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Multifunctional Lignin-Modified Jute/Bio-Epoxy Composites with MWCNT-Based Piezoresistive Self-Sensing and Structural Health Monitoring for Sustainable Housing Applications

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Abstract

This study develops a multifunctional natural-fibre-reinforced polymer composite based on alkali-treated jute fabric, bio-based epoxy, kraft lignin and a small amount of multiwalled carbon nanotubes (MWCNTs), with particular emphasis on polymer-matrix modification, fibre–matrix interfacial behaviour and intrinsic damage sensing. Kraft lignin was incorporated at 0, 2, 4 and 6 wt.% to examine its influence on polymer-chain mobility, interfacial morphology, thermal stability and mechanical response, while 0.5 wt.% MWCNTs was introduced into the selected 4 wt.% lignin formulation to impart piezoresistive functionality. FTIR results indicated enhanced hydrogen–bonding interactions within the polymer–lignin–fibre system, with the O–H band shifting from 3338 cm⁻¹ for JE to 3325 cm⁻¹ for JE-L4. SEM observations showed reduced fibre pull-out and improved polymer adhesion at 4 wt.% lignin, whereas 6 wt.% lignin promoted agglomeration, microvoid formation and incomplete fibre wetting. The JE-L4 composite exhibited a flexural strength of 118.6 MPa and flexural modulus of 5.12 GPa, corresponding to improvements of 28.4% and 22.5%, respectively, relative to the reference JE composite. Its storage modulus increased to 3.31 GPa and glass-transition temperature to 87.6°C, confirming greater restriction of epoxy-chain mobility. The JE-L4C composite further increased flexural strength and modulus to 121.9 MPa and 5.36 GPa, respectively, while providing an effective gauge factor of approximately 18.7. Overall, 4 wt.% lignin provided the most favourable balance between polymer-network modification, interfacial reinforcement and processability, while MWCNT incorporation transformed the modified bio-epoxy matrix into an electrically responsive phase capable of monitoring strain and progressive damage.

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INTRODUCTION

The transition from conventional petroleum-intensive construction materials toward lightweight, resource-efficient and lower-carbon alternatives has increased interest in natural-fibre-reinforced polymer composites. Plant fibres – such as jute,

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flax, hemp, kenaf, and sisal – offer relatively low density, renewability, reasonable specific mechanical properties and comparatively low processing energy. Their incorporation into thermosetting or thermoplastic polymer matrices has, therefore, been investigated for automotive panels, interior structures, furniture, building products and lightweight civil-engineering components [1, 2].

From a polymer-composite perspective, however, the environmental origin of reinforcement alone does not guarantee satisfactory engineering performance. The behaviour of a natural-fibre composite is governed primarily by the matrix chemistry, fibre surface condition, polymer–fibre interphase, fibre volume fraction, void content and the ability of the polymer phase to transfer stress to the reinforcement. Natural fibres contain abundant hydroxyl groups and are intrinsically hydrophilic, whereas many polymer matrices are comparatively hydrophobic. Insufficient physicochemical affinity between the two phases can, therefore, result in fibre pull-out, interfacial voids, premature debonding and deterioration of mechanical performance.

The polymeric matrix is particularly important in housing applications because it binds and protects the fibrous skeleton, transfers shear stress between fibres, controls viscoelastic response and governs the development of microcracks [3, 4]. Modification of the matrix using renewable macromolecular fillers is, therefore, an attractive strategy for simultaneously improving the sustainability and functionality of natural-fibre polymer composites.

Lignin is an aromatic, three-dimensional natural macromolecule present in lignocellulosic biomass. It contains phenolic, aliphatic hydroxyl, ether and aromatic functionalities and is generated in substantial quantities as a by-product of pulp and biorefinery processes. Its aromatic structure provides greater rigidity than many other biomass-derived polymers, while its surface functional groups permit physical and chemical interaction with thermosetting polymers and lignocellulosic fibres. Consequently, lignin has been studied as a particulate filler, reactive modifier, toughening component, compatibilizer and precursor for carbonaceous materials in polymer composites [5, 6].

The influence of lignin is nevertheless strongly dependent on concentration. At relatively low loading, well-dispersed lignin particles can modify cross-linking, restrict local polymer-chain mobility and improve stress transfer through the interphase. At excessive concentrations, lignin agglomeration and the increased viscosity of the uncured resin can restrict impregnation of the fibre architecture. Previous studies on lignin-modified fibre/epoxy systems have similarly shown that an optimum lignin concentration is required because excessive addition can reduce resin flow and promote heterogeneous interfacial regions [7].

A second challenge associated with natural-fibre polymer components is the detection of progressive damage during service. Conventional inspection identifies damage only after external inspection is performed, whereas polymer composites can develop subsurface matrix cracking, interfacial debonding and fibre damage before visible failure occurs. Structural health monitoring (SHM) is, therefore, increasingly being incorporated directly into fibre-reinforced polymers.

Among the available SHM mechanisms, piezoresistive sensing is particularly attractive for polymer composites. Electrically conductive particles incorporated into an insulating polymer matrix form interconnected or near-connected conductive pathways. Mechanical deformation changes particle separation, contact resistance and tunnelling distance, producing a measurable change in electrical resistance. At larger deformation, microcracks and interfacial damage progressively disrupt the network, enabling the same material to provide information on both elastic strain and damage accumulation.

For the present material system, MWCNTs were selected as a secondary functional filler because a low nanotube content can form an electrically responsive network without fundamentally changing the natural-fibre composite architecture. Lignin remains the principal bio-based matrix modifier and is expected to contribute to nanotube distribution by increasing matrix polarity and interfacial interaction [8, 9]. Thus, rather than attaching a separate sensor to the finished component, the polymer matrix itself becomes the sensing medium.

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The scientific concept of the present study is consequently based on a fibre–lignin–polymer–conductive filler hierarchy. Jute fibres provide the load-bearing reinforcement; bio-epoxy provides continuous-phase stress transfer; kraft lignin modifies the polymer network and fibre/matrix interphase; and a small quantity of MWCNTs creates electrically responsive pathways throughout the lignin-modified matrix.

The principal objective was to develop a sustainable natural-fibre polymer composite capable of simultaneously providing mechanical reinforcement and in-situ deformation/damage monitoring. Particular attention was given to the polymer-composite aspects of lignin concentration, filler dispersion, interfacial morphology, thermal stability, viscoelastic response and damage-induced electrical resistance variation.

Unlike broader investigations involving a large number of analytical techniques, the present work was deliberately restricted to five complementary characterization methods: Fourier-transform infrared spectroscopy (FTIR) was employed to evaluate chemical interactions within the composite system; scanning electron microscopy (SEM) was used to examine polymer/fibre interfacial morphology; thermogravimetric analysis (TGA) was carried out to assess thermal stability; dynamic mechanical analysis (DMA) was performed to investigate the viscoelastic behaviour of the polymer network; and simultaneous flexural–piezoresistive testing was conducted to evaluate both mechanical performance and structural health monitoring capability. The resulting multifunctional composite is primarily intended for semi-structural housing applications, including modular partition panels, wall-panel skins, ceiling panels, internal cladding, door panels and sandwich structures, rather than as a direct replacement for highly loaded primary structural members.

MATERIALS AND METHODS

Materials

A commercially available bio-based epoxy resin containing approximately 30–35% renewable carbon content was used as the continuous polymer matrix together with a compatible amine-based curing agent at the manufacturer-recommended stoichiometric ratio. Plain-woven jute fabric with an areal density of approximately 300 g/m² was selected as the natural-fibre reinforcement. Prior to fabrication, the jute fabrics were dried at 80°C for 12 h and subsequently treated with 5 wt.% NaOH solution for 2 h to remove weakly bonded surface constituents and improve fibre surface roughness, followed by repeated washing with distilled water until neutral pH was attained and final drying at 80°C. Commercial kraft lignin was used as a renewable polymer modifier and was vacuum-dried at 70°C before processing. Lignin was incorporated at 0, 2, 4 and 6 wt.% by partially replacing the epoxy fraction. Multiwalled carbon nanotubes (MWCNTs) were used as the conductive filler for structural health monitoring and were incorporated at a fixed concentration of 0.5 wt.% in the optimized lignin-modified formulation. Accordingly, the composite systems consisted of the reference jute/epoxy composite (JE), lignin-modified composites containing 2, 4 and 6 wt.% lignin (JE-L2, JE-L4 and JE-L6, respectively), and a self-sensing composite containing 4 wt.% lignin and 0.5 wt.% MWCNTs (JE-L4C).

Methods

Kraft lignin was dispersed in the bio-epoxy resin using mechanical stirring at approximately 1000 rpm for 20 min followed by ultrasonication for 15 min, while maintaining the resin temperature below 50°C to avoid premature curing. For the self-sensing JE-L4C formulation, MWCNTs were first dispersed in the epoxy through sequential mechanical stirring and sonication, after which 4 wt.% lignin was incorporated to obtain a homogeneous conductive lignin-modified resin system. The curing agent was subsequently added, and the mixture was vacuum-degassed prior to laminate fabrication. Composite laminates were produced using compression-assisted hand lay-up by sequentially impregnating treated jute fabric plies with the corresponding resin formulation, followed by consolidation under moderate pressure to improve fibre wetting and reduce void formation. The laminates were cured for 24 h at room temperature and post-cured at 80°C for 2 h before specimens were cut to the required dimensions. The overall laboratory processing sequence adopted for preparation of the lignin-modified jute/bio-epoxy composites and the MWCNT-containing self-sensing composite is illustrated in Figure 1.

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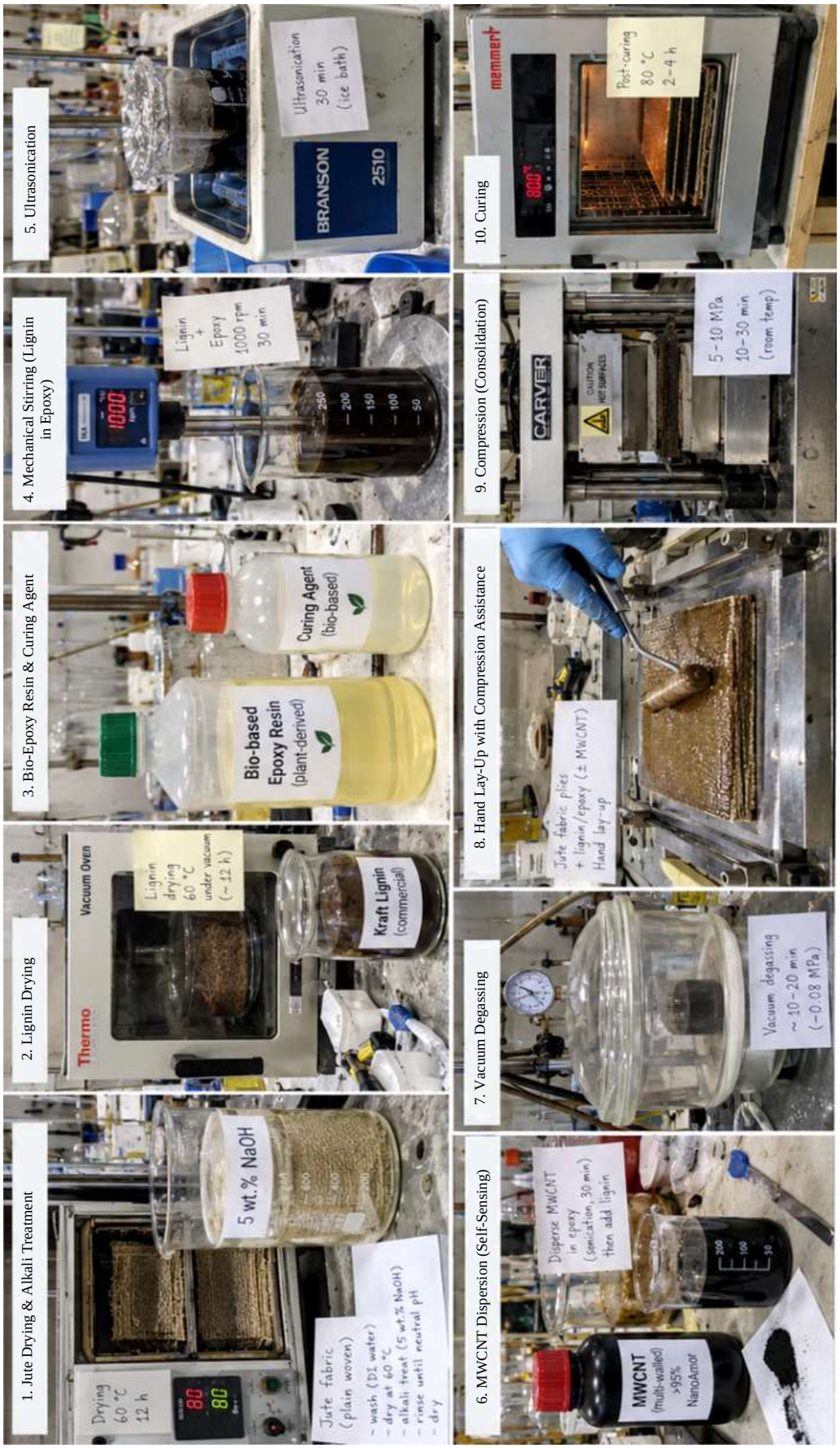


Figure 1. Processing sequence of lignin-modified jute/bio-epoxy composites with MWCNT self-sensing modification.

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The fabricated composites were evaluated using only five characterization techniques: Fourier-transform infrared spectroscopy (FTIR) to investigate chemical interactions within the polymer–lignin–fibre system, scanning electron microscopy (SEM) to examine fibre/matrix interfacial morphology and fracture characteristics, thermogravimetric analysis (TGA) to determine thermal stability and residual char formation, dynamic mechanical analysis (DMA) to assess storage modulus, damping behaviour and glass-transition characteristics, and simultaneous three-point flexural–piezoresistive testing to evaluate flexural strength, flexural modulus, normalized electrical resistance variation and strain-sensing response for structural health monitoring applications.

RESULTS AND DISCUSSION

The influence of kraft lignin incorporation on the physicochemical, interfacial, thermal, viscoelastic and mechanical behaviour of jute-fibre-reinforced bio-epoxy composites was systematically evaluated. Particular emphasis was placed on changes occurring within the polymer matrix and at the fibre-polymer interphase because these regions principally govern stress transfer, damage initiation and long-term performance of natural-fibre polymer composites. The reference jute/bio-epoxy composite, JE, was compared with composites containing 2, 4 and 6 wt.% lignin, designated JE-L2, JE-L4 and JE-L6, respectively. The multifunctional formulation JE-L4C contained 4 wt.% lignin together with 0.5 wt.% MWCNTs.

The combined results demonstrate that lignin does not behave simply as an inert particulate filler. Instead, at appropriate loading it modifies the local polymer network, alters the chemical environment surrounding the hydroxyl-rich jute fibres, restricts segmental mobility of the cured epoxy and influences fibre wetting and stress transfer. However, these advantages are dependent upon maintaining adequate lignin dispersion. Increasing the lignin concentration beyond an optimum range increases the probability of particle–particle interaction, local agglomeration and resin-viscosity-induced incomplete fibre impregnation [10, 11]. The behaviour of the composites can, therefore, be interpreted in terms of a balance between interphase strengthening and polymer-network restriction at moderate lignin loading and agglomeration, heterogeneous curing and incomplete wetting at excessive loading.

FTIR Analysis: Polymer–Lignin–Fibre Interactions

FTIR spectroscopy was employed to identify changes in the chemical environment of the bio-epoxy matrix after incorporation of jute fibres and kraft lignin. The spectra of the composites retained the principal absorption features associated with the cured epoxy network, lignocellulosic reinforcement and aromatic lignin structure, indicating that composite fabrication did not produce severe chemical degradation of any constituent.

A broad absorption band in the approximately 3200–3600 cm^{-1} region was assigned primarily to O–H stretching vibrations originating from cellulose and hemicellulose in jute, phenolic/aliphatic hydroxyl groups of lignin and hydroxyl functionalities generated during epoxy curing. The O–H band occurred at approximately 3338 cm^{-1} for JE and shifted to 3332, 3325 and 3328 cm^{-1} for JE-L2, JE-L4 and JE-L6, respectively. The approximately 13 cm^{-1} downward shift from JE to JE-L4 suggests strengthening of intermolecular interactions and modification of the hydrogen–bonding environment within the polymer–lignin–fibre system rather than the formation of a completely new polymeric phase.

The bands observed at approximately 2923 and 2854 cm^{-1} were attributed to asymmetric and symmetric C-H stretching vibrations of aliphatic $–CH_2$ and $–CH_3$ groups. Aromatic skeletal vibrations associated predominantly with kraft lignin became more distinguishable at approximately 1601 and 1512 cm^{-1}, particularly for JE-L4 and JE-L6. Bands in the 1030–1260 cm^{-1} region were associated with overlapping C–O and C–O–C stretching vibrations from lignin, cellulose and ether-containing segments of the cured epoxy network. The characteristic oxirane-ring-associated absorption near approximately 914 cm^{-1} was only weakly detectable after curing. The reduced intensity of this band for all compositions indicates substantial epoxy-ring opening during curing and suggests that

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incorporation of lignin up to 6 wt.% did not prevent formation of the cross-linked epoxy network [12–14]. The FTIR spectra of JE, JE-L2, JE-L4, JE-L6 and JE-L4C composites are presented in Figure 2. The most pronounced spectral modifications were observed for JE-L4 in the 3200–3600 cm^{-1} and 1030–1260 cm^{-1} regions. Rather than indicating formation of a completely new covalent structure, these changes are more appropriately associated with increased secondary interactions, particularly hydrogen bonding among phenolic hydroxyl groups of lignin, hydroxyl-rich jute surfaces and polar functionalities of the cured epoxy.

Such interactions are significant because the interphase of a natural-fibre composite is normally affected by the chemical dissimilarity between hydrophilic lignocellulosic reinforcement and the comparatively less hydrophilic polymer matrix. Properly dispersed lignin can modify the local chemical environment of the epoxy adjacent to the jute surface. The observed spectral changes, therefore, support improved intermolecular association and potentially more effective stress transfer through the fibre–matrix interphase. For JE-L6, the O–H band shifted slightly back toward higher wavenumber, from 3325 cm^{-1} for JE-L4 to 3328 cm^{-1}. Although the difference is relatively small, when considered together with the mechanical and morphological results it suggests that increasing lignin beyond 4 wt.% did not continuously strengthen polymer–filler interactions. At higher lignin concentration, increased lignin–lignin association and locally heterogeneous filler-rich regions become increasingly probable.

For JE-L4C, incorporation of 0.5 wt.% MWCNTs did not introduce prominent new FTIR absorption bands because of the low nanotube concentration and the relatively weak infrared response of graphitic carbon structures [15]. The principal O–H band was observed at approximately 3323 cm^{-1}, representing only a small additional displacement relative to JE-L4 and suggesting modification of the local polymer environment rather than formation of new chemical functionality.

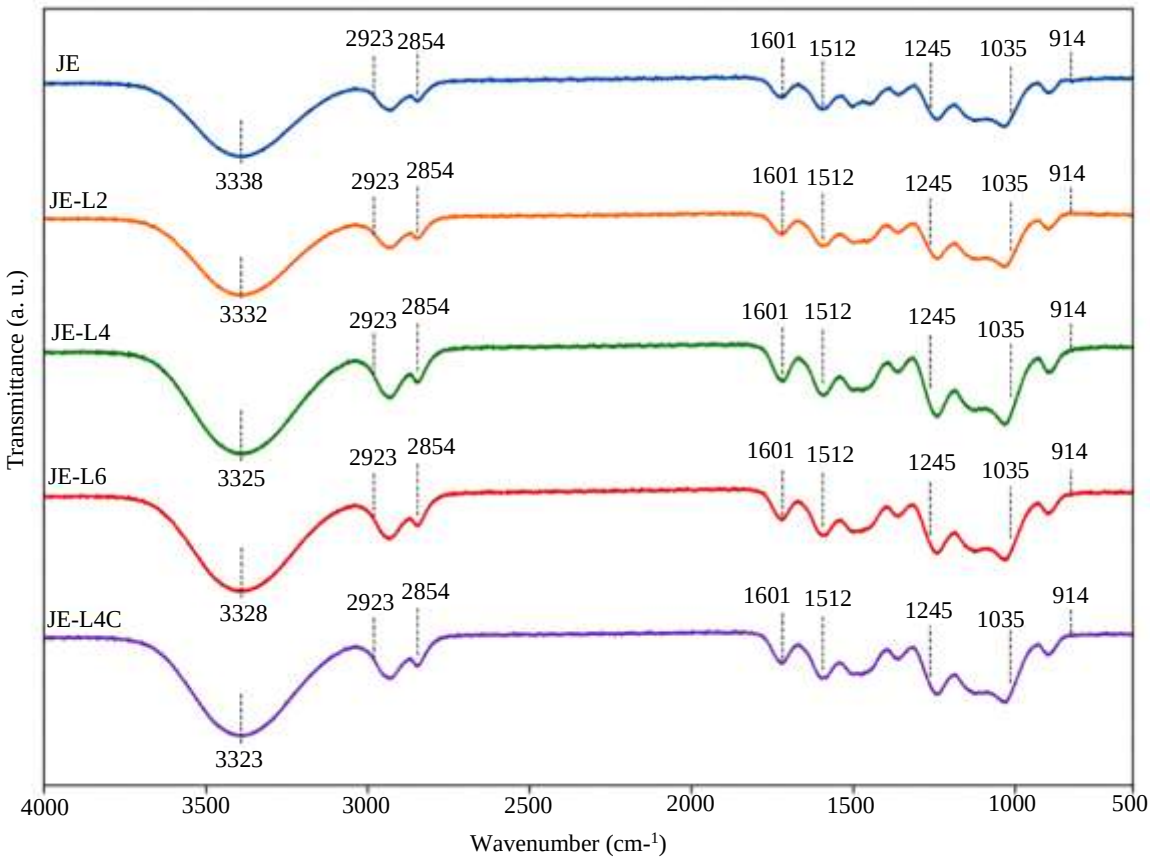


Figure 2. FTIR spectra of JE, JE-L2, JE-L4, JE-L6, and JE-L4C composites.

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SEM Analysis: Matrix Morphology and Fibre–Matrix Interphase

SEM examination revealed clear differences in fracture morphology with lignin concentration. The fracture surface of the reference JE composite exhibited relatively clean fibre pull-out, distinct fibre–matrix interfacial gaps, comparatively smooth epoxy-rich fracture regions and occasional microvoids. Several jute fibres were visible with limited residual polymer adhering to their surfaces. These features indicate that interfacial separation contributed significantly to failure of the reference composite.

The introduction of 2 wt.% lignin in JE-L2 resulted in a noticeably more irregular fracture morphology. Greater quantities of fractured polymer remained attached to the jute fibre surfaces, while the apparent width and frequency of interfacial gaps decreased. These observations suggest improved polymer wetting and increased resistance to simple fibre pull-out. The JE-L4 composite displayed the most compact and integrated fibre–matrix morphology. Fractured fibres remained substantially covered by adhered epoxy, clean fibre pull-out was considerably reduced, and large interfacial cavities were uncommon [16]. The fracture path appeared to pass through the surrounding polymer and interphase rather than propagating exclusively along the fibre boundary. This morphology suggests stronger mechanical interlocking and improved fibre-to-polymer stress transfer. The SEM micrographs of the fracture surfaces of JE, JE-L2, JE-L4, JE-L6, and JE-L4C composites are shown in Figure 3.

The improvement observed at 4 wt.% lignin can be associated with the combined effects of lignin dispersion, polymer-network modification and improved interfacial interaction. The aromatic structure of lignin provides relatively rigid domains within the epoxy, while its hydroxyl-containing surface chemistry facilitates secondary interaction with both the lignocellulosic fibre and the cured polymer network. In contrast, JE-L6 exhibited localized lignin-rich agglomerates, small microvoids, regions of incomplete wetting around jute bundles and occasional particle/matrix debonding. The agglomerated regions ranged approximately from 4 to 12 micrometres in apparent lateral dimension in the examined fracture fields. Such regions were not dominant throughout the structure but were substantially more frequent than in JE-L4.

The deterioration in morphology at 6 wt.% can be associated with an increase in uncured resin viscosity and reduced average separation between lignin particles. Increased viscosity restricts penetration of the resin into small spaces within individual jute yarns, whereas closer filler spacing promotes lignin–lignin association [17]. These heterogeneous regions subsequently act as local stress concentrators during mechanical loading. JE-L4C showed a comparatively rough and highly tortuous fracture morphology. Large MWCNT bundles were not evident over most of the examined surface, suggesting reasonably effective dispersion at the selected 0.5 wt.% nanotube concentration. Localized nanoscale/microscale polymer bridging and crack deflection were visible in several regions. This morphology is consistent with a nanotube network capable of interacting mechanically with the surrounding epoxy while simultaneously providing electrically responsive pathways.

The SEM observations, therefore, establish a direct relationship between filler concentration, polymer morphology, fibre wetting, interfacial quality and mechanical response. The results show that maximum renewable filler content does not necessarily correspond to maximum composite performance; instead, the most favourable behaviour occurred when lignin concentration was sufficiently high to modify the polymer/interphase but sufficiently low to avoid extensive agglomeration.

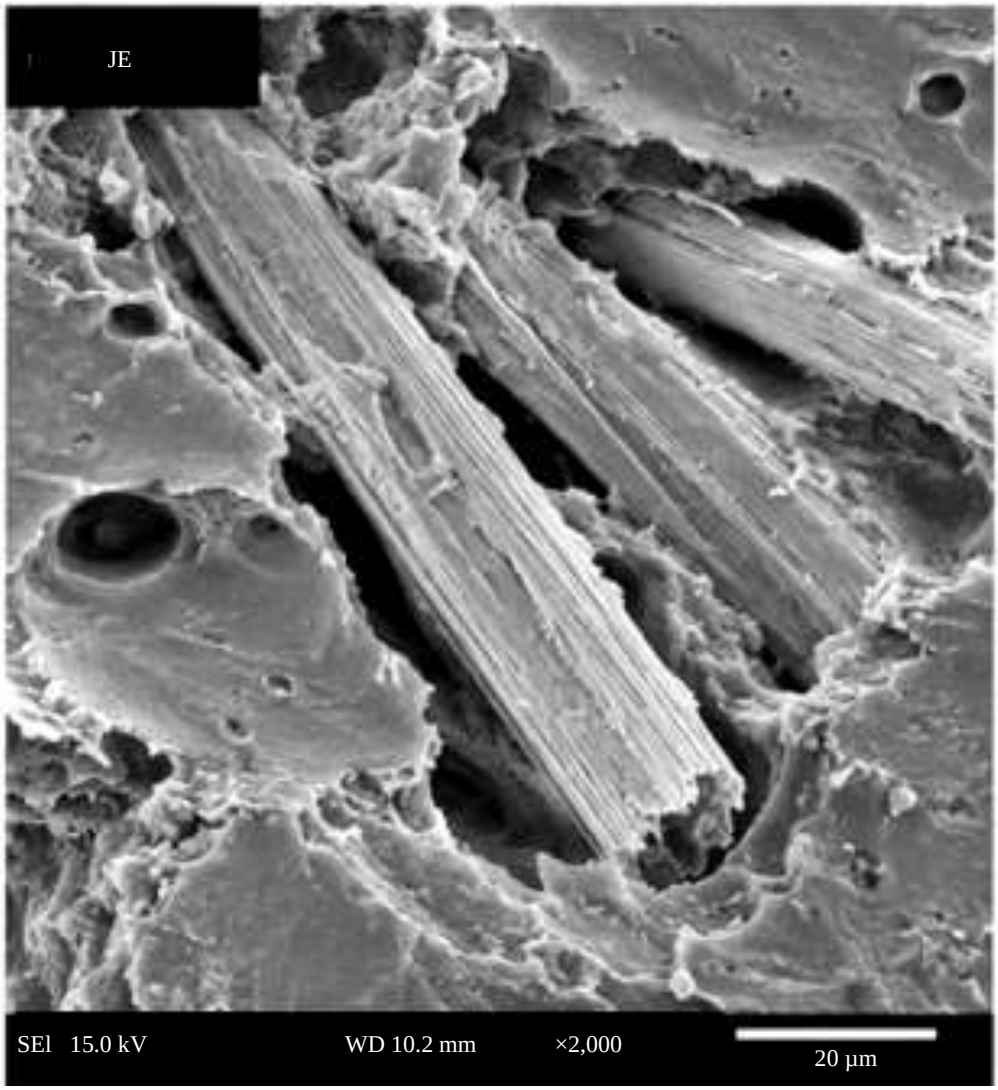
The fracture-surface observations also provide a mechanistic explanation for the measured flexural behaviour. In the reference JE composite, relatively clean fibre pull-out, interfacial gaps and limited residual polymer on the jute surface indicate that interfacial debonding and fibre pull-out contributed strongly to failure. With increasing lignin content up to 4 wt.%, the fracture morphology became progressively more integrated, with greater polymer attachment to the fibres, reduced clean pull-out and fewer large interfacial cavities. In JE-L4, the fracture path more frequently traversed the surrounding polymer and interphase rather than propagating preferentially along the fibre boundary,

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indicating improved interfacial stress transfer. These observations are consistent with the increase in flexural strength from 92.4 MPa for JE to 118.6 MPa for JE-L4 and the corresponding increase in flexural modulus from 4.18 to 5.12 GPa. At 6 wt.% lignin, the appearance of lignin-rich agglomerates, microvoids, incomplete fibre wetting and local particle/matrix debonding introduced stress-concentration sites, which explains the reduction in flexural strength to 101.3 MPa. The JE-L4C fracture surface further exhibited a rougher and more tortuous morphology, together with local polymer bridging and crack deflection, while large MWCNT agglomerates were not evident over most of the examined surface. These features are consistent with the slight additional increase in flexural strength to 121.9 MPa and modulus to 5.36 GPa. The fracture analysis, therefore, establishes a direct correlation between failure mechanism and mechanical response: interfacial separation dominates the weaker reference composite, improved fibre-matrix adhesion supports the enhanced performance at 4 wt.% lignin, and agglomeration-induced defects account for the decline observed at excessive lignin loading.

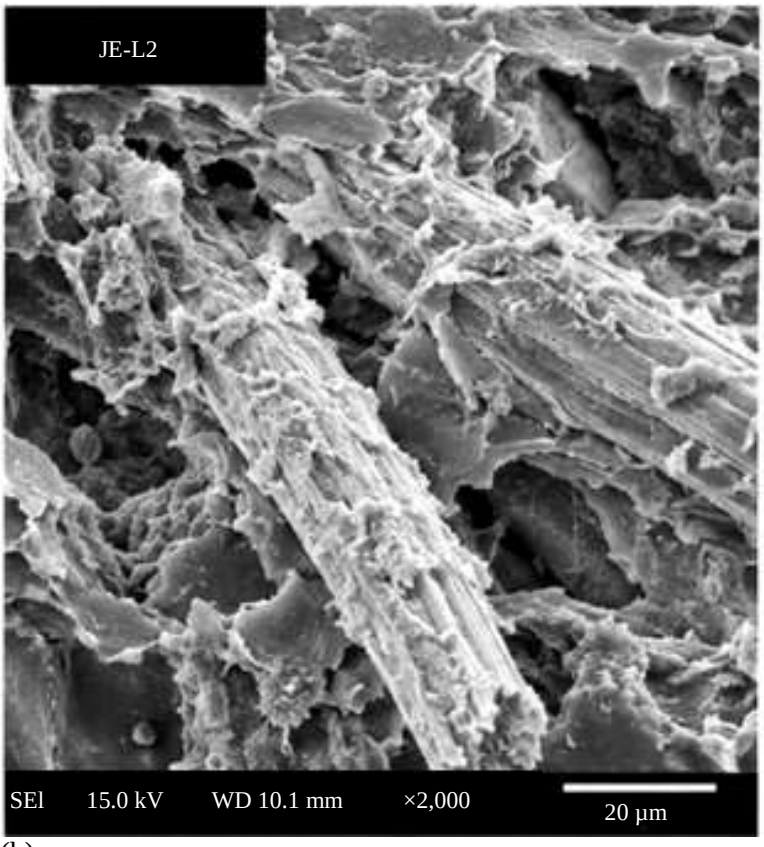
Thermal Stability and Degradation Behaviour

TGA was conducted to determine the influence of lignin modification on the thermal stability of the jute/bio-epoxy composites. All compositions displayed a relatively small initial mass loss below approximately 150°C followed by a major degradation region extending from approximately 250 to 430°C. The initial mass loss below 150°C was approximately 3.8% for JE, 3.5% for JE-L2, 3.2% for JE-L4 and 3.6% for JE-L6.

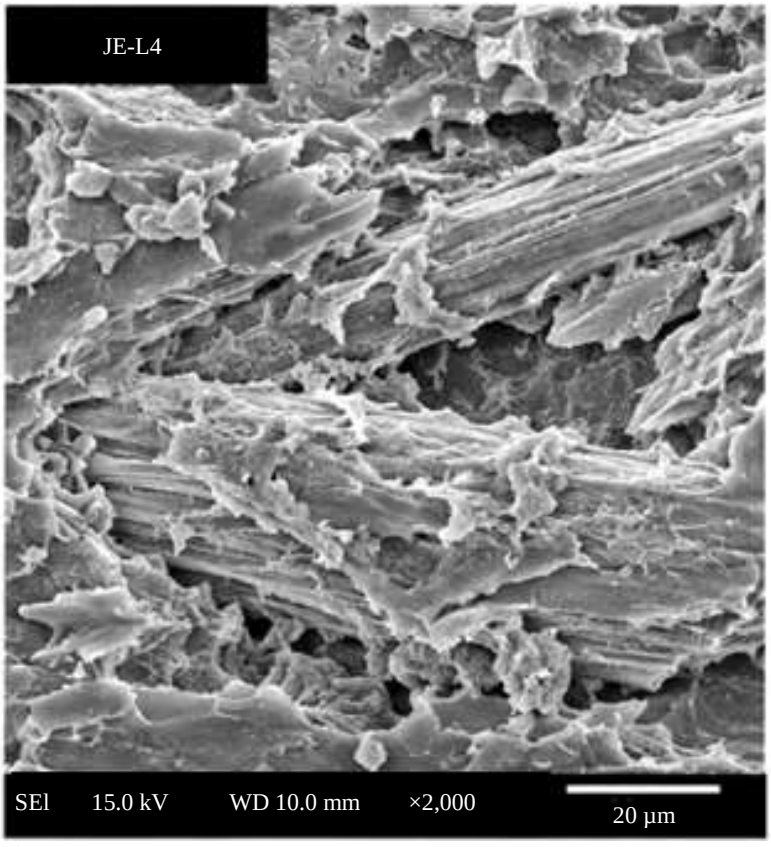


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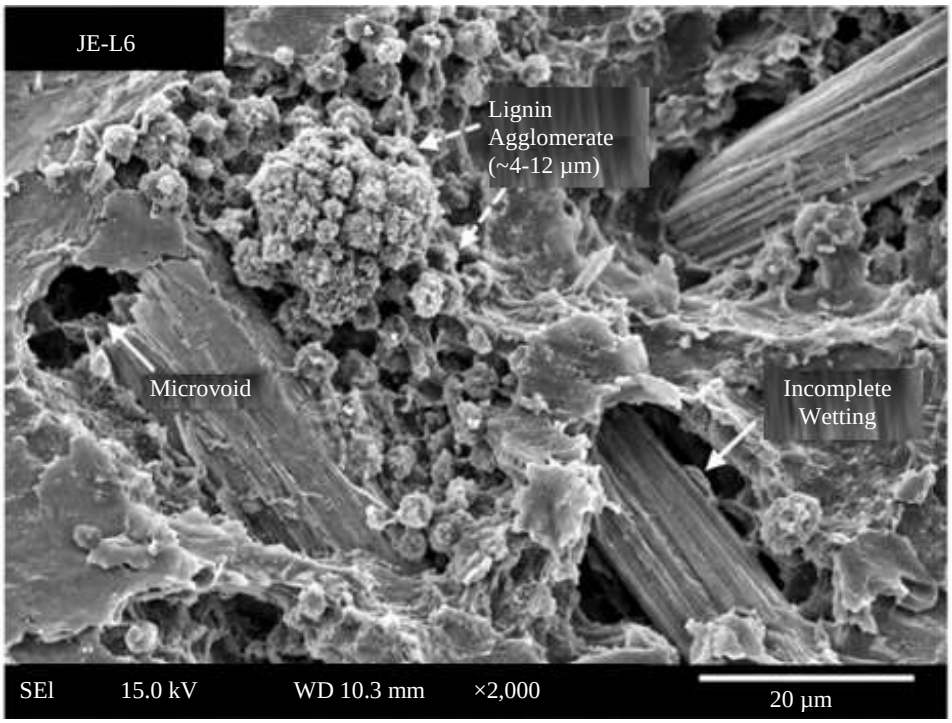


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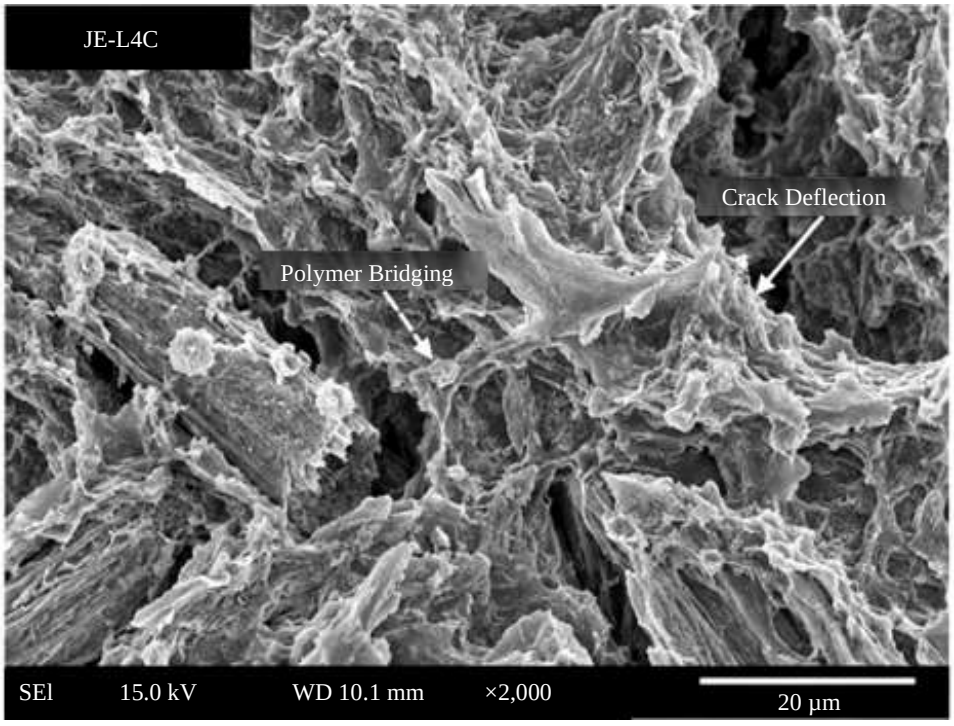


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(d)



(e)

Figure 3. SEM micrographs of fracture morphology for JE, JE-L2, JE-L4, JE-L6 and JE-L4C composites.

The slightly reduced low-temperature mass loss of JE-L4 may indicate comparatively effective encapsulation of the jute fibres by the modified polymer matrix and reduced availability of loosely bound moisture. The temperature corresponding to 5% mass loss, T5, increased from approximately 256°C for JE to 262°C for JE-L2 and 271°C for JE-L4. For JE-L6, T5 decreased slightly to

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approximately 264°C. JE-L4C exhibited a T5 of approximately 274°C. The principal maximum degradation temperature, T_{max}, followed a similar trend. T_{max} was approximately 358°C for JE, 364°C for JE-L2, 372°C for JE-L4, 365°C for JE-L6 and 375°C for JE-L4C. The TGA curves of JE, JE-L2, JE-L4, JE-L6, and JE-L4C composites are presented in Figure 4.

The approximately 14°C increase in T_{max} from JE to JE-L4 indicates that moderate lignin incorporation improves resistance of the polymer composite to thermally activated degradation. This behaviour can be associated with the aromatic structure of lignin, increased intermolecular interactions and restricted mobility of thermally activated polymer segments [18, 19]. The high-temperature residual char measured at 700°C was approximately 14.8% for JE, 17.6% for JE-L2, 21.9% for JE-L4, 24.7% for JE-L6 and 23.1% for JE-L4C.

The progressive increase in char residue with lignin loading is consistent with the relatively high aromatic carbon content of kraft lignin and its tendency to undergo condensation and carbonization during thermal decomposition. The JE-L6 composite, therefore, produced the largest residual mass despite not exhibiting the highest degradation onset temperature. This distinction is important. Higher residual char does not necessarily imply greater thermal stability throughout the entire degradation process. JE-L6 contains more lignin capable of producing carbonaceous residue, but agglomeration and polymer-network heterogeneity can initiate local degradation at somewhat lower temperature [19]. In contrast, JE-L4 combines relatively high thermal stability with substantial char generation.

The addition of 0.5 wt.% MWCNTs to JE-L4 increased T_{max} from approximately 372 to 375°C and increased the residual char from 21.9 to 23.1%. These relatively modest changes are consistent with the low concentration of nanotubes. The thermally stable graphitic network and restriction of polymer mobility near the nanotube surface can nevertheless slightly delay matrix degradation.

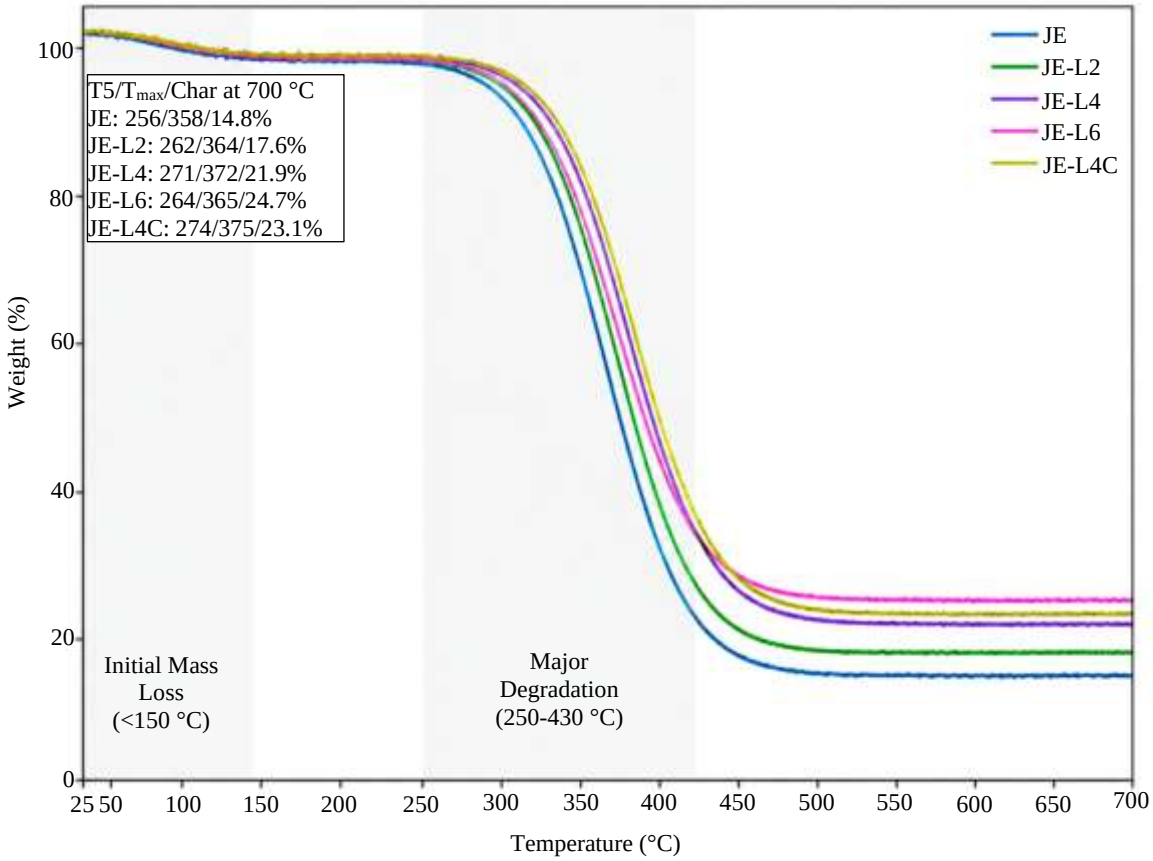


Figure 4. TGA curves of JE, JE-L2, JE-L4, JE-L6, and JE-L4C composites.

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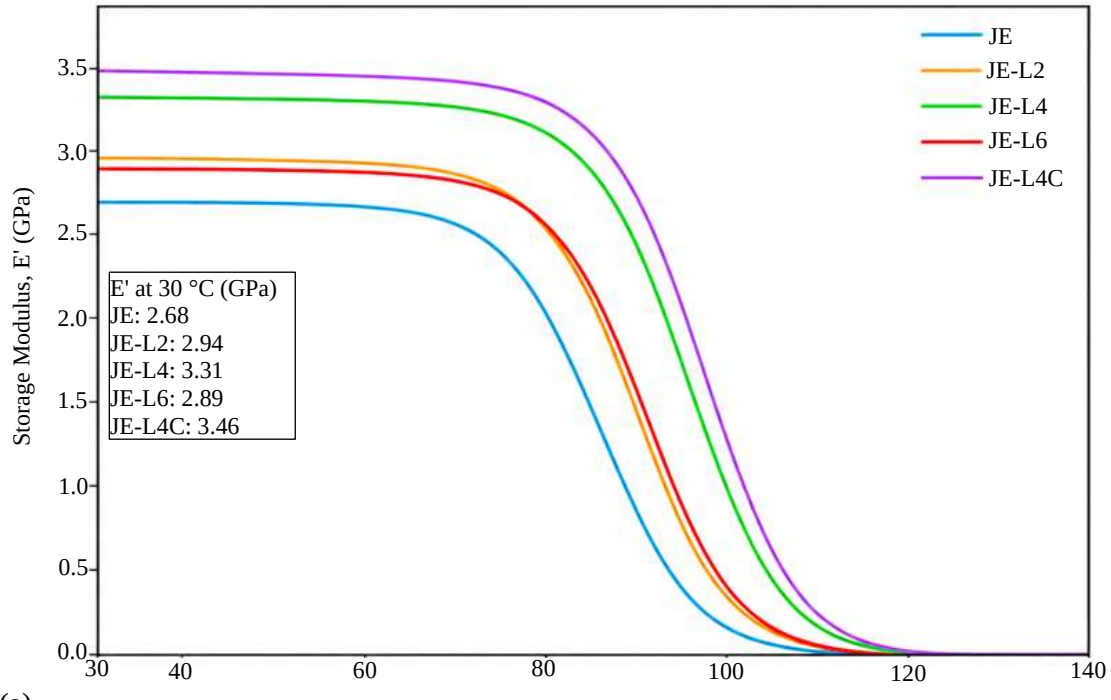
Dynamic Mechanical Behaviour

DMA was used to evaluate the influence of lignin and MWCNT incorporation on the viscoelastic behaviour and molecular mobility of the cured polymer network. At 30°C, the storage modulus, E', of the reference JE composite was approximately 2.68 GPa. The storage modulus increased to 2.94 GPa for JE-L2 and reached 3.31 GPa for JE-L4. Increasing the lignin concentration to 6 wt.% reduced the storage modulus to approximately 2.89 GPa, while JE-L4C exhibited the highest value of approximately 3.46 GPa.

Thus, incorporation of 4 wt.% lignin increased the room-temperature storage modulus by approximately 23.5% relative to the reference JE composite. The increase indicates greater elastic constraint of the epoxy-rich regions and more efficient stress transmission through the modified polymer-fibre interphase. The improvement is associated with the rigid aromatic structure of lignin and its ability to restrict segmental deformation of the cured epoxy when adequately dispersed [20, 21]. The SEM observations support this interpretation because JE-L4 also exhibited reduced fibre pull-out and improved polymer attachment to the jute surface.

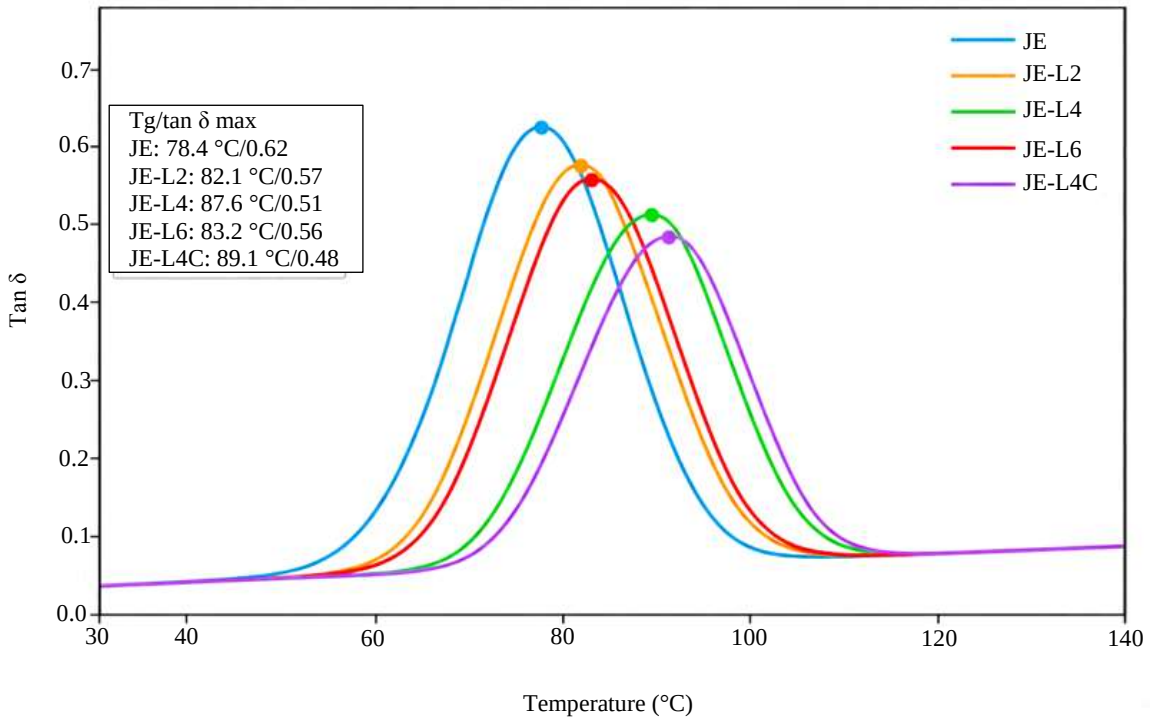
The decline in E' from 3.31 GPa for JE-L4 to 2.89 GPa for JE-L6 indicates that the stiffening effect of lignin is offset when filler loading becomes sufficiently high to promote agglomeration and incomplete fibre impregnation. The glass-transition temperature, determined from the tan delta maximum, was approximately 78.4°C for JE, 82.1°C for JE-L2, 87.6°C for JE-L4, 83.2°C for JE-L6 and 89.1°C for JE-L4C. The increase in Tg from 78.4°C for JE to 87.6°C for JE-L4 corresponds to an increase of approximately 9.2°C. This shift provides evidence that segmental mobility of the epoxy network was increasingly restricted by polymer-lignin and interfacial interactions. The storage modulus and tan δ responses of the composites are presented in Figure 5.

The maximum tan delta value decreased from approximately 0.62 for JE to 0.57 for JE-L2 and 0.51 for JE-L4. JE-L6 showed a tan delta maximum of approximately 0.56, whereas JE-L4C exhibited a value of approximately 0.48. The reduced tan delta magnitude for JE-L4 indicates that a larger fraction of the polymer was mechanically constrained and, therefore, less able to dissipate energy through unrestricted molecular rearrangement. The slightly greater tan delta of JE-L6 is consistent with increased structural heterogeneity and the presence of polymer-rich and lignin-rich regions with differing local mobilities.



(a)

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(b)

Figure 5. DMA curves of JE, JE-L2, JE-L4, JE-L6, and JE-L4C composites.

For JE-L4C, the storage modulus increased from 3.31 to 3.46 GPa and Tg increased from 87.6 to 89.1°C. These relatively modest but measurable increases suggest that the high-aspect-ratio nanotubes further restricted local epoxy-chain mobility without substantially disrupting the lignin-modified polymer network. Taken together, the DMA results provide molecular-level confirmation that the optimum lignin content is associated with suppression of excessive polymer-chain motion and more effective interfacial stress transfer.

Flexural Properties of the Lignin-Modified Polymer Composites

The reference JE composite exhibited a flexural strength of approximately 92.4 MPa and a flexural modulus of approximately 4.18 GPa. Incorporation of 2 wt.% lignin increased the flexural strength to approximately 103.7 MPa and the flexural modulus to 4.54 GPa. The maximum values among the lignin-only composites were obtained for JE-L4, which exhibited a flexural strength of approximately 118.6 MPa and a flexural modulus of approximately 5.12 GPa.

Increasing the lignin content to 6 wt.% reduced the flexural strength to approximately 101.3 MPa and the modulus to approximately 4.63 GPa. The increase from 92.4 MPa for JE to 118.6 MPa for JE-L4 represents an improvement in flexural strength of approximately 28.4%. Similarly, the increase in flexural modulus from 4.18 to 5.12 GPa corresponds to an improvement of approximately 22.5%.

The improvement at 4 wt.% lignin can be attributed primarily to modification of the epoxy matrix and fibre–matrix interphase. Under flexural loading, matrix microcracks tend to initiate within resin-rich regions or at interfacial defects.

Improved polymer–fibre interaction enables a greater proportion of the applied stress to be transferred into the woven jute reinforcement before interfacial debonding occurs [22]. The reduction in flexural strength from 118.6 MPa for JE-L4 to 101.3 MPa for JE-L6 corresponds to a decrease of approximately 14.6%. This decline agrees well with the SEM observations of lignin-rich agglomerates and incomplete wetting at the highest lignin concentration.

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The behaviour, therefore, demonstrates the characteristic optimum-filler phenomenon encountered in particulate-modified polymer composites. At moderate concentration, decreasing filler separation strengthens polymer–filler interactions and restricts matrix deformation. Beyond an optimum concentration, agglomeration and processing-induced defects begin to dominate the response.

The self-sensing JE-L4C composite exhibited a flexural strength of approximately 121.9 MPa and flexural modulus of approximately 5.36 GPa. Relative to JE-L4, incorporation of 0.5 wt.% MWCNTs, therefore, increased flexural strength by approximately 2.8% and modulus by approximately 4.7%. Compared with the reference JE composite, JE-L4C exhibited approximately 31.9% higher flexural strength and approximately 28.2% higher flexural modulus. The comparative flexural strength and modulus are shown in Figure 6.

The relatively small but positive change after MWCNT incorporation indicates that the nanotube concentration was sufficiently low to avoid severe agglomeration while still providing additional nanoscale restriction of polymer deformation. The flexural failure strain was approximately 3.28% for JE, 3.16% for JE-L2, 3.02% for JE-L4, 3.08% for JE-L6 and 2.94% for JE-L4C. The gradual decrease in failure strain associated with the stiffer formulations is consistent with increasing matrix constraint [23]. This change does not necessarily indicate inferior structural performance because it accompanies substantial increases in strength and modulus.

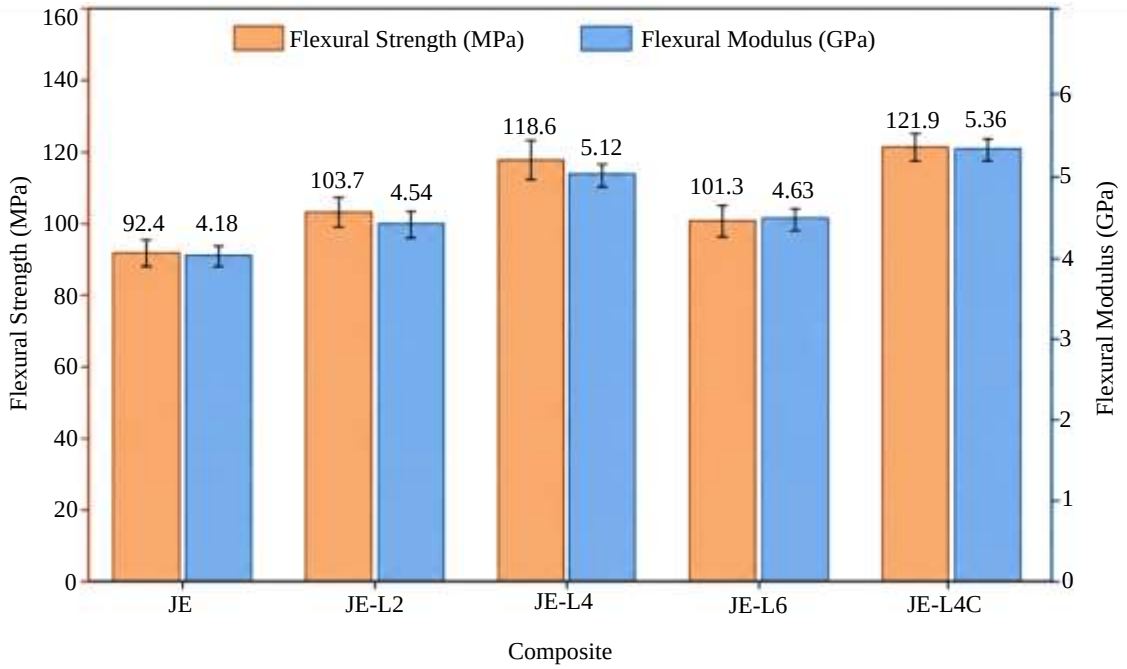


Figure 6. Flexural strength and modulus of the developed composites.

Piezoresistive Response and In-Situ Damage Monitoring

The JE-L4C composite exhibited a clear and reproducible variation in electrical resistance during three-point flexural deformation. The normalized resistance change was calculated according to $\Delta R/R_0 = (R - R_0)/R_0$, where R_0 represents the initial resistance and R represents the instantaneous resistance during loading. The initial electrical resistance of the JE-L4C specimens was approximately 18.6 k-Ohm, with specimen-to-specimen variation of approximately ±1.4 k-Ohm.

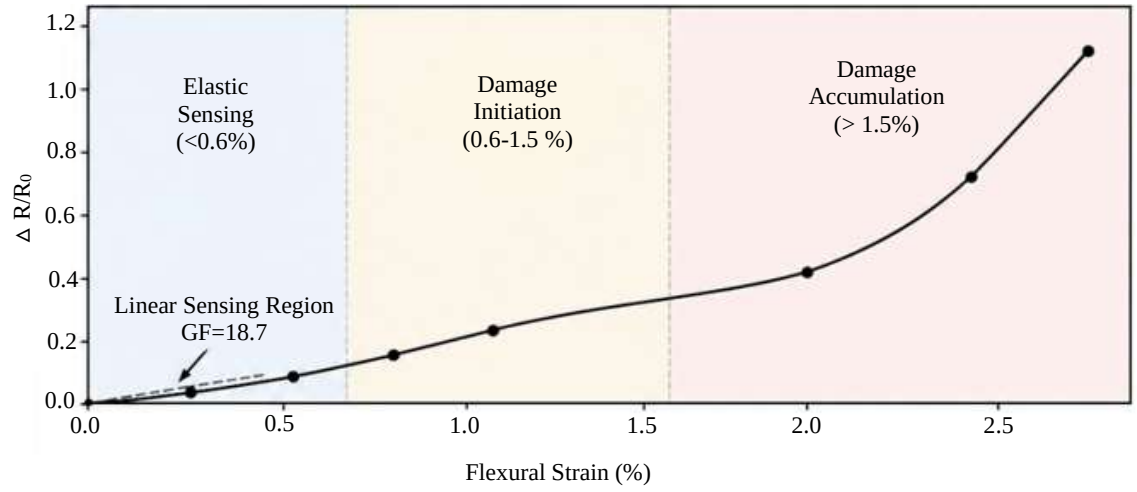
During the early stage of flexural loading, $\Delta R/R_0$ increased approximately monotonically with strain. At a flexural strain of 0.25%, the normalized resistance change was approximately 0.042, while at 0.50% strain it increased to approximately 0.096. At 0.75% strain, ΔR/R0 reached approximately 0.158,

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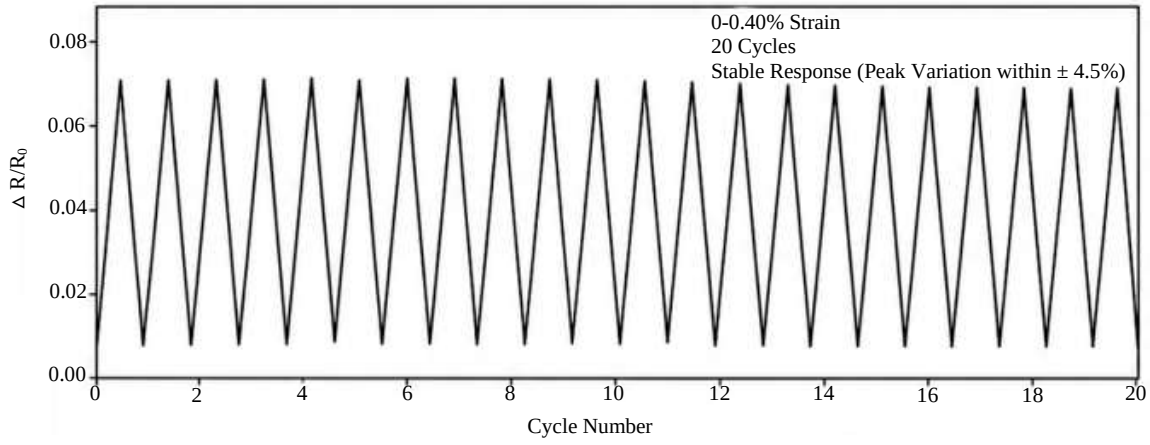
and at approximately 1.00% strain it increased to approximately 0.231. The sensitivity of the composite was expressed using the gauge factor, $GF = (\Delta R / R_0)/\varepsilon$, where ε represents the mechanical strain. Within the approximately linear low-strain interval between 0.10 and 0.50%, the calculated effective gauge factor was approximately 18.7. This comparatively high strain sensitivity originates from deformation-induced changes in the nanoscale MWCNT conductive network.

When the epoxy matrix is deformed, the average distance between adjacent nanotubes changes and some direct conductive contacts become partially separated. Because electron tunnelling resistance is strongly dependent on nanotube separation, relatively small deformation of the polymer produces a measurable change in electrical resistance [24]. The piezoresistive response of the JE-L4C composite during flexural deformation is presented in Figure 7.

At strains below approximately 0.6%, the electrical response remained substantially reversible during loading and unloading. The residual normalized resistance after unloading was less than approximately 0.012, indicating that most conductive pathways were restored after elastic recovery of the polymer matrix. At strains between approximately 0.6 and 1.2%, a progressively larger residual resistance offset was observed. The baseline ΔR/R0 increased from approximately 0.015 to 0.041 after unloading in this region. This change indicates initiation of irreversible microstructural damage rather than purely elastic rearrangement of the nanotube network.

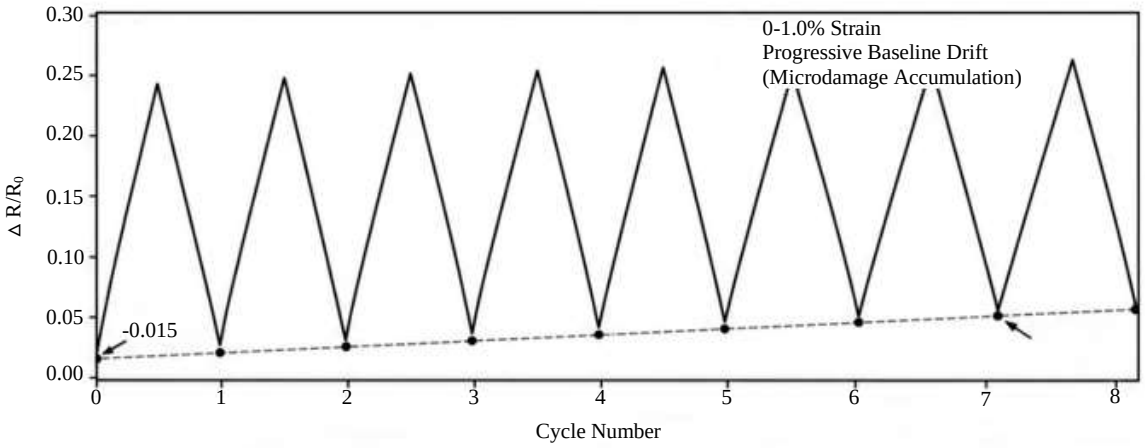


(a)



(b)

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(d)

Figure 7. Piezoresistive response of the JE-L4C composite.

Beyond approximately 1.5% flexural strain, the resistance response became increasingly nonlinear. $\Delta R/R_0$ increased to approximately 0.42 at 2.0% strain and approximately 0.73 at 2.5% strain. Near final flexural failure, normalized resistance changes exceeding approximately 1.1 were observed. The sensing behaviour can, therefore, be divided into three regimes. In the first, or elastic sensing regime, reversible nanotube separation and tunnelling-distance changes dominate. In the second, or damage-initiation regime, matrix microcracking and local fibre–matrix debonding begin to permanently disrupt conductive pathways. In the third, or damage-accumulation regime, crack propagation, interfacial separation and fibre damage produce large irreversible resistance changes. During repeated low-strain cycling between approximately 0 and 0.40% strain, the peak-to-peak variation in $\Delta R/R_0$ remained within approximately ±4.5% over 20 cycles, demonstrating comparatively stable strain-sensing behaviour. When the maximum cycling strain was increased to approximately 1.0%, progressive baseline drift became observable, consistent with accumulating microdamage.

The electrical response, therefore, confirms that JE-L4C functions as an intrinsically self-sensing polymer composite rather than as a conventional composite carrying an externally attached sensor. Importantly, the conductive response should be attributed primarily to the MWCNT network [25]. Lignin contributes indirectly by modifying the polymer environment and potentially influencing nanotube distribution, but lignin itself is not responsible for establishment of the conductive network.

Structure–Property Relationship of the Composite System

The combined FTIR, SEM, TGA, DMA, flexural and piezoresistive results reveal a consistent concentration-dependent structure–property relationship (Figure 8). The reference JE composite exhibited a flexural strength of 92.4 MPa, flexural modulus of 4.18 GPa, storage modulus of 2.68 GPa and Tg of 78.4°C. Its SEM morphology showed comparatively greater fibre pull-out and interfacial separation, indicating that fibre–matrix adhesion represented an important limitation.

Addition of 2 wt.% lignin produced moderate improvements, increasing flexural strength to 103.7 MPa, storage modulus to 2.94 GPa and Tg to 82.1°C. These changes indicate that even relatively low lignin loading begins to modify the cured polymer network and surrounding interphase [26]. The JE-L4 composition exhibited the most favourable overall response among the lignin-only composites. Flexural strength increased to 118.6 MPa, modulus increased to 5.12 GPa, storage modulus increased to 3.31 GPa, Tg increased to 87.6°C and Tmax increased to 372°C. The approximately 28.4% enhancement in flexural strength and 22.5% increase in flexural modulus relative to JE are consistent with the improved interfacial morphology and increased polymer-network constraint identified using SEM and DMA.

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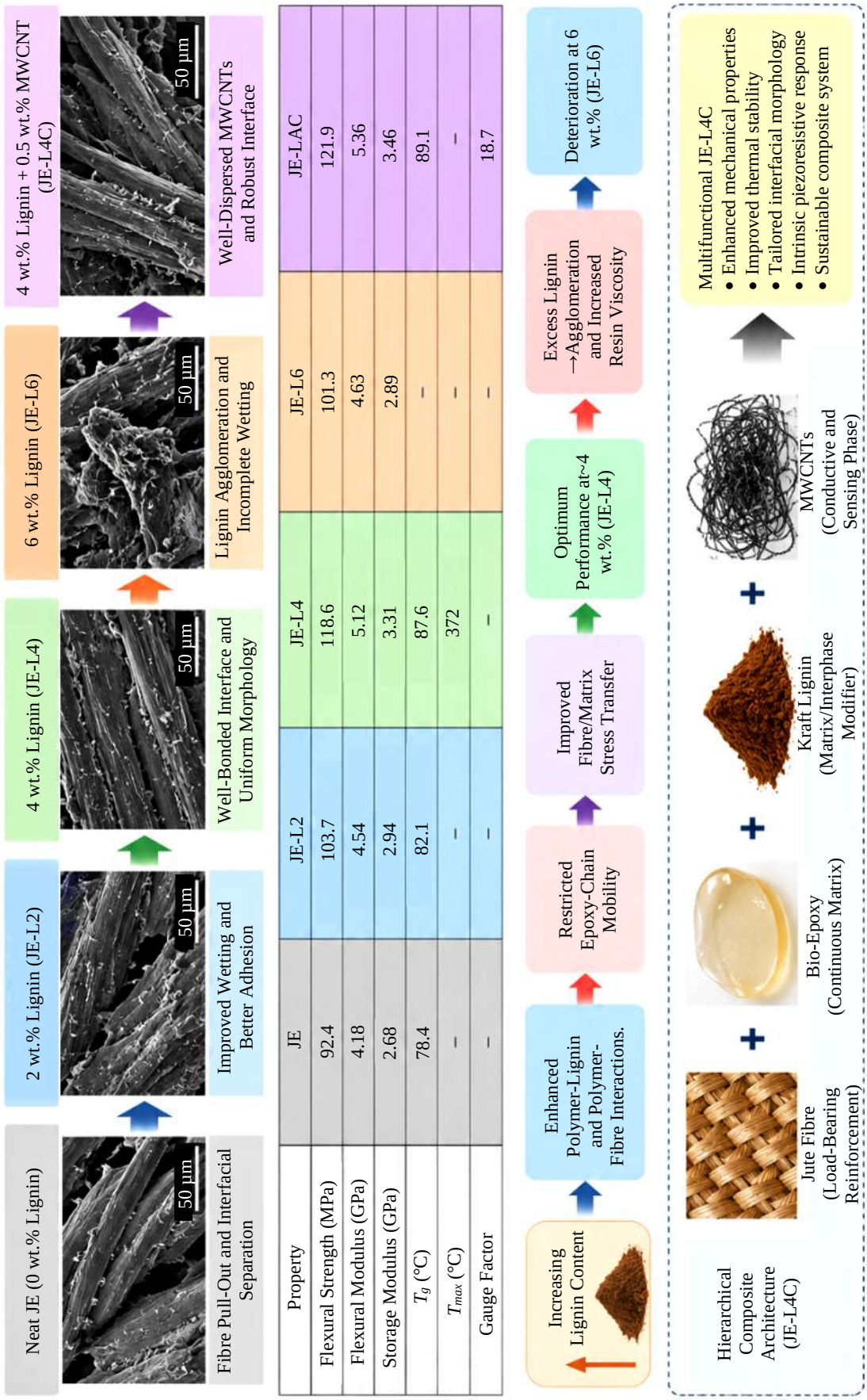


Figure 8. Structure–property relationship of the developed composite system.

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At 6 wt.% lignin, the beneficial trend was reversed. Flexural strength declined to 101.3 MPa and storage modulus to 2.89 GPa, despite the larger char residue produced during TGA. SEM observations of filler agglomeration and incomplete wetting provide a microstructural explanation for this reduction. The overall lignin concentration dependence can, therefore, be represented as: increasing lignin concentration –> increased polymer-lignin and polymer–fibre interactions –> restricted epoxy-chain mobility –> improved fibre/matrix stress transfer –> optimum performance at approximately 4 wt.% –> increasing lignin–lignin interaction and resin viscosity –> agglomeration/incomplete wetting –> deterioration at 6 wt.%. The addition of 0.5 wt.% MWCNTs to JE-L4 further increased the flexural strength to 121.9 MPa, storage modulus to 3.46 GPa and Tg to 89.1°C while simultaneously generating a gauge factor of approximately 18.7.

The multifunctional JE-L4C system can consequently be described in terms of a hierarchical polymer–composite architecture: jute fibre as the primary load-bearing reinforcement; bio-epoxy as the continuous stress-transfer polymer phase; kraft lignin as the renewable matrix/interphase modifier; and MWCNTs as the nanoscale conductive and electrically responsive phase. The material response, therefore, results from interaction among constituents at several length scales rather than from the independent contribution of any individual component.

Relevance to Sustainable Housing Polymer Composites

The present results demonstrate that the developed system is most appropriately classified as a multifunctional natural-fibre-reinforced polymer composite. The use of jute reduces dependence on conventional synthetic reinforcement, while lignin provides partial renewable modification of the polymer phase. Importantly, lignin does not function only as a sustainability additive; the observed changes in T_g, storage modulus, thermal degradation temperature and flexural performance demonstrate a direct contribution to polymer-network and interfacial behaviour. The JE-L4 formulation provided the most favourable lignin-only response, with a flexural strength of 118.6 MPa, flexural modulus of 5.12 GPa and T_g of 87.6°C. The addition of 0.5 wt.% MWCNTs produced JE-L4C with a flexural strength of 121.9 MPa, modulus of 5.36 GPa and effective strain gauge factor of approximately 18.7.

This combination of structural performance and deformation-dependent electrical resistance indicates potential for semi-structural housing applications such as modular partition-panel skins, internal wall panels, ceiling panels, door panels, cladding elements, and sandwich-panel facings. The material should nevertheless not be considered a direct substitute for highly loaded primary structural members based solely on the present characterization [27]. Long-term moisture ageing, creep, fatigue, fire performance, environmental durability and full-scale component testing would be required for structural qualification. Within the present scope, the principal outcome is the demonstration of a polymer-composite design in which 4 wt.% kraft lignin provides a favourable balance between polymer-network modification and interfacial reinforcement, while 0.5 wt.% MWCNTs introduces an electrically responsive pathway without adversely affecting the structural integrity of the composite.

The developed JE-L4C composite is not intended to replace conventional glass-fibre- or carbon-fibre-reinforced composites in highly loaded primary structural members. Instead, its relevance lies in achieving a useful balance between moderate structural performance, renewable material content and intrinsic sensing capability for semi-structural housing components. The JE-L4C composite exhibited a flexural strength of 121.9 MPa and a flexural modulus of 5.36 GPa, representing improvements of approximately 31.9% and 28.2%, respectively, compared with the unmodified jute/bio-epoxy composite. Although glass- and carbon-fibre composites generally provide higher absolute stiffness and strength, their use in applications – such as internal wall panels, partition panels, ceiling panels, door panels, and sandwich – facings may provide mechanical capacity substantially beyond the level required for service. In contrast, the present material incorporates jute as the principal load-bearing reinforcement, a bio-based epoxy containing approximately 30–35% renewable carbon and kraft lignin as a renewable matrix modifier. Only 0.5 wt.% MWCNTs is

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required to impart electrical functionality. An additional advantage is that the conductive network is integrated into the composite itself, providing strain- and damage-dependent resistance variation without requiring a separate sensing layer. The measured gauge factor of approximately 18.7 demonstrates that the material provides a combination of mechanical reinforcement and intrinsic structural-health-monitoring capability. Therefore, compared with conventional synthetic-fibre composites, the principal advantage of JE-L4C is not maximum mechanical performance alone, but the combined contribution of renewable constituents, reduced dependence on synthetic reinforcement and integrated piezoresistive sensing in a single composite architecture.

The present results also provide an initial indication of the material's suitability for protected indoor housing environments, although a specific service life in years cannot be assigned from the present laboratory-scale characterization. JE-L4C exhibited a glass-transition temperature of approximately 89.1°C, a storage modulus of 3.46 GPa, a 5% mass-loss temperature of approximately 274°C and a maximum degradation temperature of approximately 375°C. These values are substantially above temperatures normally encountered in indoor residential service and indicate that ordinary thermal exposure is unlikely to approach the thermal transition or degradation limits of the composite. In addition, the piezoresistive response was substantially reversible below approximately 0.6% flexural strain, and repeated cycling between 0 and 0.40% strain produced peak-to-peak resistance variation within approximately ±4.5% over 20 cycles, indicating short-term stability of the conductive network under low-level repeated deformation. Nevertheless, long-term durability of natural-fibre polymer composites can be affected by moisture absorption, humidity-temperature cycling, sustained loading, creep, fatigue, ultraviolet exposure and fire conditions. Consequently, the present results should be interpreted as demonstrating the potential suitability of JE-L4C for protected semi-structural housing applications rather than establishing a definitive numerical service life. Long-term environmental ageing and component-level durability assessment would be required before a design service life can be assigned.

CONCLUSION

The present study demonstrates that lignin can function as an effective polymer-matrix and interphase modifier, rather than merely as bio-based filler, in jute/bio-epoxy composites. The main outcomes are summarized below:

- Kraft lignin altered the chemical environment of the cured bio-epoxy matrix and fibre–matrix interphase. The O–H stretching band shifted from 3338 cm^{-1} for JE to 3325 cm^{-1} for JE-L4, supporting enhanced intermolecular and hydrogen–bonding interactions among lignin, jute and the cured epoxy network.
- A lignin content of 4 wt.% produced the most favourable interfacial morphology. JE-L4 showed reduced clean fibre pull-out, improved polymer adhesion and fewer large interfacial cavities, indicating improved fibre wetting and stress transfer. In contrast, 6 wt.% lignin promoted 4–12 μm agglomerated regions, microvoids and incomplete wetting, demonstrating the adverse effect of excessive filler loading on polymer-matrix homogeneity.
- Moderate lignin addition improved thermal performance while increasing char formation. JE-L4 exhibited a T_5 of approximately 271°C and Tmax of 372°C compared with 256°C and 358°C for JE. Residual char at 700°C increased from 14.8% for JE to 21.9% for JE-L4, reflecting the contribution of the aromatic lignin structure to high-temperature carbonaceous residue formation.
- Dynamic mechanical behaviour confirmed restriction of polymer-chain mobility at the optimum lignin concentration. The storage modulus increased from 2.68 GPa for JE to 3.31 GPa for JE-L4, while Tg increased from 78.4 to 87.6°C. These changes indicate greater elastic constraint of the epoxy-rich regions and stronger interaction between the matrix, lignin and reinforcement.
- JE-L4 provided the optimum balance between stiffness, strength and interfacial integrity. Its flexural strength increased from 92.4 to 118.6 MPa and its flexural modulus from 4.18 to 5.12 GPa, corresponding to improvements of approximately 28.4% and 22.5%, respectively. Increasing lignin to 6 wt.% reduced these benefits because agglomeration and higher resin viscosity impaired stress transfer and fibre impregnation.

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- MWCNT incorporation converted the lignin-modified polymer matrix into a multifunctional self-sensing phase without sacrificing structural performance. JE-L4C achieved a flexural strength of 121.9 MPa, flexural modulus of 5.36 GPa, storage modulus of 3.46 GPa and Tg of 89.1°C, together with a gauge factor of approximately 18.7.
- The developed composite exhibits a hierarchical polymer-composite architecture in which jute provides primary reinforcement, bio-epoxy acts as the continuous stress-transfer phase, lignin modifies the polymer network and fibre/matrix interphase, and MWCNTs provide electrically conductive pathways for strain and damage monitoring. This combination makes the JE-L4C system particularly promising for semi-structural polymer-composite housing components where mechanical performance, renewable material content and intrinsic condition monitoring are required simultaneously.

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