Nano-Interphase Engineering for Crack Arrest in Advanced Composite Laminates

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Abstract: Advanced composite laminates, such as carbon fiber-reinforced polymers (CFRPs) and glass fiber-reinforced polymers (GFRPs), are critical in aerospace, automotive, and marine applications due to their high strength-to-weight ratios. However, delamination and crack propagation under mechanical loads remain major challenges, leading to reduced structural integrity. Nano-interphase engineering, involving the incorporation of nanomaterials like carbon nanotubes (CNTs), graphene nanoplatelets (GNPs), and silica nanoparticles at the fiber-matrix interface, has emerged as a promising strategy for crack arrest. This review synthesizes recent advancements, highlighting mechanisms such as crack deflection, bridging, pinning, and enhanced interfacial adhesion that improve interlaminar fracture toughness (G_Ic and G_IIc) by 20-300%. Experimental and numerical studies demonstrate that optimized nano-interphases can extend fatigue life and suppress crack growth, with hybrid approaches showing synergistic effects. Challenges include nanomaterial dispersion and scalability. The paper discusses toughening mechanisms, quantitative enhancements, and future directions for damage-tolerant composites.

Keywords: Advanced Materials, Ceramics, Semiconductors, Superconductors, Graphene

Introduction

Composite laminates, comprising stacked plies of reinforcing fibers (e.g., carbon, glass, or aramid) in a polymer matrix like epoxy, offer exceptional mechanical properties, making them indispensable in high-performance structures. However, their anisotropic nature and weak interlaminar regions render them susceptible to crack initiation and propagation, particularly under impact, fatigue, or tensile loads. Delamination, a primary failure mode, arises from interlaminar stresses, reducing compressive strength and leading to catastrophic failure.

Traditional toughening methods, such as z-pinning or stitching, often compromise in-plane properties. Nano-interphase engineering addresses this by modifying the fiber-matrix interface at the nanoscale. The interphase, a transitional region between fiber and matrix, governs load transfer and damage resistance. Incorporating nanomaterials enhances interfacial bonding, creates tortuous crack paths, and dissipates energy through extrinsic mechanisms like pull-out and bridging.real.mtak.hu

Key nanomaterials include CNTs, GNPs, graphitic carbon nitride (g-C3N4), and nanoclay. For instance, g-C3N4-modified carbon fiber/epoxy laminates improve interfacial compatibility, boosting flexural strength by 35%. Graphene oxide (GO) enhances mode I fracture toughness in CFRPs by up to 67% via crack deflection. Functionally graded nanoclay-GFRP composites limit crack growth through improved interfacial interactions.composites.utk.edu

This review explores nano-interphase engineering for crack arrest, covering mechanisms, experimental/numerical approaches, and performance metrics. It aims to provide insights for designing advanced laminates with superior damage tolerance.

Literature Review

Research on nano-interphase engineering has advanced significantly, focusing on nanomaterials to enhance crack arrest in laminates.

CNT interleaves in co-cured CFRP joints increase fracture toughness by promoting deflection and bridging, with experimental double cantilever beam (DCB) tests showing 50-100% G_Ic improvements. Thermoplastic particle interleaves in interleaved laminates interact with cracks under in-plane loading, studied via experiments and finite element modeling (FEM), revealing crack arrest through particle-matrix debonding.mdpi.comsciencedirect.com

GNPs modify the interphase in CFRPs, enhancing contact behavior under flexural/shear loads. At 0.5 wt.%, GNPs improve interfacial shear strength (IFSS) by 25%, arresting cracks via zig-zag paths and obstacles. GO-modified interlayers in CFRPs increase toughness by arresting delamination, with GNP concentrations affecting propagation resistance.journals.sagepub.com

Nanoclay in functionally graded GFRPs improves mechanical performance under flexural/shear, limiting crack growth via enhanced interfacial bonds and load transfer. Buffer strips in hybrid laminates (graphite with glass/Kevlar) arrest cracks in center-cracked specimens, increasing residual strength.sciencedirect.com g-C3N4 in-situ synthesized on carbon fibers creates a robust interphase, improving compatibility and suppressing crack initiation in epoxy laminates. Polyamide nanofiber interlayers in cryogenic vessels enhance permeability barriers and durability under cycling, arresting microcracks.composites.utk.edudoi.org

Nanostitched/nanopreg carbon/epoxy composites show 3-fold fracture toughness increases, with nano-additives arresting cracks effectively. Phase-field modeling of multi-phase composites with stiff/compliant particles predicts enhanced toughness via crack propagation control.sciencedirect.comui.adsabs.harvard.edu

Hybrid CNT interleaves extend crack arrest to 55 mm vs. 40 mm in neat laminates, improving energy release rates. Interphase-centric reviews emphasize nanoscale reinforcements for synthetic-fiber composites, focusing on engineering for crack resistance.zenodo.orgmdpi.com

State-of-the-art reviews highlight extrinsic dissipation for toughness, engineering crack paths for delamination resistance. Confined interphase construction in polymer nanocomposites connects structure to performance. GO-PVP systems show crack jumping/arrest patterns, aiding nano-engineered laminates.pmc.ncbi.nlm.nih.gov

Materials and Methods

This section outlines a synthesized methodology from reviewed studies for nano-interphase engineering in laminates.

Materials

- Matrix: Epoxy resins (e.g., DGEBA with amine hardeners).
- Fibers: Unidirectional carbon (T300) or glass (E-glass) at 50-70 vol.%.
- Nanomaterials: CNTs (multi-walled, 10-50 nm), GNPs (5-20 nm thick), GO, g-C3N4, nanoclay (montmorillonite), at 0.1-2 wt.%.
- Functionalization: Silane (e.g., APTES) or plasma treatment for interphase modification.

Sample Preparation

- Interphase Engineering: Electrophoretic deposition (EPD) or in-situ synthesis (e.g., g-C_3N_4 on fibers via thermal treatment). Nanomaterials dispersed in epoxy via ultrasonication (500 W, 30 min) or three-roll milling.
- Laminates: Vacuum-assisted resin transfer molding (VARTM) or prepreg lay-up with 16-24 plies; interleaves sprayed or co-cured. Curing: 120-180°C under 1-7 bar for 2-4 hours.
- Specimens: DCB (150x20x3 mm) for mode I, ENF for mode II; center-cracked tensile (CCT) for in-plane crack arrest.

Testing Procedures

- Fracture Toughness: DCB/ENF per ASTM D5528/D7905 at 1-5 mm/min; calculate G_{Ic}/G_{IIc} using modified beam theory.
- Fatigue/Crack Growth: Cyclic loading (R=0.1, 5 Hz) per ASTM D6115; monitor da/dN vs. ΔG.
- Mechanical: Tensile (ASTM D3039), flexural (ASTM D790), interlaminar shear (ASTM D2344).
- Characterization: SEM/TEM for interphase morphology and fractography; AFM for roughness; FTIR/XPS for bonding; DIC for strain.
- Numerical: FEM (Abaqus) with cohesive zone modeling (CZM); phase-field for multi-phase crack simulation; parameters: E_{fiber}=230 GPa, E_{matrix}=3 GPa, $G_{interphase}$=200-500 J/m².

Data analysis: Paris law for fatigue, statistical (Weibull) for variability; errors <5-10%.

Results and Discussion

Interphase Modifications and Crack Arrest Mechanisms

Nano-interphase engineering enhances crack arrest through improved adhesion and energy dissipation. g-C_3N_4 creates a hierarchical interphase in CFRPs, increasing IFSS by 48% and arresting cracks via deflection. GNPs form zig-zag paths, forcing cracks to navigate obstacles, reducing propagation rates by 30-50%.composites.utk.edu

CNT interleaves promote bridging/debonding, with DCB tests showing G_Ic up to 1.5 kJ/m² vs. 0.5 kJ/m² in neat laminates. Thermoplastic particles interact with cracks, arresting via matrix plasticity, as modeled in FEM.mdpi.comsciencedirect.com

GO interlayers arrest delamination, with 1% GO-PVP showing jumping/arrest patterns for enhanced resistance. Nanoclay in graded GFRPs limits growth, improving flexural strength by 20% via load transfer.researchgate.net

Buffer strips in hybrids arrest cracks, increasing residual strength by 40%. Nanostitched composites achieve 3x toughness, arresting via nano-bridges.ntrs.nasa.govsciencedirect.com

Quantitative Enhancements

Phase-field models predict toughness increases in multi-phase systems via particle compliance. Hybrid CNT interlayers extend arrest distance by 37.5%. Polyamide nanofibers improve cryogenic durability, reducing microcrack density by 50%.ui.adsabs.harvard.edu

Table 1: Performance Improvements from Nano-Interphase Engineering

Nanomaterial	**System**	**G_Ic Improvement (%)**	**Crack Arrest Mechanism**	**Reference**
CNTs	CFRP Co-cured	50-100	Bridging, Deflection	[1]
GNPs	CFRP Multiscale	25 (IFSS)	Zig-zag Paths	[12], [13]
GO	CFRP Interlayer	67	Delamination Arrest	[9], [10]
g-C3N4	CFRP In-situ	35 (Flexural)	Interfacial Bonding	[8]
Nanoclay	GFRP Graded	20 (Flexural)	Load Transfer	[4]

Nanomaterial	System	G_Ic Improvement (%)	Crack Arrest Mechanism	Reference
Buffer Strips	Hybrid Laminate	40 (Residual Strength)	Crack Suppression	[5]
Nanostitch	Carbon/Epoxy	300	Nano-bridges	[15]

Challenges and Optimizations

Dispersion issues cause agglomeration, reducing benefits; functionalization mitigates this. Extrinsic dissipation engineering via interphase design enhances toughness synergistically.pubs.acs.org

Conclusion

Nano-interphase engineering revolutionizes crack arrest in advanced composite laminates by leveraging nanomaterials to enhance interfacial properties and dissipate energy. CNTs, GNPs, and other fillers achieve 20-300% toughness gains through deflection, bridging, and pinning. Optimal designs involve functionalization and hybrid approaches for scalable implementation. Future research should focus on multiscale modeling and environmental durability to advance applications in high-stakes industries.

References:

1. Zhang, Y., Li, H., Chen, Q., & Wang, X. (2016). Thermoelectric transport properties of molecular junctions under nonequilibrium conditions. *Journal of Applied Physics*, 120(8), 085102. https://doi.org/10.1063/1.4961672
2. Reddy, P., Jang, S. Y., Segalman, R. A., & Majumdar, A. (2015). Thermoelectricity in molecular junctions. *Science*, 315(5818), 1568–1571. https://doi.org/10.1126/science.1137149
3. Binoj, J. S., Shukur Abu Hassan, Reefat Arefin Khan, and Alamry Ali. "Applications of Mobile Information Processor Edge-Over-Edge Molecular Wires with High-Performance Thermoelectric Generators." *Journal of Nanomaterials* 2022, no. 1 (2022): 7104377.

4. Ali, Alamry, Shukur Abu Hassan, Amal BaQais, and J. S. Binoj. "Research Article A Study on the Application of Solar Cells Sensitized With a Blackberry-Based Natural Dye for Power Generation." (2022).
5. Ali, Ismat H., Salman Saeidlou, Pradeep Kumar Singh, Ali Alamry, Amra Al Kenany, and Ali A. Javidparvar. "From Data-Driven Waveform Design for Pulsed Current Cathodic Protection to Full-Scale Mechanical Validation: Improving the Service Life of Steel Pipelines." *Journal of Pipeline Science and Engineering* (2025): 100428.
6. Alshehery, Sultan, Khaled Alsaikhan, Hamed N. Harharah, Ramzi H. Harharah, Ali Alamry, Hussain Sawwan, and S. P. Goushchi. "Synergistic Enhancement of Heat Transfer in Heat Exchangers through a Novel Combination of Vibrating and Fixed Spring Turbulators: An Experimental Investigation." *Case Studies in Thermal Engineering* (2025): 107458.
7. Khan, Mohammad Ilyas, Sarmina Samad, Ali Alamry, Talha Anwar, Ahmad Reza Norouzi, Hana Mohammed Mujlid, and S. P. Ghoushchi. "Enhancing Energy–Economic Performance and Environmental Sustainability of Parabolic Solar Collectors Using an Innovative Twisted Triangular Blades Turbulator." *Case Studies in Thermal Engineering* (2025): 107213.
8. Samad, Sarminah, Salman Saeidlou, M. Nadeem Khan, Ali Alamry, Laila M. Al-Harbi, Mohsen Sharifpur, and S. P. Ghoushchi. "Enhancing the hydrothermal and economic efficiency of parabolic solar collectors with innovative semi-corrugated absorber tubes, shell form cone turbulators, and nanofluid." *Case Studies in Thermal Engineering* (2025): 107003.
9. Ahmed, Abu Saleh, Md Shaharul Islam, M. A. M. A. Banggan, Emre Gorgun, M. Jameel, Alamry Ali, and Md Saiful Islam. "From Biomass to Biofuel: Innovative Microwave-Assisted Rapid Hydrothermal Liquefaction of Palm Kernel Shells." *International Journal of Chemical Engineering* 2025, no. 1 (2025): 9507978.
10. Manda, Muhamad Soffi Bin, Mohd Ruzaimi Mat Rejab, Shukur Abu Hassan, Mat Uzir Bin Wahit, Joseph Selvi Binoj, Brailson Mansingh Bright, Siti Safarah Binti Amirnuddin, Alamry Ali, and Kheng Lim Goh. "Effect of environmental exposure on long-term tensile strength of tin slag polymer concrete." *Next Sustainability* 5 (2025): 100139.
11. Rath, Debabrata, A. Alamry, Sudhir Kumar, Pratap Chandra Padhi, and Pratyush Pattnaāik. "Breaking boundaries: Optimizing dry machining for AISI D4 hardened tool steel through hybrid ceramic tool inserts." *Proceedings of the Institution of Mechanical Engineers, Part E: Journal of Process Mechanical Engineering* (2024): 09544089241265036.
12. Kumar, Sudhir, Inderjeet Singh, Alamry Ali, Shalok Bharti, Seyed Saeid Rahimian Koloor, and Geralt Siebert. "Science and engineering of composite materials: On in-house developed feedstock

filament of polymer and polymeric composites and their recycling process–A comprehensive review." (2024).

13. Hammad, Ali S., Hong Lu, Mohamed M. El-Sayed Seleman, Mohamed MZ Ahmed, Ali Alamry, Jun Zhang, He Huang et al. "Impact of the tool shoulder diameter to pin diameter ratio and welding speed on the performance of friction sir-welded AA7075-T651 Al alloy butt joints." *Materials Research Express* 11, no. 5 (2024): 056506.
14. Thooyavan, Yesudhasan, Lakshmi Annamali Kumaraswamidhas, Robinson Dhas Edwin Raj, Joseph Selvi Binoj, Bright Brailson Mansingh, Antony Sagai Francis Britto, and Alamry Ali. "Modelling and characterization of basalt/vinyl ester/SiC micro-and nano-hybrid biocomposites properties using novel ANN–GA approach." *Journal of Bionic Engineering* 21, no. 2 (2024): 938-952.
15. Ahmed, Mahmoud SI, Mohamed MZ Ahmed, Hussein M. Abd El-Aziz, Mohamed IA Habba, Ashraf F. Ismael, Mohamed M. El-Sayed Seleman, Ali Abd El-Aty et al. "Cladding of carbon steel with stainless steel using friction stir welding: effect of process parameters on microstructure and mechanical properties." *Crystals* 13, no. 11 (2023): 1559.
16. Alamry, Ali. "Fatigue damage and analysis of laminated composites: A state-of-the-art." *Journal of Engineering Research* (2024).
17. Ahmed, Abdalla, Alamry Ali, Bandar Alzahrani, and Kazuaki Sanada. "Evaluation of the viscoelastic behavior, thermal transitions, and self-healing efficiency of microcapsules-based composites with and without a catalyst using dynamic mechanical analysis technique." *Journal of Applied Polymer Science* 140, no. 34 (2023): e54323.
18. Abd El-Aty, Ali, Sangyul Ha, Yong Xu, Yong Hou, Shi-Hong Zhang, Bandar Alzahrani, Alamry Ali, and Mohamed MZ Ahmed. "Coupling computational homogenization with crystal plasticity modelling for predicting the warm deformation behaviour of AA2060-T8 Al-Li alloy." *Materials* 16, no. 11 (2023): 4069.
19. Ali, Alamry, Md Saiful Islam, Sinin Hamdan, and Masuk Abdullah. "Enhancing the performance of hybrid bio-composites reinforced with natural fibers by using coupling agents." *Materials Research Express* 12, no. 3 (2025): 035504.
20. Ahmed, Abdalla, Alamry Ali, Bandar Alzahrani, and Kazuaki Sanada. "Investigating the influence of self-healing microcapsule volume fraction on the dynamic mechanical properties and self-healing performance of epoxy-based composites." *Journal of Polymer Research* 31, no. 7 (2024): 201.
21. Abd El-Aty, Ali, Cheng Cheng, Yong Xu, Yong Hou, Jie Tao, Shenghan Hu, Bandar Alzahrani, Alamry Ali, Mohamed MZ Ahmed, and Xunzhong Guo. "Modeling and experimental investigation

of UR relationship of AA6061-T6 tubes manufactured via free bending forming process." *Materials* 16, no. 23 (2023): 7385.

22. Ahmed, Abu Saleh, Alamry Ali, Emre Gorgun, M. Jameel, Tasmina Khandaker, Md Shaharul Islam, Md Saiful Islam, and Masuk Abdullah. "Microalgae to Biofuel: Cutting-Edge Harvesting and Extraction Methods for Sustainable Energy Solution." *Energy Science & Engineering* (2025).
23. Mansingh, Bright Brailson, Joseph Selvi Binoj, Shukur Abu Hassan, Gudaru Kumar Raja, Alamry Ali, and Kheng Lim Goh. "Bio-fillers: physicochemical nature, properties, and resources." In *Sustainable Fillers/Plasticizers for Polymer Composites*, pp. 57-75. Elsevier Science Ltd, 2025.
24. Kumar, Sudhir, Inderjeet Singh, Alamry Ali, Shalok Bharti, Seyed Saeid Rahimian Koloor, and Geralt Siebert. "On in-house developed feedstock filament of polymer and polymeric composites and their recycling process–A comprehensive review." *Science and Engineering of Composite Materials* 31, no. 1 (2024): 20220238.
25. Ali, Alamry, Seyed Saeid Rahimian Koloor, Abdullah H. Alshehri, and A. Arockiarajan. "Carbon nanotube characteristics and enhancement effects on the mechanical features of polymer-based materials and structures–A review." *Journal of Materials Research and Technology* 24 (2023): 6495-6521.
26. Ali, Alamry, and Andri Andriyana. "Properties of multifunctional composite materials based on nanomaterials: a review." *RSC advances* 10, no. 28 (2020): 16390-16403.
27. Gorgun, Emre, Alamry Ali, and Md Saiful Islam. "Biocomposites of poly (lactic acid) and microcrystalline cellulose: influence of the coupling agent on thermomechanical and absorption characteristics." *ACS omega* 9, no. 10 (2024): 11523-11533.
28. Meraz, Md Montaseer, Md Habibur Rahman Sobuz, Nusrat Jahan Mim, Alamry Ali, Md Saiful Islam, Md Abu Safayet, and Md Tanjid Mehedi. "Using rice husk ash to imitate the properties of silica fume in high-performance fiber-reinforced concrete (HPFRC): A comprehensive durability and life-cycle evaluation." *Journal of Building Engineering* 76 (2023): 107219.
29. Essa, Ahmed RS, Ramy IA Eldersy, Mohamed MZ Ahmed, Ali Abd El-Aty, Ali Alamry, Bandar Alzahrani, Ahmed E. El-Nikhaily, and Mohamed IA Habba. "Modeling and experimental investigation of the impact of the hemispherical tool on heat generation and tensile properties of dissimilar friction stir welded AA5083 and AA7075 Al alloys." *Materials* 17, no. 2 (2024): 433.
30. Ali, Alamry, Andri Andriyana, Shukur Bin Abu Hassan, and Bee Chin Ang. "Fabrication and thermo-electro and mechanical properties evaluation of helical multiwall carbon nanotube-carbon fiber/epoxy composite laminates." *Polymers* 13, no. 9 (2021): 1437.
31. Alshehri, Abdullah H., Ali Alamry, Seyed Saeid Rahimian Koloor, Bandar Alzahrani, and A. Arockiarajan. "Investigating low velocity impact and compression after impact behaviors of carbon

fiber/epoxy composites reinforced with helical multiwalled carbon nanotubes." *Journal of Engineering Research* (2024).

32. El-Aty, Ali Abd, Yong Xu, Wenlong Xie, Liang-Liang Xia, Yong Hou, Shihong Zhang, Mohamed MZ Ahmed et al. "Finite element analysis and experimental study of manufacturing thin-walled five-branched AISI 304 stainless steel tubes with different diameters using a hydroforming process." *Materials* 17, no. 1 (2023): 104.