ECO-FRIENDLY COMPOSITES: FABRICATION AND CHARACTERIZATION OF POLYMER COMPOSITES MADE FROM BIO-BASED CURING AGENT

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ABSTRACT

Thermoset polymer composites, known for their outstanding thermal, mechanical, and chemical properties, have found applications in diverse fields, including aerospace and automotive industries. These polymers, once cured, cannot be recycled, making the end-of-life management of these composites very difficult and posing an environmental challenge. Conventional recycling methods are unsuitable for thermosets, forcing their accumulation in landfills and raising environmental concerns. One possible solution to overcome this concern is to use resins or curing agents, or both, made from biodegradable materials. This study explores the fabrication and characterization of polymer composites using a commercially available green curing agent made from biomass. The composite laminates were fabricated using HVARTM (Heated Vacuum Assisted Resin Transfer Molding) process. In this process, heat pads are used to increase the temperature of both the epoxy resin and the plain weave carbon fiber laminate to a desired temperature, providing ease of flow to the resin. Small coupons were cut from the laminate using a water jet machine to study the flexural behavior of the composite in accordance with ASTM testing standards and compared with composite coupons fabricated using conventional epoxy resin.

Keywords: Mechanical Properties, Flexural Properties, Bio-Binder, Sustainability
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1. INTRODUCTION

Composite materials are engineered or naturally occurring materials made up of two or more constituent materials with different physical and chemical properties. Each component in a composite material brings its own unique properties, and when merged, the material produced has properties that are lighter, stronger, and more versatile than the parent materials. High strength-to-weight ratio, corrosion resistance, and tailor-ability are some of the many advantages of composite materials. These factors make them prime materials for various applications in industries such as aerospace, automotive, sports equipment, construction, as well as other composite industries. Different types of composites include fiber-reinforced composites, particle-reinforced composites, and laminar composites. Fiber-reinforced composites [1] are widely used types of composites. Commonly used fiber reinforcements include carbon, aramid, and glass fibers. The continued development and application of composites are crucial in various industries where performance and efficiency are of high importance.

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Generally, composite materials are made from different epoxy resin and hardeners or curing agents. These resins and hardeners are products of the chemical industry that are toxic and make use of non-renewable energy resources. Recent environmental challenges have raised toxicity and sustainability issues, compelling the search for alternative solutions that can meet these challenges. The composite market has recently seen the introduction of green epoxy resin made from biodegradable materials but faces challenges such as the availability of paired curing agents, cost, and mechanical performance. Another possible solution to cope with this challenge is using bio-degradable materials for the production of curing agents. Past investigations [2-4] have already shown the possibility of curing epoxy resins with bio-based curing agents. Additionally, the use of bio-based curing agents [3] can fulfill the performance requirements like mechanical, thermal, as well as chemical properties like those using traditional curing agents.

2. FABRICATION

2.1. Fabrication Methodology

In this study, to address the issue of epoxy resin's low viscosity at room temperature, which can lead to the production of substandard panels, a Heated Vacuum Assisted Resin Transfer Molding (HVARTM) [5] process was employed. The process involved preheating the entire setup, including the mold, fabric, peel ply, and plastic bag, to 120°F. This temperature elevation was critical to attaining the resin's optimal viscosity and flow. Temperature monitoring was crucial, with thermocouples placed at strategic points: one atop the insulating material and another at the mold's base. The goal was to minimize the temperature gradient from the mold's bottom to the topmost layer of the outer bag, ensuring even heating for uniform resin coating and distribution.

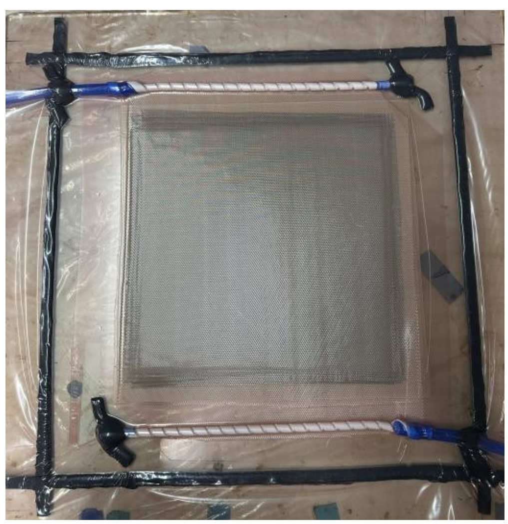


Fig 1: Panel Fabrication

A 24 inches × 24 inches silicone rubber laminated heating pad from Omega was utilized in the HVARTM process. The vacuum bag in this study was slightly larger than the laminate, specifically 7 inches longer and 2 inches wider. Inside this bag, laminate was placed. For the mold, a 24 inches × 24 inches glass sheet, 0.5 inches thick, was used. To create a vacuum-tight seal, sealant tape was applied to the glass. A mold release agent, Henkel-Frekote 770-NC, was then evenly applied within the sealed area on the glass using a paper towel, allowing it to dry. This agent prevents epoxy or plastic from adhering to the glass, facilitating the removal of the composite and vacuum bag after fabrication. A plastic film was laid on the mold within the sealed area to further protect the mold surface or glass from epoxy exposure, except around the edges between the plastic and the sealant tape. The resin distribution system consisted of a nylon mesh, which served as a spacer between the plastic and the Teflon peel ply layers on both the top and bottom. This setup allowed for even resin distribution as it flowed through the mesh. Teflon release fabric was placed between the distribution medium and the fabric, optimizing the resin flow and distribution throughout the fabrication process as shown in Figure 1.

The composite panel with dimensions of 12 inches x 12 inches was fabricated, consisting of twelve layers of plain weave carbon fibers (CF). The construction of the panel utilized a precise bagging process. This process required the strategic placement of inlet and outlet ports along the seal's perimeter, which were integral for resin infusion and vacuum establishment within the mold inside the bag. Attached to these ports were the resin and vacuum distribution lines, composed of silicone spiral-cut tubing. The length of this tubing was tailored to match the top side width of the vacuum bag. These distribution lines were strategically positioned above the distribution media, running along two sides of the fabric lay-up, and confined within the limits of the bag. The resin line was securely sealed at one end and connected to a resin supply at the other. To achieve an airtight seal, sealant tape was meticulously wrapped around the silicone tube, stopping approximately an inch from the end. This wrapping included the spiral-cut tube, with the tape overlapping and self-sealing, thereby forming an effective sealed system. Such attention to detail in the setup was critical for the successful infusion of resin and the maintenance of a consistent vacuum throughout the fabrication [5-7] process.

2.2. Panel Fabrication

Two distinct 12 inches x 12 inches composite panels, both constructed from plain weave carbon fiber, were fabricated. The first panel used a curing process using Epon-862 resin combined with Epikure-W hardener. The second panel was similarly cured but utilized Epon-828 resin and a commercially available bio-based curing agent. For each panel, the resin-to-hardener mixing ratios were precisely followed as per the manufacturer's [8] specifications by weight, 100:26.4 for the Epon-862/Epikure-W combination and 100:55 for the Epon-828/bio-curing agent mix. The curing cycle for both the panels was identical and both the panels were cured at a temperature of 300°F. This approach ensured consistency in the curing process, allowing for an accurate comparison between the two different resin-hardener systems.

3. CHARACTERIZATION

3.1. Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials ASTM D3039

The fabricated composite panels were precisely processed into tensile test coupons, adhering to the specifications outlined in ASTM D3039 [9]. This was accomplished using a water jet cutting machine, ensuring accuracy and consistency in the dimensions of the samples. Subsequently, each coupon was mounted with strain gauges in preparation for mechanical testing. The testing followed the ASTM D3039 standard and was conducted using an Instron electromechanical testing system.

The strain rate for the tests was set at 2 mm/min, aligning with standard testing protocols and ensuring reliable and comparable results across all samples.

(a) (b) (c)

Fig 2: (a)Sample Preparation, (b) Tensile Strength Test setup, (c) Sample failure after test

3.2. Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials ASTM D790

Flexural properties are essential in determining the material behavior under bending stresses. The fabricated composite panels were cut into coupons for the three-point bending test. The dimensions for the test coupons were according to ASTM D790 [10] standard. This cutting was executed using a water jet machine to ensure precision and uniformity in the coupon dimensions. In alignment with the testing standard, a span-to-thickness ratio of 16:1 was meticulously maintained. For both sample types, the width was 0.51 inches. However, a slight variation was observed in their thickness. Epon 862/Epikure-W samples were 0.17 inches, whereas the Epon 828/bio-curing agent samples were marginally thinner at 0.158 inches. To assess their flexural strength, six specimens from each sample category were subjected to the flexural test. The test setup design is shown in Figure 3. The rate of crosshead motion was set to 0.072 in/min for the Epon862/Epikure-W sample and 0.067 in/min for the Epon828/bio-curing agent mix sample.

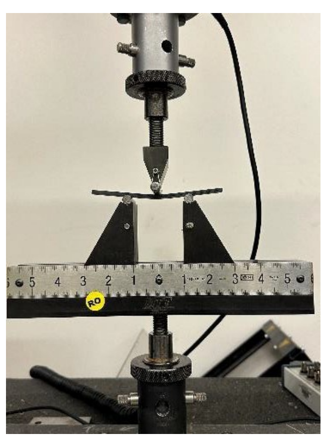
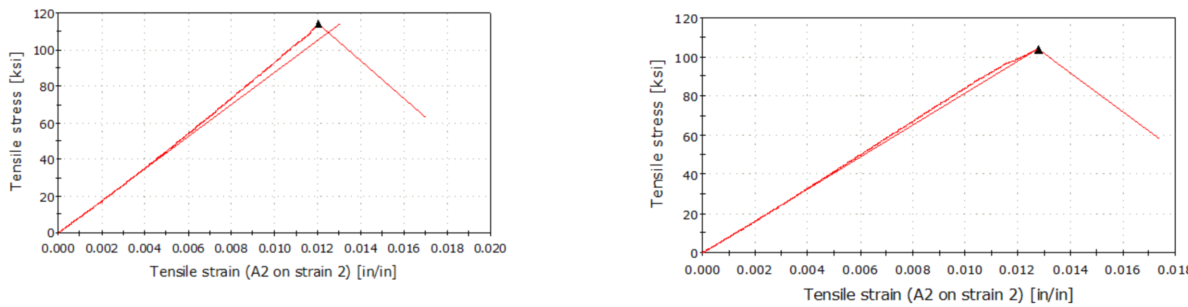


Fig 3: Test Setup

4. RESULTS & DISCUSSIONS

Figure 4 shows the stress-strain curve, while Table 1 shows the strength, modulus of elasticity, and strain at failure for both types of coupons. Analyzing both the stress-strain curve and the data presented in the results table, it becomes evident that the tensile properties of the materials tested exhibit a high degree of similarity.



(a) (b)

Fig 4: Tensile Stress-Strain curve of coupons produced using (a) Epon 862/Epikure-W, (b) Epon 828/Bio-Curing Agent

This comparability in tensile characteristics is clearly reflected in the consistent patterns observed in the stress-strain relationships and the corroborating numerical data. The tensile strength and elastic modulus of the test coupons are closely matched, showcasing comparable mechanical properties. Specifically, the coupon treated with Epon-862/Epikure-W exhibited a tensile strength of 114.07 ksi and an elastic modulus of 8.79 Msi. In comparison, the coupon processed with Epon-828/Bio-curing agent demonstrated a slightly lower tensile strength of 104.12 ksi and an elastic modulus of 8.20 Msi, as detailed in Table 1.

Table 1: Tensile Properties of coupons

Coupon Material	Tensile Strength (ksi)	Tensile Modulus of Elasticity (Msi)	Tensile Strain at failure (%)
Epon-862 and Epikure-W	114.07	8.79	1.2
Epon-828 and Bio-curing agent	104.12	8.20	1.36

Figure 5 shows Flexural Stress vs Flexural Strain as well as the Load Vs Extension curve. From the Load vs Extension curve, it can be observed that the load bearing of both the samples are very much comparable.

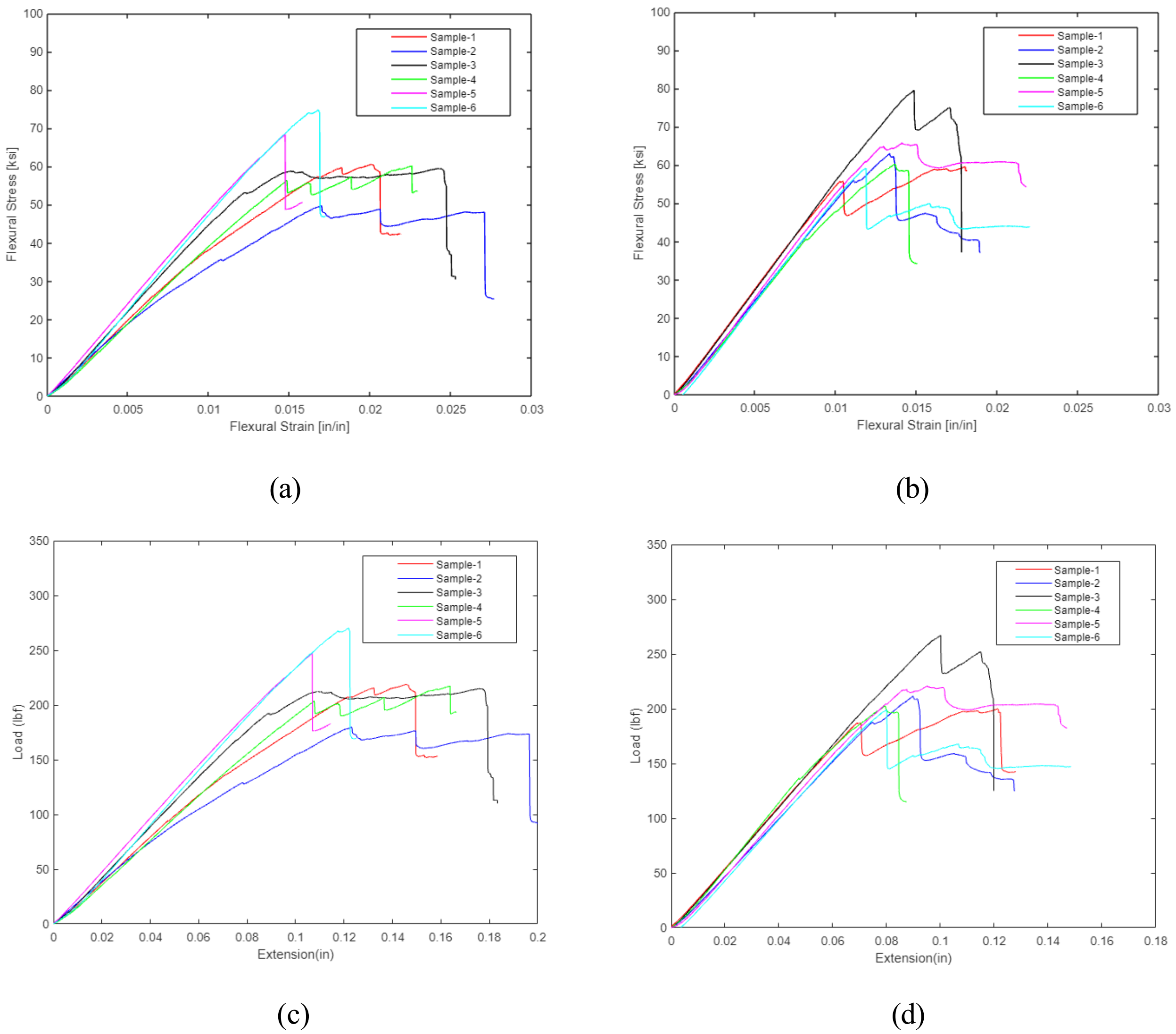


Fig 5: Flexural Stress-Strain curve of coupon made from (a) Epon 862/EpikureW, (b) Epon 828/Bio-curing agent; Load vs Extension curve of coupon made from (c) Epon 862/EpikureW, (d) Epon 828/Bio-curing agent

The maximum flexural stress and flexural modulus of elasticity are calculated using the following equations [14]:

$$\sigma_f = \frac{3PL}{2bd^2} \qquad\qquad \epsilon_f = \frac{6Dd}{L^2} \qquad\qquad \text{……………………(1)}$$

where,

P = load at a given point [lbf],

L = Support span [in.],

b = Width of beam tested [in.],

d = Depth of beam tested [in.]

D = Maximum deflection of the center of the beam [in.]

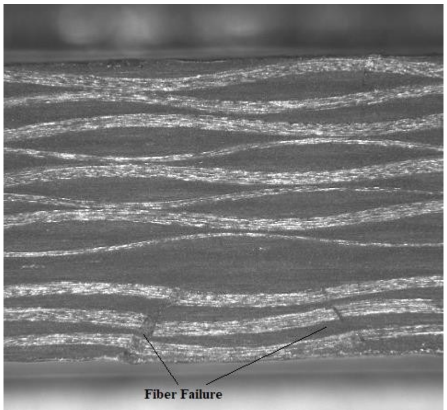
Table 2 shows the flexural strengths of six samples each cured with conventional and bio-based curing agents.

Table 2: Flexural properties stiffness and strength comparison

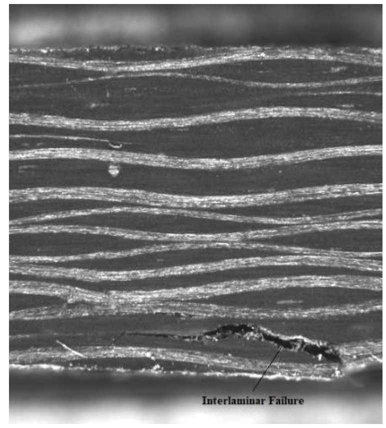
Coupon Material	Epon-828 and Bio-curing agent		Epon-862 and Epikure-W	
Sample	**Flexural Strength(ksi)**	**Flexural Modulus(Msi)**	**Flexural Strength(ksi)**	**Flexural Modulus(Msi)**
1	59.58	5.67	60.53	4.24
2	63	5.23	49.76	3.77
3	79.52	5.69	59.48	4.72
4	60.25	5.25	60.14	4.05
5	65.8	5.61	68.37	4.97
6	59.16	5.55	74.78	5.03
Mean	64.55	5.50	62.18	4.46
Std. Deviation	7.75	0.21	8.55	0.52
C.O.V.	12.01	3.77	13.75	11.62

Table 2 clearly indicates that flexural strength and stiffness of both the laminates (Epon-828/Bio-Curing agent and Epon-862/Epikure-W) are comparable. According to ASTM D790, a set of at least six specimens is necessary for the three-point bending test to assess flexural properties. When analyzed statistically, the flexural strength of the specimens created using Epon-828/Bio-curing agent was determined to be 64.55 ± 7.75 ksi with a flexural modulus of 5.50 ± 0.21 Msi. Conversely, specimens made with Epon-828/Epikure-W showed a slightly lower flexural strength of 62.18 ± 8.55 ksi and a flexural modulus of 4.46 ± 0.52 Msi. The results indicate that the flexural strengths of the two types of specimens are quite comparable.

While performing flexural tests on conventional epoxy resin samples and biobinder based samples, there was an interesting observation. It was observed that the failure mechanism in two different systems (conventional curing agent and bio-based curing agent) was entirely different. Figure 6 shows the image of samples cured with Epikure-W and Bio-based curing agent after performing the flexural test. It was observed that the samples cured with bio-based curing agent consistently showed interlaminar failure.



(a)



(b)

Fig 6: (a) Epon862/Epikure-W sample having fiber failure; (b) Epon828/Bio-based curing agent showing interlaminar failure.

Since interlaminar failure occurs due to debonding between the layers of material and can considerably reduce the interlinking strength between the laminates. The present research clearly indicates that additional investigation is warranted to understand behavior of coupons fabricated using bio-based curing agent using ASTM D2344 short beam shear strength standard. Furthermore, if samples fabricated using bio-based curing agent shows consistent interlaminar failure, then before this bio-based system is adopted in real life application, it may be necessary to improve the interlaminar strength by using various techniques such as surface treatments, employing through-thickness reinforcements or z-pinning or utilizing nanofibers place in the interlaminar region [11-13].

5. CONCLUSIONS

This paper investigated the tensile and flexural properties of composite coupons using conventional curing agent and bio-based curing agent. It was seen that the coupons cured with bio-curing agent have both tensile and flexural properties comparable to those cured using conventional curing agent. Also, the failure under flexural loading for the laminates fabricated using the bio-based curing agent was entirely different as compared to the conventional laminates that were manufactured using Epikure-W curing agent. The major difference was consistent interlaminar failure in laminates which were fabricated using bio-based curing agent. The present investigation shows that bio-curing agents have a potential for achieving the sustainability goal of using green composites for various applications.

6. ACKNOWLEDGEMENTS

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7. REFERENCES

1) *Polymers* 2023, *15*(2), 385; DOI: https://doi.org/10.3390/polym15020385

2) Dayo, A. Q., Wang, J., Wang, A. R., Lv, D., Zegaoui, A., Derradji, M., & Liu, W. B. (2018). Mechanical and thermal properties of a room temperature curing epoxy resin and related hemp fibers reinforced composites using a novel in-situ generated curing agent. *Materials Chemistry and Physics*, *203*, 293-301. DOI: https://doi.org/10.1016/j.matchemphys.2017.10.004
3) Baroncini, E. A., Kumar Yadav, S., Palmese, G. R., & Stanzione III, J. F. (2016). Recent advances in bio-based epoxy resins and bio-based epoxy curing agents. *Journal of Applied Polymer Science*, *133*(45). DOI: https://doi.org/10.1002/app.44103
4) Makh, N.S., Zhang, L. and Kelkar A.D. (2023). Replacement of epoxy curing agent with bio-binder 1,4-Diaminobutane-A green solution. SAMPE, Seattle, WA, United States. https://doi.org/10.33599/nasampe/s.23.0175
5) Jadhav, V. S., & Kelkar, A. D. (2019). Effect of curing temperature on the fundamental properties of laminated composites fabricated using plain weave and non-crimp fiber and epoxy resin. CAMX 2019, 23-26 Sept.
6) D. K. Shinde and A. D. Kelkar, "Effect of TEOS Electrospun Nanofiber Modified Resin on Interlaminar Shear Strength of Glass Fiber / Epoxy Composite," *Int. J. Chem. Mol. Nucl. Metallurgical Eng.*, vol. 8, no. 1, pp. 54–60, 2014.
7) A. D. Kelkar, J. S. Tate, and R. Bolick, "Introduction to Low-Cost Manufacturing of Composite Lamina," Proc. 2003 Am. Soc. Eng. Educ. Annu. Conf. Expo., p. 1482, 2003.
8) Resolution Performance Products, Product bulletin EPIKOTE™ Resin 862/EPIKURE™ Curing Agent W System.
9) ASTM Standard D3039-17, 2017, "Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials" ASTM International, West Conshohocken, PA, 2017. DOI: 10.1520/D3039_D3039M-17
10) ASTM Standard D790-17, 2017, "Standard Test Method for Flexural properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials" ASTM International, West Conshohocken, PA, 2017. DOI: 10.1520/D0790-17
11) Islam, A. I., & Kelkar, A. D. (2017). Prospects and challenges of nanomaterial engineered prepregs for improving interlaminar properties of laminated composites—a review. *MRS Communications*, *7*(2), 102-108.
12) Storck, S., Malecki, H., Shah, T., & Zupan, M. (2011). Improvements in interlaminar strength: a carbon nanotube approach. *Composites Part B: Engineering*, *42*(6), 1508-1516.
13) Kelkar, A. D., Tate, J. S., & Bolick, R. (2006). Structural integrity of aerospace textile composites under fatigue loading. *Materials Science and Engineering: B*, *132*(1-2), 79-84.
14) Shinde, D. K., White, F. T., & Kelkar, A. D. (2014, November). Flexural behavior of fiberglass polymer composite with and without TEOS electrospun nanofibers. *In ASME international mechanical engineering congress and exposition* (Vol. 46637, p. V014T11A037). American Society of Mechanical Engineers. DOI: https://doi.org/10.1115/IMECE2014-38304