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CONFERENCE PAPER · LOW-CARBON BUILDINGS

Lignin-Based Reinforcement of HDPE–EPDM Blends for Low-Carbon and Energy-Efficient Applications

Muhammad Khatami[1,2*], H.P.S. Abdul Khalil[3], Hazizan Md Akil[4], Mohammad Aliff Shakir[1], Mardiana Idayu Ahmad[1]

[1] School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia
[2] Department of Industrial Engineering, Faculty of Science and Technology, Universitas Samudra, Langsa 24416, Indonesia
[3] Fakulti Perniagaan, Hospitaliti dan Teknologi, Universiti Islam Melaka, Kuala Sungai Baru, 78200 Melaka, Malaysia
[4] School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia

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ABSTRACT

The integration of bio-based fillers into thermoplastic elastomer blends has garnered considerable interest within the scientific community. However, quantitative assessments of lignin micro fillers in high-density polyethylene (HDPE) and ethylene-propylene-diene monomer (EPDM) systems remain sparse. This study systematically investigates the influence of lignin micro filler incorporation on the mechanical, thermal, and morphological properties of HDPE/EPDM blends. The findings reveal that incorporating lignin significantly enhances both stiffness and strength. Notably, the tensile strength exhibited an increase from 7.474 MPa to 9.490 MPa, while the tensile modulus demonstrated a substantial rise from 44.086 MPa to 152.403 MPa. This pronounced increase in modulus is attributed to the intrinsic rigidity of lignin particles, which facilitates improved stress transfer within the polymer matrix. Morphological analyses indicated a relatively uniform dispersion of lignin at optimized loading levels, thereby enhancing interfacial interactions among the constituents. However, elevated lignin content was observed to diminish ductility, likely due to constraints on polymer chain mobility. This investigation addresses the existing gap in the quantitative understanding of lignin-reinforced HDPE/EPDM blends. It demonstrates that optimized lignin incorporation can substantially augment mechanical stiffness while retaining acceptable strength levels. The findings emphasize the potential of lignin-based composites for sustainable automotive components, sealing materials, and eco-friendly structural materials that require superior rigidity and thermal resistance.

KEYWORDS Lignin micro filler, HDPE/EPDM blends, Mechanical properties, Sustainable composites

1. Introduction

High-density polyethylene (HDPE) and ethylene-propylene-diene monomer (EPDM) rubber are widely combined to produce thermoplastic elastomer (TPE) blends that merge the processability of a semi crystalline thermoplastic with the flexibility and impact resistance of an elastomeric phase. HDPE/EPDM systems of this type are used in automotive weather-seals, industrial gaskets, and flexible packaging components, where a balance between stiffness, toughness, and long-term durability is required. As the plastics industry moves toward reducing its reliance on petroleum-derived materials, there is growing interest in reinforcing thermoplastic elastomer matrices with renewable, low-cost bio-based fillers rather than conventional mineral or carbon-based fillers (Ridho *et al.* 2022).

* Corresponding author. Email address: dalimunthe.khatami@student.usm.my (Muhammad Khatami)

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Lignin, the second most abundant terrestrial biopolymer after cellulose, is generated in large volumes as a low-value by-product of the pulp, paper, and biorefinery industries. Its rigid, highly aromatic macromolecular architecture, together with its comparatively low density, gives lignin considerable promise as a reinforcing micro-filler in polyolefin and elastomer matrices (Ridho *et al.* 2022; Orebom *et al.* 2021). Several studies have reported that lignin incorporation into HDPE improves stiffness through enhanced stress transfer between the rigid filler particles and the surrounding polymer matrix, although the magnitude of the effect depends strongly on the lignin source, particle size, filler loading, and the degree of interfacial adhesion achieved during melt compounding (Menta *et al.* 2022; Ikhtiarini *et al.* 2026; Xie *et al.* 2026). In EPDM-based elastomers, lignin has similarly been explored as a partial or full replacement for carbon black, with reported improvements in modulus and tear strength, generally accompanied by trade-offs in elongation and thermal stability (Poyraz *et al.* 2022; Poyraz *et al.* 2023; Yuan *et al.* 2024).

Despite this growing body of literature on lignin-filled HDPE composites and lignin-filled EPDM elastomers considered separately, quantitative studies that specifically address lignin micro-fillers incorporated into HDPE/EPDM thermoplastic elastomer blends remain comparatively scarce. Most previous work has focused either on rigid, HDPE-dominated systems (Menta *et al.* 2022; Ikhtiarini *et al.* 2026; Xie *et al.* 2026) or on crosslinked EPDM rubber compounds (Poyraz *et al.* 2022; Poyraz *et al.* 2023; Yuan *et al.* 2024), leaving the intermediate thermoplastic elastomer regime, in which a crystalline thermoplastic phase and a rubbery phase coexist, relatively unexplored from the standpoint of lignin reinforcement. This gap is significant, since the two-phase morphology of HDPE/EPDM blends is expected to respond differently to a rigid micro-filler than either single-component system, particularly with respect to stress transfer across the HDPE/EPDM interface and the resulting balance between stiffness and ductility.

The present study therefore aims to systematically investigate the effect of lignin micro-filler loading on the tensile, flexural, and water-absorption behaviour of HDPE/EPDM blends, and to relate the observed mechanical and physical trends to the underlying mixing, dispersion, and interfacial mechanisms. By quantifying how lignin content shifts the trade-off between stiffness/strength and ductility in this specific blend system, this work addresses the existing gap in the quantitative understanding of lignin-reinforced HDPE/EPDM composites and provides guidance for the design of sustainable, lignin-based materials for automotive sealing components, gaskets, and other applications that demand a favourable combination of rigidity, toughness, and moisture resistance (Poyraz *et al.* 2022; Poyraz *et al.* 2023; Balaji Ayyanar *et al.* 2024; Ayyanar *et al.* 2024).

2. Material and Methods

2.1. Materials

High-density polyethylene (HDPE) was chosen because it is often used and easy to recycle. The HDPE came from Lot 29.01, Level 29, 1 Powerhouse, No. 1, Persiaran Bandar Utama, Bandar Utama, 47800 Petaling Jaya, Selangor Darul Ehsan, Malaysia. The density of the HDPE was 0.92 g/cm^3, and its melt flow index (MFI) was 0.93 g/10 min at 230 °C. (Atiqah *et al.* 2015). Ethylene propylene diene monomer (EPDM) of grade Vistalon 2504N was obtained from ExxonMobil Chemical. Lignin microparticles (LMPs) were made using a method called acid precipitation to extract crude lignin from the leftover liquid of coconut fiber waste (Alfatah and Abdul Khalil 2024). The LMPs were then passed through sieve meshes numbered 500 and 270 to get particles that were about 25–53 micrometers in size. After that, the LMPs were dried in an oven at 50 degrees Celsius for a full night and kept in a desiccator to stay stable until they were needed for use (Balaji Ayyanar *et al.* 2024).

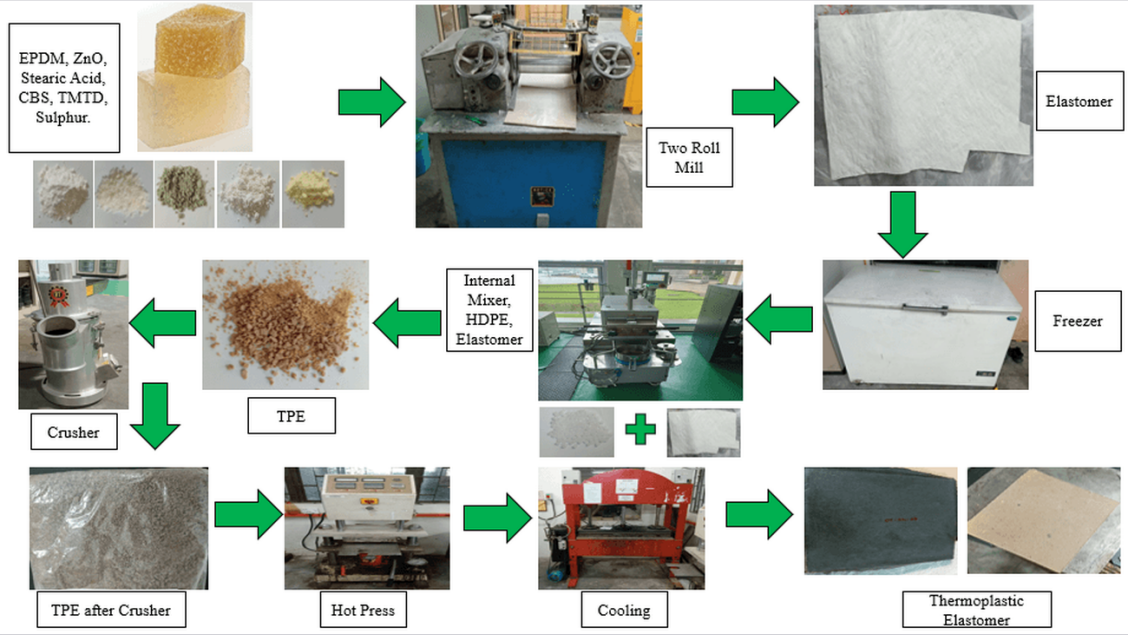

Figure 1. The overall process of preparing thermoplastic elastomer (TPE).

Table 1. Formulation of HDPE/EPDM composites with lignin.

Materials	Wt. (%)
HDPE + EPDM (Control)	50:50
HDPE + EPDM + Lignin	45:45:10
HDPE + EPDM + Lignin	40:40:20
HDPE + EPDM + Lignin	35:35:30
HDPE + EPDM + Lignin	30:30:40
HDPE + EPDM + Lignin	25:25:50

2.2. Methods

Mixing procedure

A Haake Reomix PolyDrive was used to prepare the composites. Mixing was done at 50 rpm and 180 °C. To start the melt mixing process, HDPE was first filled. Lignin was added two minutes later, and then elastomer at the six-minute mark. Three more minutes were spent mixing. After ten minutes, the composite was removed and ground at a nip setting of 2.0 mm in a lab mill. An electrically heated hydraulic press was used to compress the composite samples. The hot-pressing process entails 10 minutes of preheating at 250 °C and 20 minutes of cooling under pressure.

Measurement of tensile properties

The Instron 3366 was subjected to tensile tests in accordance with ASTM D 412. The Wallace die cutter was used to cut dumb bell specimens from the moulded sheets that were 115 mm long overall (the gauge length was 25 mm), 6 mm wide, and 3 mm thick. The test was conducted at 25 ± 3 °C with a cross head speed of 50 mm/min.

Measurement of flexural properties

In research on the effects of lignin micro-filler in HDPE-EPDM (high-density polyethylene-ethylene propylene diene monomer) thermoplastic elastomers, flexural properties—such as flexural strength and modulus—are often evaluated to understand how lignin addition influences the material's stiffness and resistance to bending. Flexural testing of HDPE-EPDM with lignin micro-filler reveals that moderate lignin additions enhance stiffness and bending strength, though excessive filler can weaken flexural properties due to poor dispersion and reduced interfacial bonding.

Optimizing lignin content is essential to maintain balanced mechanical properties. Flexural tests were carried out according to ASTM D 790. Test specimens with a cross-section of 3.2 mm × 12.7 mm and a length of 127 mm were used.

Impact Test

Specimen for the Izod impact evaluation followed ASTM D256 geometry, with each bar cut to a length of 63.5 mm and a width of 12.7 mm, matching the dimensions Ling et al. used for their Izod-type test bars prepared under ASTM D256-10 (Ling *et al.* 2024). Every specimen carried a standard V-notch on one face before testing, since the notch concentrates stress at a controlled point and forces crack initiation to occur where the pendulum strike will register the energy absorbed rather than at a random flaw elsewhere in the bar. Testing itself followed the pendulum-impact principle central to ASTM D256, in which a calibrated hammer swings through a fixed arc and strikes the notched face of a clamped specimen, with the energy consumed in breaking the bar read directly from the instrument scale and normalized by specimen thickness.

Water absorption test

The composite samples were placed in a beaker containing 100 mL of water at a temperature of 22 °C for 9 days. The initial weight of the samples was recorded, and then after 0, 1, 3, 5, 7 and 9 days. They were removed from the beaker, quickly wiped and dried, and weighed immediately. The amount of water absorbed by the samples was determined by calculating the percentage increase in mass. Five replicates were carried out to obtain a mean value for each set of samples. The composite samples were put into distilled water at room temperature. The amount of water they absorbed was checked by weighing them at set times. This follows the standard ASTM D570 test method for measuring water absorption in plastics, which involves soaking the samples in distilled water for 24 hours. The prepared samples should be placed in a container with distilled water that is kept at a steady temperature.

3. Results and discussion

3.1. Mixing Procedures

Torque-time curves for HDPE-EPDM and HDPE/EPDM/Lignin composites with filler amounts of 0%, 10%, 20%, 30%, 40%, and 50%. All the torque-time curves for each composite follow a similar pattern. At first, HDPE is added to the Haake machine, and the rotor starts turning. There is a quick rise in torque because HDPE resists the rotor. As HDPE begins to melt, the torque starts to go down. Right after adding the filler, at the third minute, there is a sharp drop in torque because the filler acts as a lubricant. At the sixth minute, when cold EPDM is added to the mixer, there's a sudden increase in torque. This spike shows that HDPE is being loaded and melted. Once HDPE is fully melted, the torque slowly decreases because the material becomes less thick, until it reaches a steady level. It is clear that as more filler is added, the torque stabilizes at a higher level. This means that adding more filler makes the composite thicker when melted.

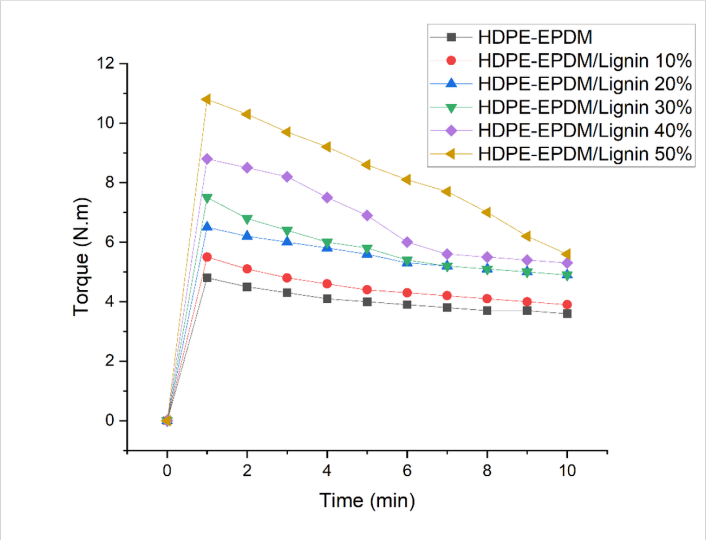

Figure 2. Torque vs time of TPE, and TPE-L.

3.2. Tensile Properties

ASTM D412 is a widely used standard for testing the tensile properties of rubber and elastomeric materials. The test involves pulling a sample of the material until it breaks to determine several mechanical properties that describe its behaviour under tension. These properties are critical for determining the suitability of rubber and elastomers in applications that require flexibility, strength, and durability under tensile loads.

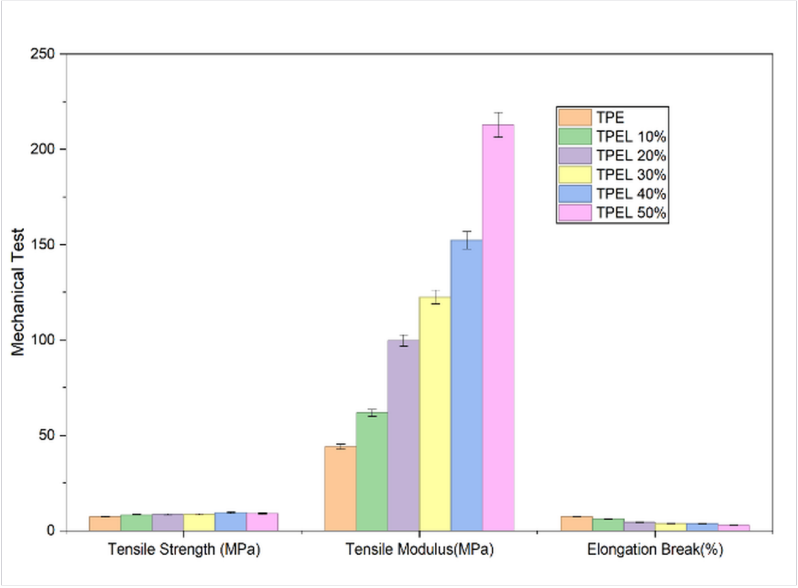

Figure 3. Tensile test of TPE-L.

Tensile strength of TPE-L climbed from 7.5 MPa at 0 wt.% lignin to a peak of 9.4 MPa at 40 wt.%, then dropped to 9.0 MPa at 50 wt.%. Menta et al. reported the identical pattern in tobacco-lignin/HDPE blends: strength held steady up to 15 wt.% and only fell once loading passed 30 wt.%, a shift they traced to particle agglomeration overtaking the reinforcing effect of well-dispersed lignin (Menta *et al.* 2022). Wang et al. found comparable strength gains at low lignin fractions in silica-modified HDPE composites, crediting improved interfacial bonding between filler and matrix (Wang *et al.* 2021). Kayaba et al. observed the mirror-image decline at high biomass loading in palm-kernel-shell/HDPE composites, where weak filler-matrix adhesion produced voids and stress concentrations

under load (Kayaba *et al.* 2025). Lignin below 40 wt.% therefore reinforces the HDPE-EPDM matrix through efficient stress transfer; beyond that point, agglomeration and poor wetting between hydrophilic lignin and the non-polar matrix erode the gain.

Tensile modulus told a different story, rising almost fivefold from 42 MPa to 215 MPa across the same range, with the sharpest gains past 20 wt.% lignin. Feng et al. saw the same stiffening in lignin-reinforced EPDM, where rigid lignin domains restricted chain mobility and pushed composite stiffness above that of carbon-black-filled EPDM (Feng *et al.* 2024). Mahmoud et al. recorded a matching jump in HDPE loaded with clay minerals, where 6 wt.% filler raised elastic modulus by over 100% (Mahmoud *et al.* 2024). Elongation at break moved the opposite direction, falling from 7.5% to 2.8%, a loss Jurczyk et al. attributed in biochar-filled polyester blends to restricted matrix mobility and rising composite brittleness at high filler fractions (Jurczyk *et al.* 2024). Stiffness and ductility in TPE-L trade against each other as lignin content rises, exactly as they do in these unrelated filler systems.

The three curves point to 30-40 wt.% lignin as the loading that balances strength, stiffness, and residual ductility in TPE-L. Shorey et al. frame this kind of trade-off as the price of turning a genuinely low-carbon, high-carbon-content byproduct into a functional filler, since lignin's aromatic structure delivers real reinforcement but its polarity mismatch with polyolefin and elastomer matrices caps how far loading can go before performance degrades (Shorey *et al.* 2024). Bjurström et al. measured the same pattern directly: pushing lignin content to 44 wt.% in an ABS matrix cut greenhouse-gas emissions by up to 40% while trading a modest strength loss for a stiffness gain. That convergence across unrelated matrices suggests the 30-40 wt.% window is not a coincidence of this dataset but a structural limit set by lignin chemistry itself.

3.3. Flexural Properties

The flexural test under the standard ASTM D790 is used to measure the flexural properties of unreinforced and reinforced plastics and other materials, including polymers, composites, and wood-based materials. This test provides valuable information on how a material behaves when subjected to bending forces. This test is essential for understanding the bending performance of materials in practical applications, particularly for load-bearing structures.

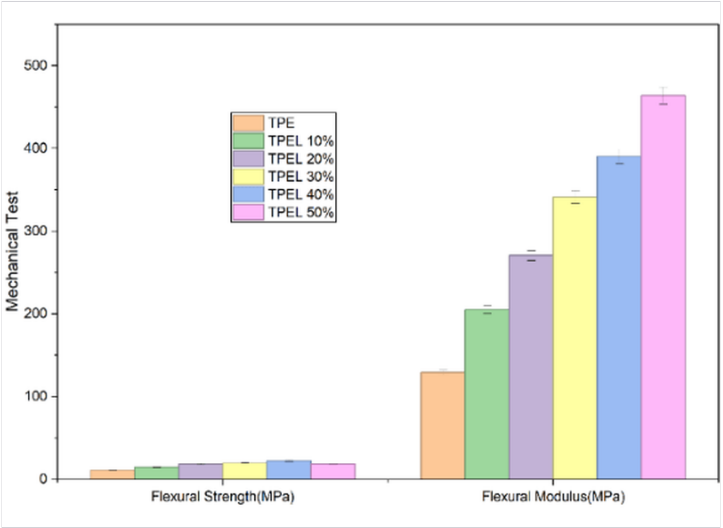

Figure 4. Flexural test of TPE-L.

Flexural modulus climbed steadily across the entire lignin range, from roughly 130 MPa at 0 wt.% to about 470 MPa at 50 wt.%, more than tripling without any reversal. Türkan et al. found the same one-way climb in lignocellulosic-filled polypropylene, where flexural modulus gained up to 18% as filler content rose, a change they linked to restricted polymer chain mobility once rigid particles

occupy the matrix (Türkan *et al.* 2026). Piyanirund et al. reported a matching stiffening trajectory in epoxy-lignin composites, where storage modulus rose steadily with lignin loading before the effect leveled off near 10 phr, since the filler's own higher modulus simply outweighs that of the surrounding resin (Piyanirund *et al.* 2021). Sorze et al. observed comparable behavior in PA6-lignin microcomposites, where increasing lignin content pushed tensile and viscoelastic stiffness upward through strong physical interactions and efficient interfacial adhesion between lignin particles and the polymer chains (Sorze *et al.* 2026). None of these systems showed a modulus decline at high loading, which fits Fig. 4: stiffness accumulates almost in direct proportion to how much rigid lignin the matrix carries, largely independent of the agglomeration effects that limit strength.

Flexural strength followed a different, rising from about 11 MPa at 0 wt.% to a peak near 23 MPa at 40 wt.%, then falling to roughly 18 MPa at 50 wt.%. Bukhary et al. documented the identical shape in PHBV-kenaf biocomposites reinforced with lignin microfillers: flexural strength improved by 15% at an intermediate loading of 5 wt%, then mechanical performance dropped once loading reached 7 wt% because particle agglomerations began to dominate (Bukhary *et al.* 2025). Alshahrani et al. traced a similar optimum in bamboo-fiber polyester composites, where flexural strength and modulus both peaked at a specific lignin fraction before excess filler disrupted the reinforcing effect (Alshahrani and Arun Prakash 2022). This rise-then-fall pattern points to a loading ceiling somewhere near 40 wt.% in TPE-L, past which lignin particles can no longer disperse evenly enough to keep transferring stress efficiently across the HDPE-EPDM matrix.

Set side by side, the two curves describe a composite that keeps getting stiffer well past the point where it stops getting stronger, a dissociation between modulus and strength widely seen once biomass filler content moves past what a given matrix can wet and disperse properly. Ni et al. reached flexural strengths of 30-35 MPa in lignin/HDPE composites only once a maleic-anhydride compatibilizer was added, underscoring that untreated lignin's polarity mismatch with a non-polar matrix caps how much strength further loading can deliver even as stiffness keeps rising (Ni *et al.* 2026). For TPE-L, this suggests 30-40 wt.% lignin as the loading window worth targeting for flexural performance, with compatibilization or lignin surface modification as the likely route to pushing strength higher at 50 wt.% without giving up the stiffness gains already banked at that loading.

3.4. Impact Test

Unfilled TPE recorded the highest absorbed energy in Fig. 5, at roughly 43,000, and every lignin addition pulled that number down, dropping to about 25,500 at just 10 wt.% before continuing a steady decline to 11,500 at 50 wt.%. Pregi et al. reported the same directional effect in polypropylene-lignin-flax composites, noting plainly that lignin addition deteriorates impact resistance and that a secondary fiber phase, in their case flax, was needed just to compensate for that loss (Pregi *et al.* 2022). Obielodan et al. found a matching decline in PLA-lignin biocomposites, where Izod impact resistance dropped as loading rose from 30 wt.% to 40 wt.%, a range that sits well within the steepest part of the TPE-L curve (Obielodan *et al.* 2022). Both studies point to the same underlying cause: lignin is a rigid, non-deformable aromatic filler, and once it is dispersed through an otherwise ductile matrix it interrupts the matrix's ability to absorb impact energy through plastic deformation, so toughness falls even while stiffness climbs.

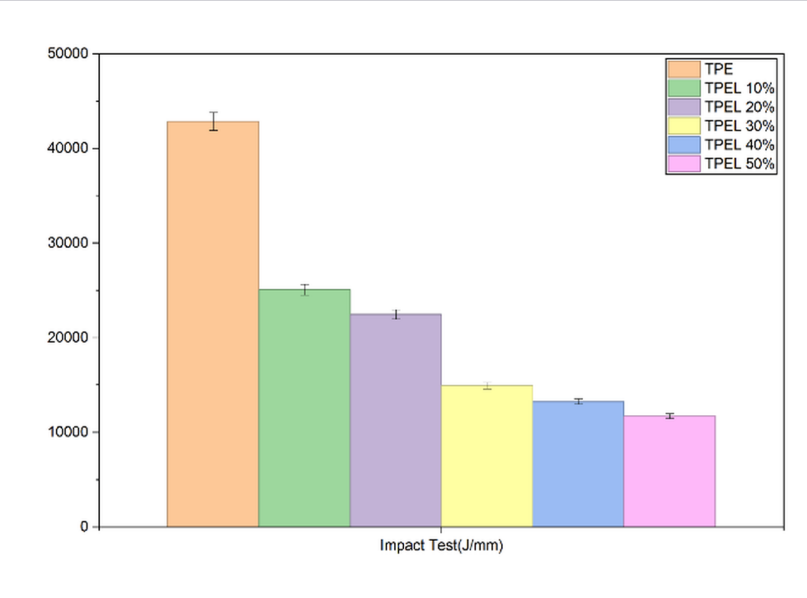

Figure 5. Impact test of TPE-L.

The decline does not level off at higher loading in TPE-L, which distinguishes it from systems where a second reinforcing phase intervenes. Harmaen Ahmad Saffian et al. found that combining modified lignin with kenaf core fiber in a PBS matrix actually produced the highest Izod impact values in their study, because the fibers deflected and diverged crack propagation that would otherwise run straight through the brittle lignin domains (Saffian *et al.* 2021). TPE-L carries no such secondary fiber phase, so cracks initiated at lignin particles propagate with less resistance as particle density increases from 20 wt.% to 50 wt.%, consistent with the near-linear drop visible across that range. Grząbka-Zasadzińska et al. observed a related pattern in PLA loaded with hybrid TiO2-lignin filler, where impact strength moved in the opposite direction from bending strength and hardness as filler content rose, reinforcing that stiffness gains and impact-toughness losses tend to move as a pair once a rigid filler exceeds a few weight percent in a polymer matrix (Grząbka-Zasadzińska *et al.* 2024).

Set against the objective of a low-carbon, energy-efficient TPE-L, this impact data marks the clearest constraint on how far lignin loading can be pushed. The tensile and flexural results favored 30-40 wt.% lignin as a sweet spot, but toughness has already fallen by more than half at 20 wt.% and keeps falling from there, so any formulation targeting impact-sensitive applications would need to sit closer to 10-20 wt.% lignin or borrow the fiber-based compensation strategy Pregi et al. and Harmaen Ahmad Saffian et al. both used to recover toughness without giving up lignin's carbon and cost advantages (Pregi *et al.* 2022; Grząbka-Zasadzińska *et al.* 2024). For applications where impact resistance is secondary to stiffness and material substitution, the 40-50 wt.% range remains defensible.

3.5. Water Absorption

Water absorption stayed close to neat TPE at low lignin content, dipping slightly from 1.08 at 0 wt.% to 0.98 at 10 wt.%, before climbing past the baseline as loading increased further, reaching 1.18 at 50 wt.%. Younesi-Kordkheili et al. saw the same early-stage dip in wood-plastic composites, where lignin content raised from 1 to 5 wt.% steadily lowered water absorption and thickness swelling, an effect they credited to lignin filling interfacial gaps and improving dispersion within the matrix (Younesi-Kordkheili *et al.* 2024). That protective effect appears to run out once loading crosses roughly 20-30 wt. % in TPE-L, a shift Zhiltsova et al. connected to hydrophilic hydroxyl groups on natural fillers becoming more exposed as filler content rises, drawing water into the composite through the growing filler-matrix interface (Zhiltsova *et al.* 2024). Mulana et al. reported the identical direction in

fiber-reinforced polyester composites, where a higher ratio of natural filler to synthetic component consistently pushed water absorption upward (Mulana *et al.* 2023). The 10 wt.% minimum in Fig. 6 therefore marks the point where lignin's void-filling benefit still outweighs its hydrophilicity; beyond that point, the balance tips the other way.

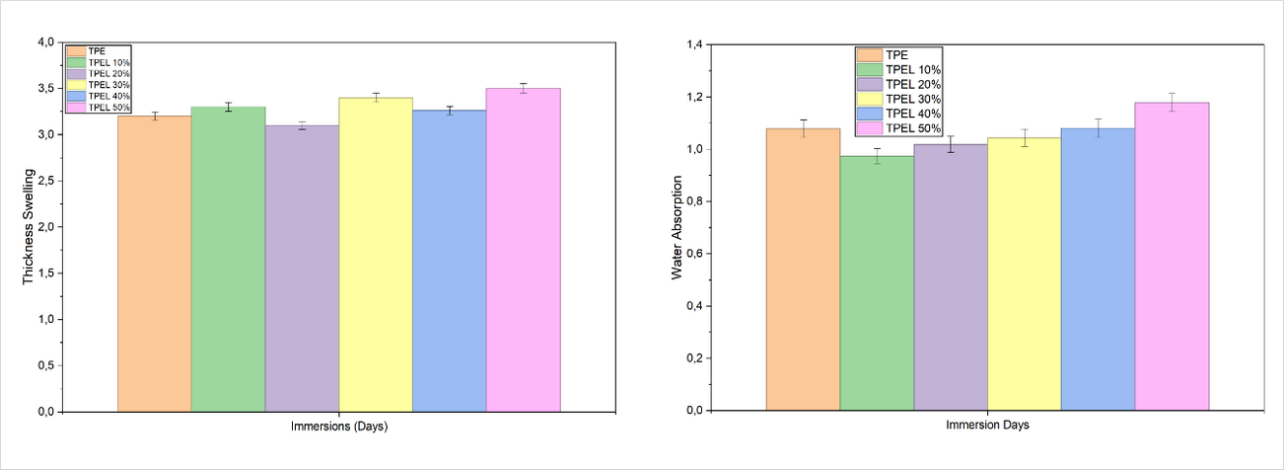

Figure 6. Water absorption and thickness swelling of TPE-L.

Thickness swelling traces a comparable path, dropping from 3.25 at 0 wt.% to a low of about 3.10 at 20 wt.%, then rising to a maximum of 3.50 at 50 wt.%. Gumowska et al. found a similar threshold behavior in lignocellulosic particle composites, where increasing biopolymer binder content improved dimensional stability only up to a point, after which further additions produced no meaningful gain or even reversed the trend (Gumowska *et al.* 2021). The renewed swelling past 30 wt. % lignin in TPE-L likely stems from the same mechanism reported for agro-waste-filled polypropylene, where fiber loading beyond a critical fraction created microvoids and capillary pathways that let water penetrate deeper into the composite structure during prolonged immersion (Zhiltsova *et al.* 2024). Since thickness swelling reflects moisture-driven expansion of the polymer network rather than simple mass uptake, its rise alongside water absorption at high lignin content confirms that the matrix is losing its ability to constrain filler-associated moisture once lignin content exceeds the 20-30 wt.% range.

These two curves place a practical ceiling on how much lignin TPE-L can carry before dimensional stability becomes a liability, a constraint every lignin-composite study eventually runs into once filler content climbs high enough. Selver et al. showed that this ceiling is not fixed: the inherently hydrophobic aromatic structure of lignin can still lower moisture uptake in a polymer matrix by up to 6% when dispersion and interfacial bonding are well controlled, particularly at higher loadings (Selver *et al.* 2025). Kim et al. reached a related conclusion in bio-based polyurethane, where lignin's effect on water resistance depended heavily on how well its aromatic domains interacted with the surrounding polyol network rather than on lignin content alone (Kim *et al.* 2025). For TPE-L, this points toward surface modification or compatibilization of lignin as the likely path to keeping the 40-50 wt.% loading range mechanically attractive without paying the water-absorption and thickness-swelling penalty currently visible in Fig. 5.

4. Conclusion

This study set out to develop a lignin-reinforced HDPE-EPDM thermoplastic elastomer, TPE-L, and to evaluate how its physical and mechanical performance shifts across a range of lignin loadings from 0 to 50 wt.%. That first objective has been met: lignin blends into the HDPE-EPDM matrix without processing difficulty and produces a composite whose properties respond predictably to filler

content, confirming that TPE-L is a technically workable route to replacing part of a petroleum-based elastomer blend with a low-cost, high-carbon-content biomass by-product.

The second objective, evaluating mechanical and physical performance across loadings, is answered by a consistent pattern running through every test. Tensile and flexural strength both climbed with added lignin and peaked around 40 wt.%, then eased back at 50 wt.%, while tensile and flexural modulus rose almost the entire way from 0 to 50 wt.% as the stiff lignin particles restricted movement within the matrix. Elongation at break and impact energy told the opposite story, falling steadily as lignin content increased, since a matrix carrying more rigid filler has less room left to deform or absorb a sudden blow before fracturing. Water absorption and thickness swelling added one more layer to this picture: both dipped slightly at low lignin content, where the filler appears to fill interfacial gaps rather than open them, before climbing past the unfilled baseline once loading passed roughly 20-30 wt.%, at which point lignin's hydrophilic character starts drawing in more moisture than the matrix can keep out.

Reading these results side by side, 30-40 wt.% lignin stands out as the loading that keeps TPE-L on the right side of every property measured here, offering the highest strength values alongside a real stiffness gain, moderate ductility loss, and water uptake that has not yet run away from the baseline. Fifty percent loading buys further stiffness and a lower material cost but gives up strength, toughness, and dimensional stability all at once, so it suits applications where rigidity matters more than durability under impact or moisture exposure. On balance, TPE-L delivers on both objectives: a low-carbon, energy-efficient composite is achievable, and 30-40 wt.% lignin is where that composite performs best without asking too much of any single property.

Declarations

Author contributions. Muhammad Khatami: Writing (original draft), Data curation, Formal analysis, Investigation, Methodology, Conceptualization. H.P.S. Abdul Khalil: Writing (review and editing). Hazizan Md Akil: Writing (review and editing). Mohammad Aliff Shakir: Writing (review and editing). Mardiana Idayu Ahmad: Writing (review and editing).

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Conflicts of interest. The authors declare no competing interests.

Data availability. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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