostitch 1.0 VACNTs, b) nanostitch 2.0 buckled VACNTs

B. Nanoengineered Glass Fiber Laminate

The CNT architectures were integrated into Hexcel NVE 913 E-glass unidirectional autoclave-grade prepreg laminates. The manufacturing of the IMC consisted of a 16-ply quasi-isotropic lay-up with multifunctional CNT architectures integrated in the interlaminar regions between each ply and the CNT film integrated on the top and bottom of the laminate as seen in Figure 2. Two 150 mm x 150 mm laminate plates, a baseline and the IMC, were manufactured in a $[0/90/45/-45]_{2s}$ quasi-isotropic configuration. The baseline plate was manufactured with the glass fiber reinforced polymer with no CNT integration. The IMC plate had three specimen regions, as seen in Figure 2, with various CNT architectures as listed in Table 1. After the integration of the interlaminar CNTs, the both laminates were cured in an autoclave following the manufacturer recommended cure cycle. The cure cycle began under vacuum at 0.7 bar (20 in. Hg) as the pressure rose to 6-7 bar (85-100 psig) which was then held throughout the cure cycle. The vacuum was vented when the pressure reached 1.4 bar (20 psig). The temperature is increased at a steady rate of not more than 5°C/min (9 °F/min) till 90°C (195°F), held at 90°C (195°F) for 30 minutes, increased at a steady rate of not more than 5°C/min (9 °F/min) till 120-130°C (250-265°F), held at 120-130°C (250-265°F) for 60 minutes, and then decreased at $\leq 3^\circ\text{C}/\text{min}$ (5°F/min). The pressure is released to 0 bar (0 psig) when the temperature reaches 65°C (150°F).

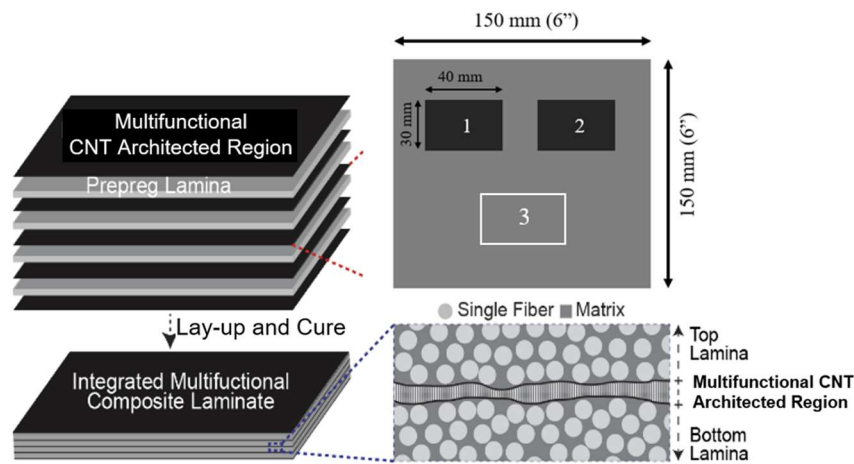


Fig. 2 Integrated multifunctional composite manufacturing with various CNT architectures placed between prepreg lamina

Table 1 Multifunctional CNT architectures in plates and specimens

Plate Name	Specimen name	Multifunctional nanostructured reinforcement architecture in the interlaminar region	Multifunctional nanostructured reinforcement architecture: CNT height (μm)	Multifunctional nanostructured layer on the top and bottom of laminate
Baseline Plate	Baseline Specimen	None	-	None
CNT Plate	CNT Specimen 1	Nanostitch 1.0	20	CNT commercial film
	CNT Specimen 2	Nanostitch 2.0	40	CNT commercial film
	CNT Baseline Specimen 3	None	-	CNT commercial film

III. Experimental Results

The integration of the various multifunctional CNT architectures into the IMC produced void-free laminates with better quality than the baseline manufactured in the same autoclave cure cycle, which had also been proven in a preliminary study of the same composite system [16]. In this study, the baseline plate was observed to have voids trapped in the laminate while the IMC CNT plate had no void formations in the laminate. As is common with prepreg-based composite laminates, the interlaminar region between plies is the Achilles' heel of the laminate as it is a resin-rich area which fails when the composite undergoes shear. With the integration of nanostitch 1.0 and nanostitch 2.0 in the interlaminar regions of the laminate, the interlaminar region is reinforced which enhances the interlaminar strength of the laminate. The interlaminar shear strength of the IMC specimens have been studied by performing short beam shear tests on the composite.

A. Short Beam Shear Test

Short beam shear (SBS) testing was performed on IMC specimens according to ASTM standard D2344 [17] for testing the short beam shear strength. SBS specimens from each specimen region of the IMC were made of dimensions 12.6 mm x 4.2 mm x 2.1 mm (length x width x thickness) and prepared by polishing to remove stress concentrations and notches in the surfaces that could act as damage initiators, enabling premature failure of the specimens. The test was performed on a Zwick/Roell Z010 mechanical testing machine with a three-point bend fixture with a 1mm/min crosshead movement rate. Figure 3 compares the short beam shear strength values of the IMC CNT plate specimens 1 and 2 with the baseline plate specimen and the IMC CNT plate baseline specimen 3. This comparison highlights the effect of the CNTs in the interlaminar region. The baseline plate, which had no CNT architecture reinforcement, had voids in the laminate and had a short beam shear strength of 81.56 ± 1.60 MPa (mean and standard error). The CNT baseline specimen 3 which has unreinforced interlaminar regions and CNT commercial film on the top and bottom of the laminate with no voids has a short beam shear strength of 84.82 ± 1.66 MPa and is considered, here, the most proper baseline. CNT specimen 1, reinforced with nanostitch 1.0 in the interlaminar region and with commercial CNT film on the top and bottom of the laminate, showed a statistically significant increase in short beam shear strength (89.53 ± 0.30 MPa) by 5.55% compared to the CNT baseline specimen and a statistically significant increase of short beam shear strength by 7.97% compared to the baseline specimen. CNT specimen 2, reinforced with nanostitch 2.0 in the interlaminar region and with commercial CNT film on the top and bottom of the laminate, showed a statistically insignificant increase in short beam shear strength (86.63 ± 1.70 MPa) by 2.06% compared to the CNT baseline specimen and a statistically significant increase of short beam shear strength by 6.22% compared to the baseline specimen. The highest short beam shear strength was observed in IMC CNT specimen 1 with nanostitch 1.0 interlaminar reinforcements which is also seen in the preliminary study performed on this composite system [16]. The nanostitch CNT interlaminar reinforcement strengthens the weakest part of the composite hence strengthening the composite and enhancing the life cycle of the composite.

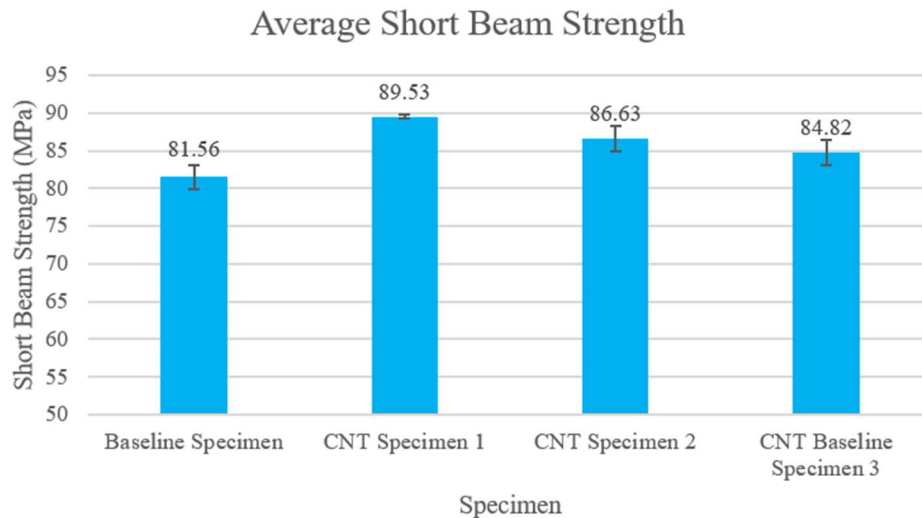


Figure 3 Average short beam shear strength of specimens with standard error

IV. Conclusion

The life-cycle enhancement of a glass fiber prepreg-based composite system was explored by the integration of various CNT multifunctional architectures in the interlaminar region as well as the top and bottom of the composite laminate. The study of two different CNT architectures, namely nanostitch 1.0 and 2.0, for interlaminar reinforcement was conducted and it was concluded that nanostitch 1.0, comprising of VACNTs, increased the short beam shear strength of the composite by a statistically significant 5.55%. While the nanoengineering of the composite aims to provide multiple multifunctionalities to the composite system, this study proved the life-cycle enhancement multifunctionality while maintaining the primary structural function. Having concluded the study on enhancing the life-cycle of the integrated multifunctional composite, the next multifunctionalities will be explored in the future.

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References

- [1] De Villoria, R.G., Hallander, P., Ydrefors, L., Nordin, P. and Wardle, B.L., 2016. In-plane strength enhancement of laminated composites via aligned carbon nanotube interlaminar reinforcement. *Composites Science and Technology*, 133, pp.33-39.
- [2] Kalfon-Cohen, E., Kopp, R., Furtado, C., Ni, X., Arteiro, A., Borstnar, G., Mavrogordato, M.N., Sinclair, I., Spearing, S.M., Camanho, P.P. and Wardle, B.L., 2018. Synergetic effects of thin plies and aligned carbon nanotube interlaminar reinforcement in composite laminates. *Composites Science and Technology*, 166, pp.160-168.
- [3] Ni, X. and Wardle, B.L., 2019. Experimental investigation of interlaminar fracture micro-mechanisms of aligned carbon nanotube-reinforced aerospace laminated composites. In *AIAA Scitech 2019 Forum* (p. 1201).
- [4] Ni, X., Furtado, C., Fritz, N.K., Kopp, R., Camanho, P.P. and Wardle, B.L., 2020. Interlaminar to intralaminar mode I and II crack bifurcation due to aligned carbon nanotube reinforcement of aerospace-grade advanced composites. *Composites Science and Technology*, 190, p.108014.
- [5] Ni, X., Kalfon-Cohen, E., Furtado, C., Kopp, R., Fritz, N.K., Arteiro, A., Valdes, G., Hank, T., Borstnar, G., Mavrogordato, M. and Spearing, S.M., 2017, August. Interlaminar reinforcement of carbon fiber composites using aligned carbon nanotubes. In *21st Int. Conf. Compos. Mater* (pp. 40-43).
- [6] Ni, X., Furtado, C., Kalfon-Cohen, E., Zhou, Y., Valdes, G.A., Hank, T.J., Camanho, P.P. and Wardle, B.L., 2019. Static and fatigue interlaminar shear reinforcement in aligned carbon nanotube-reinforced hierarchical advanced composites. *Composites Part A: Applied Science and Manufacturing*, 120, pp.106-115.
- [7] Ni, X., Acauan, L.H. and Wardle, B.L., 2020. Coherent nanofiber array buckling-enabled synthesis of hierarchical layered composites with enhanced strength. *Extreme Mechanics Letters*, 39, p.100773.
- [8] Ni, X. and Wardle, B.L., 2020. Aerospace-grade Advanced Composites with Buckling-densified Aligned Carbon Nanotubes Interlaminar Reinforcement. In *AIAA Scitech 2020 Forum* (p. 0156).
- [9] Lee, J., Stein, I.Y., Kessler, S.S. and Wardle, B.L., 2015. Aligned carbon nanotube film enables thermally induced state transformations in layered polymeric materials. *ACS applied materials & interfaces*, 7(16), pp.8900-8905.
- [10] Lee, J., Ni, X., Daso, F., Xiao, X., King, D., Gómez, J.S., Varela, T.B., Kessler, S.S. and Wardle, B.L., 2018. Advanced carbon fiber composite out-of-autoclave laminate manufacture via nanostructured out-of-oven conductive curing. *Composites Science and Technology*, 166, pp.150-159.
- [11] Lee, J., Daso, F., Kessler, S.S. and Wardle, B.L., Carbon Fiber Prepreg Composite Laminates Cured via Conductive Curing Using Nanoengineered Nanocomposite Heaters.
- [12] Lee, J. and Wardle, B.L., 2019. Nanoengineered In Situ Cure Status Monitoring Technique Based on Carbon Nanotube Network. In *AIAA Scitech 2019 Forum* (p. 1199).
- [13] Lee, J., Stein, I.Y., Antunes, E.F., Kessler, S.S. and Wardle, B.L., 2015. Out-of-oven curing of polymeric composites via resistive microheaters comprised of aligned carbon nanotube networks.
- [14] Lee, J., Kessler, S.S. and Wardle, B.L., 2020. Void-Free Layered Polymeric Architectures via Capillary-Action of Nanoporous Films. *Advanced Materials Interfaces*, 7(4), p.1901427.
- [15] Lee, J., Brampton, C.J., Bowen, C.R., Wardle, B.L. and Kim, H.A., 2015. Investigation of Aligned Conductive Polymer Nanocomposites for Actuation of Bistable Laminates. In *23rd AIAA/AHS Adaptive Structures Conference* (p. 1725).
- [16] Patel, P., Furtado, C.F., Cooper, M., Acauan, L., Lomov, S., Akhatov, I., Abaimov, S., Lee, J. and Wardle, B., 2021. Nanoengineered Glass Fiber Reinforced Composite Laminates with Integrated Multifunctionality. In *Proceedings of the American Society for Composites—Thirty-Sixth Technical Conference on Composite Materials*.
- [17] STM D 2344 / D 2344M-16, “Standard Test Method for Short Beam Strength of Polymer Matrix Composite Materials and Their Laminates,” West Conshohocken, PA: ASTM International