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Synergistic Effects on Morphology and Properties of Polyurethane-Rubber foam Composite

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Abstract: Polyurethane (PU) foams are widely applied in cushioning, packaging, and construction, but their reliance on petroleum-based feedstocks and limited durability raises sustainability concerns. This study investigates the incorporation of ground tyre rubber (GTR) into flexible PU foams to enhance performance while promoting circular economy goals. Foams were synthesized via the free-rise method using polyethylene glycol (PEG 400) and isophorone diisocyanate (IPDI), with GTR fillers (≤0.4 mm) and silicone oil surfactant. Fourier-transform infrared spectroscopy confirmed successful urethane network formation, while scanning electron microscopy revealed that GTR disrupted cellular morphology, increasing porosity and heterogeneity. Mechanical analysis showed a substantial decline in compressive strength and modulus with increasing filler loading, attributed to poor filler matrix compatibility and uneven dispersion. Thermal analysis highlighted a dual effect where GTR suppressed the glass transition temperature, reflecting reduced microphase separation, yet improved thermal stability by delaying degradation onset and increasing residual char yield. Silicone oil partially mitigated structural collapse by refining cell morphology and enhancing compressive behavior. The results showed a trade-off between sustainability-driven waste rubber utilization and foam integrity. While GTR incorporation advances responsible resource recovery and contributes to Sustainable Development Goals (SDGs), unmodified fillers compromise mechanical reliability, limiting high-performance applications. Future work should emphasize interfacial engineering, including surface modification and compatibilizer integration, to reconcile environmental imperatives with performance requirements.

Keywords: polyurethane foam, compressive test, glass transition temperature, degradation temperature, morphology, crumb rubber

I. Introduction

Polyurethane (PU) foams are widely used in packaging, construction, furniture, and automotive sectors due to their lightweight structure (Gama et al., 2018), insulation ability, and versatile mechanical properties (Suwan et al., 2025). However, conventional PU foams are synthesized from petroleum-based raw materials, leading to sustainability concerns such as resource depletion, greenhouse gas emissions (Singh et al., 2025), and waste accumulation (Kosmela et al., 2021). These issues have driven research into incorporating alternative or recycled materials into PU foams, aligning with the Sustainable Development Goals (SDGs), particularly SDG 12 (Responsible Consumption and Production) and SDG 13 (Climate Action).

Blending PU with rubber, especially recycled or waste rubber, offers a promising route to reduce environmental burdens while improving performance (Lechuga-Islas et al., 2025). Piszczyk et al. (2015) reported that incorporating recycled tyre rubber into flexible PU foams enhanced compressive strength and energy absorption, showing potential for durable cushioning applications. Similarly, Perera et al. (2025) demonstrated that blending rigid PU waste with nitrile butadiene rubber (NBR) improved mechanical and thermal properties, highlighting waste utilization opportunities. Other studies have explored PU composites with natural fibers and powders from foam scraps, finding that recycled fillers can partially replace virgin polyols without significant property loss (Maamoun et al., 2025).

Sustainability efforts also extend to bio-based PU foams, such as those derived from palm oil and water hyacinth fibers, which improve acoustic absorption and support circular economy models (Sukhawipat et al., 2022). A systematic review by (Zhang et al., 2025) further emphasizes that recycling PU foams and incorporating waste materials are critical strategies to safeguard the environment and achieve SDGs (Haba et al., 2025). Despite these advances, fewer studies have systematically examined the performance of PU foams blended specifically with ground tyre rubber (GTR). Therefore, this research aims to evaluate the properties of PU foam blended with GTR and to demonstrate how such innovations contribute toward advancing the SDGs by promoting sustainable materials and reducing environmental impact.

II. Methods

Flexible PU foam was synthesized using polyethylene glycol 400 (PEG 400, Merck), which has a hydroxyl number of 267–295 mg KOH/g and an average molecular weight of 380–420 g/mol. Isophorone diisocyanate (IPDI, Sigma-Aldrich), containing 98% NCO

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groups, was employed as the isocyanate component. Tin (II) 2-ethylhexanoate (Nacalai Tesque) served as the catalyst, silicone oil as the silicon-based surfactant, and distilled water as the blowing agent. Waste fillers in the form of styrene-butadiene rubber (SBR) ground tyre rubber (crumb rubber, ≤0.4 mm) were incorporated to modify the PU foam. The foams were prepared using the free-rise method with a two-component (A and B) system, featuring an NCO/OH molar ratio of 2:1.

Component A (polyol mixture) was prepared by combining PEG 400 (15 mL), catalyst (100 µL), surfactant (100 µL), distilled water (500 µL), and filler (1, 2, or 3 g) in a 16 oz polypropylene cup. The mixture was homogenized using a mechanical stirrer at 1000 rpm for 60 s. Component B (IPDI, 15 mL) was then added and stirred at 1000 rpm for 3 min. The resulting mixture was allowed to rise in the cup freely and subsequently cured in an oven at 60 °C for at least 24 hours.

Characterization of the PU foams was conducted using several analytical techniques. ATR-FTIR spectra were recorded with a Perkin Elmer Spectrum One in the range of 4000–650 cm^{-1} at room temperature, using a resolution of 8 cm^{-1} and four scans. Morphological analysis was performed with a Hitachi TM3030Plus scanning electron microscope (SEM) at 50× magnification. Apparent density was measured using a gravity densimeter on samples with dimensions of 2 × 2 × 2 cm. Compressive strength was evaluated according to ASTM D3574 using a tensile tester at a crosshead speed of 4 mm/min until 80% deformation. Thermal properties were studied by differential scanning calorimetry (DSC) under a nitrogen atmosphere from 30 to 300 °C at a heating rate of 10 °C/min, and by thermogravimetric analysis (TGA) in the range of 30–600 °C under nitrogen at a heating rate of 20 °C/min.

III. Results

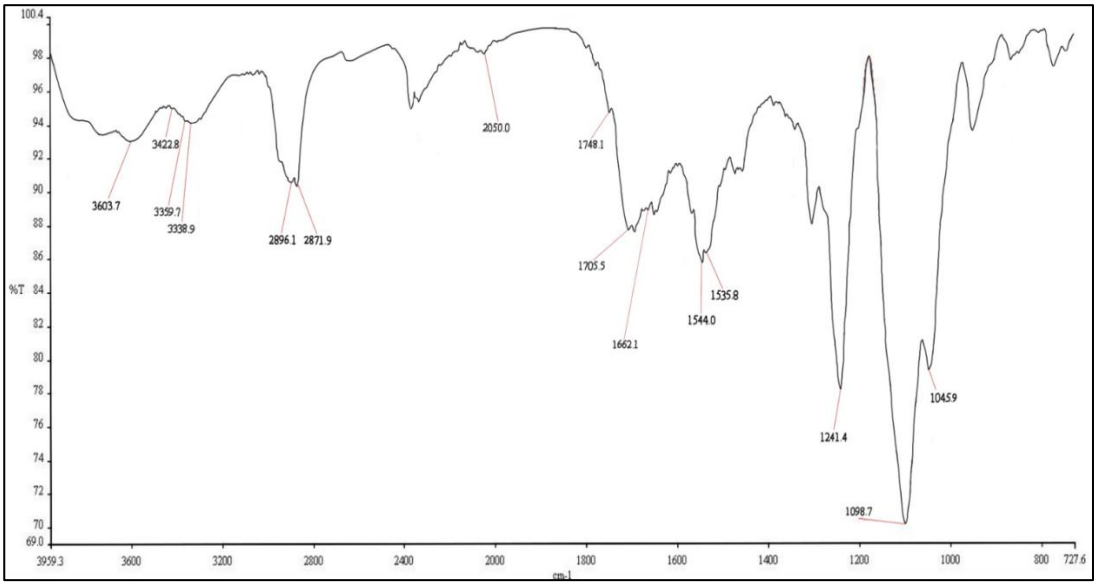

Figure 1: FTIR spectrum of synthesized PU foam

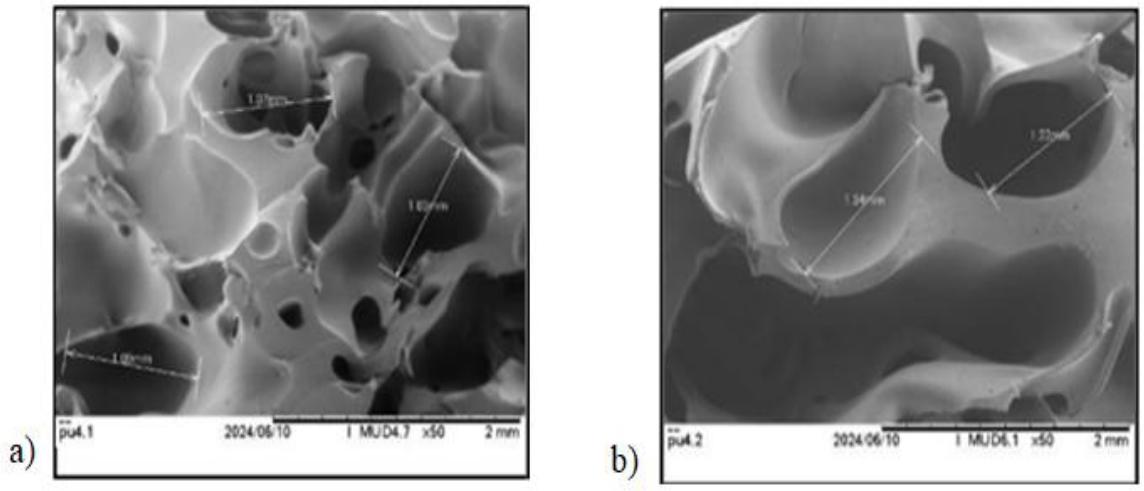

Figure 2: Average cell size of unfilled PU foam (a) without silicone oil (b) with silicone oil

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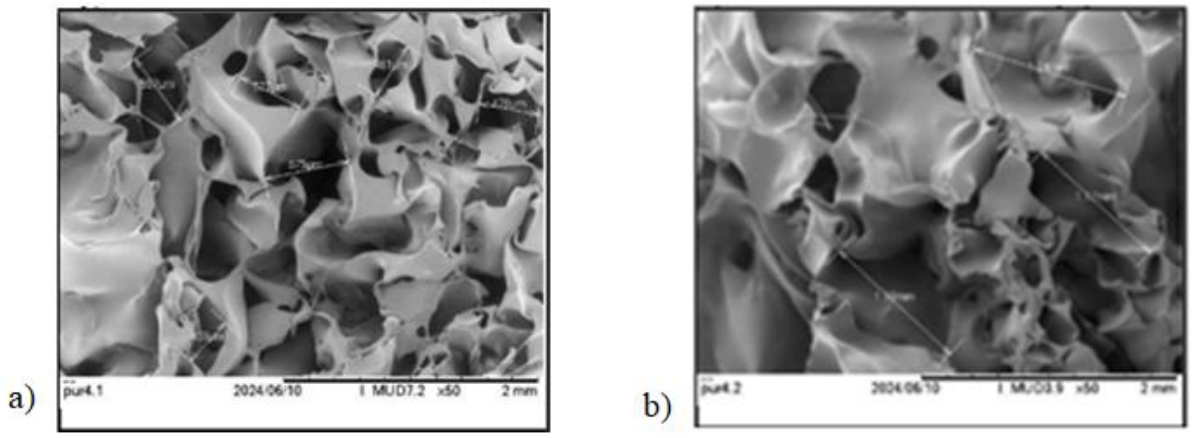

Figure 3: Average cell size of GTR-filled PU foam (a) without silicone oil (b) with silicone oil

Table 1: Effect of different amounts of loading filler in PU foam on compressive modulus, compressive stress and density (4.1: without silicone oil, 4.2: with silicone oil)

	PU4.1	PU4.2	PUR4.1(1)	PUR4.2(1)	PUR4.1(2)	PUR4.2(2)	PUR4.1(3)	PUR4.2(3)
Compressive stress at peak (MPa)	0.70	2.46	0.70	0.11	0.04	0.21	0.99	0.17
Compressive modulus (MPa)	1.22	6.04	5.37	0.49	0.09	0.31	1.60	0.22
Density (g/cm^3)	0.591	0.694	0.438	0.352	0.321	0.278	0.399	0.301

Table 2: Characteristics of thermal properties of PU foams with filler loading and surfactant content (4.1: without silicone oil, 4.2: with silicone oil)

Samples	Glass transition temperature, T_g (°C)	Onset degradation temperature (°C)	Mass loss (%)				
			Temperature (°C)				
			T_2	T_5	T_{10}	T_{50}	Residual mass (%)
PU4.1	147.37	294.0	94.932	189.932	299.932	349.932	0.54
PU4.2	136.20	267.8	93.131	173.131	303.131	353.131	0.22
PU4.1 (1)	130.70	297.0	73.130	208.130	303.130	353.130	2.87
PU4.2 (1)	-	319.5	129.199	279.199	309.199	354.199	3.32

IV. Discussion

FTIR Analysis

The ATR-FTIR spectra of the synthesized polyurethane (PU) foams provided clear evidence of successful urethane network formation. The broad absorption at 3420 cm^{-1} corresponds to O–H stretching vibrations from residual hydroxyl groups of PEG, while the characteristic NCO peak at 2049 cm^{-1}, originating from isophorone diisocyanate (IPDI), was absent in the final PU spectrum, confirming complete consumption of isocyanate groups during polymerization (Gao et al., 2023). The presence of the N–H stretching band at 3345 cm^{-1} and the carbonyl absorptions at 1748 cm^{-1} (urethane C=O), 1706 cm^{-1} (hydrogen-bonded urethane C=O), and 1662 cm^{-1} (urea C=O) further validate the formation of urethane and urea linkages, characteristic of segmented PU networks. Additionally, the band at 1098 cm^{-1} corresponds to C–O–C stretching in the PEG soft segment, signifying successful incorporation of polyether chains into the PU matrix (Lu et al., 2025).

The coexistence of free and hydrogen-bonded carbonyl groups suggests heterogeneous microphase separation between hard and soft domains. This feature strongly influences the mechanical and thermal response of PU foams. The dominance of hydrogen-bonded urethane carbonyls (1706 cm^{-1}) over free carbonyls (1748 cm^{-1}) indicates extensive inter- and intra-molecular hydrogen bonding within the hard domains, consistent with the findings of Mailhot et al. (2001), who reported that hydrogen bonding enhances rigidity but can compromise elasticity.

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For crumb rubber-filled PU foams, no new absorption peaks were observed, indicating that the interaction between PU and rubber is primarily physical rather than chemical. However, minor shifts and intensity variations in the urethane carbonyl bands suggest partial interference of rubber particles with hydrogen bonding equilibrium within the PU matrix (Gan et al., 2024). Similar phenomena were reported by Piszczyk et al. (2015), who noted that rubber fillers disrupt hard-segment packing and microphase separation, thereby altering foam morphology and mechanical integrity.

As a summary, FTIR results confirm the successful synthesis of PU networks and reveal that the incorporation of crumb rubber (GTR) modifies the hydrogen bonding structure without forming new chemical linkages. This disruption is expected to correlate with the reduced compressive performance and altered thermal stability observed in subsequent analyses, highlighting the critical role of filler–matrix interactions in determining the multifunctional properties of PU composites.

Morphology Analysis (SEM)

Figures 2 and 3 elucidate the distinct effects of silicone oil and GTR incorporation on the cellular morphology of flexible PU foams. In Figure 2, the unfilled PU foams demonstrate that the absence of silicone oil produces large, irregular, and poorly stabilized open cells, characteristic of weak foaming stabilization and insufficient surfactant activity. These unstable structures are prone to coalescence and collapse, explaining the lower compressive performance observed in mechanical testing. When silicone oil is introduced, the average cell size decreases substantially, and the foam exhibits more uniform cellular morphology. This is attributed to the ability of silicone surfactants to lower surface tension, stabilize bubble walls, and promote the formation of finer, more evenly distributed closed-cell structures, consistent with observations by Domingos et al. (2019).

Figure 3 highlights the morphological evolution upon incorporation of GTR fillers. In the absence of silicone oil, GTR-filled foams exhibit markedly smaller cell sizes but with significantly higher porosity compared to their unfilled counterparts. The increased viscosity imparted by rubber particles disrupts gas diffusion and cell expansion, yielding denser, irregular morphologies and indicating interference with the foaming kinetics (Kosmela et al., 2021). The resulting heterogeneity in pore structure contributes to inconsistent compressive behavior, as regions of dense filler aggregation act as stress concentration sites (Mukherjee et al., 2020). When silicone oil is present alongside GTR fillers, the foam microstructure appears more refined, with improved distribution of smaller cells and enhanced cell wall integrity. This suggests that surfactant-assisted stabilization partially mitigates the disruptive influence of GTR, although interfacial incompatibility between rubber particles and the PU matrix still limits structural homogeneity (Zarezadeh et al., 2024).

Compression Test

Table 1 provides quantitative evidence of how filler loading and the presence of silicone oil influence the compressive properties and density of flexible PU foam. The unfilled PU foams establish the baseline behavior, where the foam prepared with silicone oil (PU4.2) exhibited a substantially higher compressive stress (2.46 MPa) and modulus (6.04 MPa) compared to the foam without surfactant (PU4.1, 0.70 MPa and 1.22 MPa, respectively). This remarkable enhancement highlights the essential function of silicone oil in reinforcing cell wall stability, lowering surface tension, and facilitating a finer and more uniform cellular structure, thereby improving the load-bearing capacity of the foam. These results align with the findings of Domingos et al. (2019), who emphasize surface-assisted reinforcement of foam morphology as a key driver of improved compressive resistance.

When GTR fillers were introduced, a significant reduction in both compressive and modulus was observed across most formulations, concomitant with decreased foam density. For instance, PU4.1 (2) exhibited compressive stress and modulus values as low as 0.04 MPa and 0.09 MPa, respectively, accompanied by a density of only 0.321 g/cm^3. This reduction reflects the disruptive influence of rubber particles on the foam network, where poor filler dispersion and weak interfacial adhesion between the hydrophobic rubber and the PU matrix compromise stress and structural cohesion. Similar effects have been reported by Farooq et al. (2025), who demonstrated that rubber fillers often act as structural defects rather than reinforcing agents in flexible PU systems unless surface modification or compatibilization strategies are employed.

Interestingly, certain formulations deviate from the general weakening trend. For example, PUR4.1 (3) retained a relatively higher compressive stress (0.99 MPa) despite its reduced density (0.399 g/cm^3). Such variances likely arise from localized densification due to uneven filler distribution, leading to regions of enhanced stiffness embedded within an otherwise porous structure (Sun et al., 2024). However, localized effects introduce heterogeneity and unpredictable mechanical performance, which are undesirable for advanced applications requiring reproducibility and consistency (Dutta, 2018).

Thermal Analysis

Table 2 presents the thermal response of PU foams with varying filler and surfactant compositions, offering insights into how GTR and silicone oil influence the structural stability of the polymer network. The glass transition temperature (Tg) of the unfilled foams reflects the stabilizing influence of silicone oil, with PU4.1 showing a Tg of 147.37 °C that decreased to 136.20 °C in PU4.2. This reduction demonstrated the plasticizing effect of surfactant, which increases molecular chain mobility by reducing intermolecular interactions in the soft segment domains (Formela et al., 2017). The incorporation of GTR fillers further suppressed Tg, with values as low as 130.70 °C for PUR4.1(1) and undetectable in PUR4.2(1), indicating significant disruption of microphase separation between hard and soft segments. Such suppression of Tg suggests that rubber particles interfere with the hydrogen-bonding equilibrium within the hard domains, a phenomenon similarly reported by Piszczyk et al. (2015) in PU/rubber composites.

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The thermogravimetric analysis (TGA) results reinforce these observations. All samples exhibited a single major degradation stage between 250–350 °C, corresponding to the breakdown of urethane linkages and polyether chains, consistent with previous studies by Dong et al. (2024). The unfilled PU foams demonstrated lower onset degradation temperatures (294.0 °C for PU4.1 and 267.8 °C for PU4.2), reflecting the inherent thermal instability of flexible PU matrices. Notably, the incorporation of GTR in the presence of silicone oil enhanced thermal stability, with PUR4.2(1) showing an onset degradation temperature of 319.5 °C. This improvement can be attributed to the char-forming ability of rubber particles, which act as thermal barriers and slow down volatile release during decomposition (Jiang et al., 2022).

Residual mass at 600 °C further supports this mechanism, with filled foams retaining significantly higher residues (2.87% for PUR4.1(1) and 3.32% for PUR4.2(1)) compared to nearly complete decomposition in the unfilled systems (<1%). The elevated char yield indicates that rubber fillers contribute thermally stable components that persist beyond the degradation of the PU matrix (Demewoz & Yeh, 2022). However, the absence of a distinct second degradation stage suggests that the rubber is not chemically integrated but rather physically entrapped, which limits its reinforcing potential (Guo et al., 2024).

Collectively, the SEM analysis (Figures 2 and 3) and thermal–mechanical data (Tables 1 and 2) reveal a critical trade-off between sustainability-driven filler incorporation and the structural integrity of polyurethane foams. While the addition of ground tire rubber (GTR) increases porosity, reduces cell size, and lowers compressive strength due to poor filler–matrix compatibility, silicone oil acts synergistically to stabilize cell morphology and partially recover mechanical resilience (Liang et al., 2024). This morphological evidence strongly correlates with the reduced compressive performance and altered thermal transitions, where GTR incorporation compromises chain mobility and decreases Tg but simultaneously enhances thermal stability through increased char residue and delayed onset degradation. Together, these findings underscore the complex interplay between surfactants, fillers, and interfacial interactions in dictating foam performance. To reconcile sustainability objectives with the structural and functional requirements of high-performance PU foams, future strategies must focus on interfacial engineering via surface modification of rubber particles, compatibilizer incorporation, or optimized dispersion to enable the utilization of waste rubber without sacrificing mechanical integrity or thermal resilience.

V. Conclusion

This study demonstrated that incorporating GTR into flexible PU foams introduces a clear trade-off between sustainability gains and material performance. While FTIR confirmed the successful synthesis of urethane linkages, SEM analysis revealed that GTR disrupts cell morphology, increasing porosity and reducing uniformity, which directly correlates with the significant decline in compressive strength and modulus. Thermal analysis further highlighted this dual effect: GTR reduced the glass transition temperature due to interference with microphase separation but enhanced thermal stability through higher char residue and delayed onset of degradation. The synergistic role of silicone oil in stabilizing cell walls partially mitigated these negative mechanical effects, yet interfacial incompatibility between rubber particles and the PU matrix remained a critical limitation.

The findings emphasize that unmodified GTR, though environmentally attractive, compromises the structural reliability of PU foams, restricting their suitability for high-performance cushioning applications. To align with the Sustainable Development Goals (SDGs), particularly in advancing resource recovery and climate action, future work should prioritize interfacial engineering strategies such as filler surface modification, compatibilizer integration, and optimized dispersion techniques. These approaches are essential to unlock the full potential of recycled rubber as a functional filler, enabling the development of durable, sustainable PU composites that meet both performance and environmental imperatives.

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