Graphene Oxide and Natural Silk as Reinforcements for Epoxy Resin Matrix: Fabrication, Sample Preparation, and Testing

K N Sanjeev Kumar[1], Sanjeev Sharma[2], Dr. Abdel-Hamid I. Mourad[3], PB Sharma[4]

[1]PhD Student, ASET, Amity University, Gurgaon, India
[2]Professor, Head of Mechanical Engineering, ASET, Amity University, Gurgaon, India
[3]Professor, Mechanical Engineering Department, College of Engineering, United Arab Emirates University, UAE
[4]Vice Chancellor, Amity University, Gurgaon, India

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Abstract - The integration of graphene oxide (GO) and natural silk into epoxy matrices offers a promising approach to developing high-performance composites for structural applications. This study examines the fabrication processes, focusing on sample preparation and testing of GO and natural silk-reinforced epoxy composites. A comprehensive literature review spanning 1990 to 2024 provides insights into the necessity of Vacuum-Assisted Resin Transfer Molding (VARTM) at 30 PSI, the role of ultrasonication in achieving homogeneous GO dispersion, strategies to prevent agglomeration, optimal reinforcement proportions, and challenges encountered during hybridization. Key findings, including the impact of high silk volume fractions (60–70%) on mechanical properties, are incorporated, offering a comprehensive guide for composite fabrication. The findings underscore the critical parameters for successful composite fabrication and performance enhancement.

***Key Words*:** Graphene Oxide, Natural Silk, Epoxy Composites, VARTM, Ultrasonication, Hybrid Reinforcements, Structural Applications

1.INTRODUCTION

Hybrid composites combining graphene oxide (GO) and natural silk fibers offer a synergistic approach to improving the mechanical, thermal, and environmental properties of epoxy resin matrices. The use of these reinforcements necessitates precise fabrication techniques to overcome issues such as GO agglomeration and improper silk fiber dispersion, which can adversely affect composite performance. Vacuum-Assisted Resin Transfer Molding (VARTM) offers a scalable solution by ensuring uniform resin impregnation, reduced void content, and enhanced fiber-matrix adhesion. Ultrasonication further aids in dispersing GO uniformly, addressing agglomeration and improving interfacial bonding with the epoxy matrix.

This study reviews existing research to evaluate the role of VARTM, the effects of GO dispersion methods, and the proportional influence of reinforcements on composite properties. Challenges during hybridization are addressed, and recommendations for effective processing are provided. It also examines the interplay between GO and SF, emphasizing fabrication techniques, hybridization challenges, and optimization strategies for achieving superior composite performance.

2. LITERATURE REVIEW

2.1. Fabrication Techniques for GO-Silk-Epoxy Composites

Vacuum-Assisted Resin Transfer Molding (VARTM) has been widely employed for fabricating epoxy composites due to its ability to produce high-quality, void-free structures. Research suggests that maintaining a vacuum pressure of 30 PSI during resin infusion ensures uniform matrix infiltration into the reinforcement fibers, reducing porosity and enhancing mechanical properties [1, 2].

Ultrasonication is critical for dispersing GO into the epoxy resin. GO tends to agglomerate due to Van der Waals forces, leading to uneven distribution and reduced performance [3]. Studies show that ultrasonication for 30–60 minutes, depending on the GO concentration, breaks apart agglomerates and ensures homogeneous dispersion, significantly improving the mechanical properties of the resulting composites [4, 5]. The body of the paper consists of numbered sections that present the main findings. These sections should be organized to best present the material.

2.2. Avoiding Agglomeration Effects on GO

Agglomeration of GO sheets is a persistent challenge in composite fabrication. To mitigate this, surfactants like Triton X-100 and coupling agents such as silanes are often added during ultrasonication [6]. Additionally, functionalizing GO with carboxyl or hydroxyl groups enhances its compatibility with the epoxy matrix, promoting better bonding and distribution [7].

2.3. Optimal Proportions for Reinforcement

The proportion of GO in epoxy composites plays a critical role in determining performance. Literature indicates that GO concentrations of 0.3–0.5 wt.% provide optimal improvements in tensile strength and fracture toughness [8]. Beyond 0.5 wt.%, GO tends to aggregate, leading to stress concentration points that degrade mechanical properties [9].

For silk fibers, studies such as Kang Yang et al. demonstrate that high silk content (30–70% by volume) significantly enhances tensile strength, flexural strength, and fracture toughness due to improved load transfer and energy absorption mechanisms [10]. This contrasts with earlier studies that suggested lower silk fractions (<15%) were optimal, highlighting the impact of fabrication methods and silk alignment.

2.4. Challenges in Hybridization

Hybridization of GO and silk fibers introduces unique challenges, such as ensuring uniform dispersion of GO in the epoxy matrix while maintaining proper alignment and wetting of silk fibers. One significant issue is the competition between the hydrophilic silk and hydrophobic epoxy, which can hinder adhesion [10].

Surface treatments on silk fibers, such as alkali or silane coupling, enhance compatibility with the epoxy matrix [11]. Additionally, staggered addition of GO during resin mixing ensures the nanofiller is evenly distributed before introducing silk fibers, avoiding clumping and ensuring a balanced reinforcement effect [12].

3. FABRICATION AND TESTING PROTOCOL

3.1. Fabrication Steps

The fabrication of GO-silk fiber epoxy composites adopts a hybridization approach to address challenges such as reinforcement dispersion, volume fraction optimization, and environmental durability. The pressurized Vacuum-Assisted Resin Transfer Molding (VARTM) process, chosen over the wet-lay method, ensures superior composite quality and industrial scalability. VARTM offers key advantages, including enhanced fiber-matrix bonding, reduced void content, and uniform reinforcement impregnation, making it suitable for structural applications demanding consistent mechanical and thermal properties.

The hybrid laminate consists of silk fiber (SF) layers at two volume concentrations (40% and 60%), reinforced with an epoxy matrix infused with graphene oxide (GO) at weight fractions of 0%, 0.5%, 1%, 1.5%, 2%, 3%, and 5%. Operating at a controlled pressure of 30 psi, the VARTM process ensures precise resin flow, uniform impregnation, and minimized void formation, enhancing both structural integrity and scalability.

Pre-treatment of SF with a 2% NaOH solution enhances interfacial bonding by increasing surface roughness, while drying at 60°C for 24 hours and storage in a desiccator ensure low moisture content. Ultrasonication of GO at 25°C and 30% amplitude for 2 hours achieves uniform dispersion, mitigating agglomeration challenges observed in prior studies. Integrating these pre-treatments with the VARTM process facilitates effective reinforcement and enables the fabrication of composites with superior mechanical properties and reproducibility.

The composite laminate undergoes a two-stage curing process: initial curing at room temperature for 24 hours ensures proper resin infiltration, followed by post-curing at 80°C for 3 hours to achieve optimal crosslinking and enhance mechanical and thermal properties.

3.2. Testing Protocols

- **Tensile Strength:** ASTM D638 standard is used to evaluate the composite’s tensile performance.
- **Flexural Properties:** ASTM D790 standard is employed to measure flexural strength and modulus.
- **Impact Resistance:** Charpy impact tests are conducted to determine toughness**.**
- **Microscopy:** Scanning Electron Microscopy (SEM) is used to analyse the dispersion of GO and fiber-matrix interactions.

4. DISCUSSION

4.1. Influence of GO Dispersion

Uniform dispersion of GO significantly improves the mechanical performance of epoxy composites. Ultrasonication ensures even distribution and functionalization, which minimizes agglomeration and enhances load transfer. Proper dispersion results in a 25–30% improvement in tensile strength compared to neat epoxy [13].

4.2. Role of Silk Fibers

High silk volume fractions (30–70%) significantly enhance critical mechanical properties such as tensile strength and fracture toughness by facilitating efficient load transfer and energy absorption [14]. Alkali pretreatment of silk fibers further improves interfacial adhesion by increasing surface roughness, enabling better mechanical interlocking and reducing delamination [14]. However, challenges such as reduced resin infiltration at higher silk content can be effectively mitigated through optimized VARTM techniques, which ensure void-free laminates and result in composites with superior toughness [2].

4.3. Hybridization Challenges and Solutions

The hybridization of GO and SF introduces complexities in ensuring balanced dispersion and alignment. Hybridization challenges, including material compatibility and dispersion, are mitigated through techniques like GO functionalization and silk fiber surface treatments. These measures ensure composites with balanced mechanical properties and enhanced durability [15].

4.4 Scalability and Durability

Scaling up the fabrication process while maintaining consistent resin infiltration presents a significant challenge. This study employs VARTM at a controlled pressure of 30 Psi, ensuring void-free laminates and uniform reinforcement integration [16]. However, prolonged exposure to environmental factors such as UV radiation and humidity can degrade silk fibers (SF), compromising the composite's performance. Protective coatings or modified epoxy matrices are recommended to enhance environmental resistance [17]. Optimizing resin infiltration parameters, particularly for high fiber content composites, is critical. Controlling resin viscosity and vacuum pressure during the VARTM process is essential to minimize void content and ensure consistent mechanical performance [18]. Addressing these challenges can significantly improve the scalability and durability of hybrid composites, making them suitable for long-term structural applications in demanding environments.

5. CONCLUSIONS

The fabrication of graphene oxide (GO) and natural silk fiber (SF)-reinforced epoxy composites requires meticulous control of processing parameters to achieve optimal mechanical, thermal, and environmental performance. The use of Vacuum-Assisted Resin Transfer Molding (VARTM) at 30 PSI ensures uniform resin infiltration, reduced void content, and scalability, while ultrasonication effectively disperses GO, preventing agglomeration and enhancing fiber-matrix bonding. High silk volume fractions (30–70%) demonstrate superior mechanical properties, such as tensile strength and impact resistance, provided challenges like resin infiltration and fiber alignment are adequately addressed. This study highlights the potential of hybrid GO-SF composites in structural applications. Furthermore, it offers valuable insights into fabrication techniques and hybridization strategies, laying a strong foundation for future research.

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