

HYBRID WASTE MODIFIERS FOR PERFORMANCE ENHANCEMENT OF ASPHALTIC CONCRETE: A SUSTAINABLE APPROACH USING LIGNITE, POLYTHENE, AND TYRE DERIVATIVES

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ABSTRACT

There is growing interest in sustainable pavement materials, necessitating the wider use of waste-derived modifiers in asphalt concrete. This paper reports on the mechanical and microstructural performance of asphalt mixtures stabilized with lignite, waste polythene (WPT), and worn-out tyre rubber (WOT), which were designed using the Latin hypercube sampling (LHS) method. Thirty mixture formulations were created to investigate the interactive effects of the waste modifiers on mixture performance parameters of Marshall stability, flow, indirect tensile strength (ITS) and resilient modulus (RM). Hybrid modification demonstrated competing effects of binder stiffening on the matrix's stiffness and viscoelasticity. The performance spectrum of the mixture formulations showed 7% bitumen, 2% lignite, 2% WPT, and 5% WOT (R17), providing better-balanced performance with high strength and ductility, and 20.6% higher tensile strength (ITS) with 17.9% flow reduction for crack resistance and deformation control. In comparison, RM at 1470 kN/m was high yet comparable (+6.5%) to the control, and the Marshall stability of 6.60 kN was also consistent. Correlations between performance parameters suggested a high dependence of stability on RM ($R^2 = 0.683$; $p < 0.001$), but none of the mixture parameters was highly associated with one another. While tensile strength was weakly associated with stability ($R = 0.133$), its strong relationship with binder modification indicated that it is an indicator of resistance to cracking. Microstructural XRD, XRF, and SEM-EDS results revealed silica-rich mineral phases ($\text{SiO}_2 = 47.85\text{-}64.80\%$) coating the fused mineral particles and bonding the constituent mixtures, thereby densifying the matrix and improving the dispersion of carbon and mineral particles, which contributed positively to the observed mixture performance. Overall, hybrid modification was demonstrated to enable performance optimisation rather than parameter maximisation, producing a mixture that offers balanced stiffness, flexibility and strength, with good adhesion and durability. The optimal mixture composition was identified as 7% bitumen, 2% WPT, 2% lignite and 5% WOT (R17).

Keywords: Hybrid asphalt; Lignite; Waste polythene; Worn-out tyre; Sustainable pavement; Mechanical performance

1. | Introduction

The escalation of axle loads and the rapid proliferation of commercial vehicles have already put immense pressure on pavement systems, culminating in the advancement of

rutting, fatigue cracking, and surface ravelling (Junaid *et al.*, 2018; Sudarsanan and Kim, 2022). These pavement anomalies are further accentuated by temperature variability and material ageing, which undermine the underlying durability of typical asphaltic concrete systems.

Over the last 20 years, extensive research has focused on enhancing the structural and environmental durability of asphalt concrete by modifying the binder and aggregate constituents (Li *et al.*, 2022; Sharma *et al.*, 2022). Polymeric modifiers such as styrene-butadiene-styrene (SBS), polyethylene, and crumb rubber have been employed to foster ductility and remediating temperature effects, while hybrid polymer systems of crumb rubber and conventional polymer blends have been successfully utilized to mitigate high temperature rutting issues while additionally providing a means for characterizing a diversely recycled rubber modifier (Joohari and Giustozzi, 2021). The inclusion of carbonaceous materials and coke derivatives has further addressed the challenges of softening point, fatigue loading, and long-term rutting resistance inherent in asphalt systems (Hashami *et al.*, 2025). Adjunct mineral admixtures such as fly ash, silica fume and steel slag have also been used to enhance the adequacy of filler-binder interaction (Safiuddin *et al.*, 2016; Sofi and Sukhmanjit, 2017). The environmental concerns associated with the mining and processing of these virgin mineral systems have, however, led to an immediate need to adopt more holistic and circular methods of pavement infrastructure design (Wong and Peng, 2022).

The utilisation of waste materials, including plastics and tyre additives as asphalt binder modifiers, has thus become an identified route to a more sustainable, low-carbon approach to infrastructure construction (Abdel-Shafy and Mansour, 2018; Bamigboye *et al.*, 2021). Plastics and tyre wastes have been effectively employed in asphalt modification due to their mechanical enhancements of performance and concomitant benefits for residual waste management (Bastidas-Martinez *et al.*, 2022; Huan *et al.*, 2019). Notably, lignite, which is characterized as a low ranking carbonaceous material as a result of its high mineral composition and carbon content, has recently been used as a binder extender in low rank regions such as Nigeria following improvements in

activation and pyrolysis steps to generate novel filler materials that result in stiff binders, reduced ageing potential and optimised binder-filler interaction (Ungureanu *et al.*, 2025; Malpani *et al.*, 2023). Waste polythene (WPT), generally generated from bulk films, sachet water bags and food sachet wrapping, is commonly employed in asphalt modification owing to its interaction with the binder to impart hydrophobic property as well as resistance to both physical and chemical oxidation (Aziz *et al.*, 2015; Huan *et al.*, 2019). Similarly, worn-out tyre rubber (WOT) imparts the resilient properties of elasticity and energy absorption to end products, thereby improving impact and directional variance fatigue failure (Juveria *et al.*, 2023; Singaravel *et al.*, 2024). While exceptional performance benefits have been realised by adding each constituent, incorporating more than necessary can lead to adverse trade-offs (reductions in stiffness and strength), which only controlled optimisation can address.

Indeed, limited efforts have been made to explore the synergistic effects of the coupled addition of lignite, WPT, and WOT within asphalt mixtures, in contrast to the individual benefits previously reported (Abdel-Shafy and Mansour, 2018; Bastidas-Martinez *et al.*, 2022). More recently, significant strides have been made using multivariate and statistically based optimisation techniques to achieve a sustainable and substantial reduction in experimental burden while improving performance (Zakerzadeh, 2025; Nkomo, 2022). Furthermore, the valorisation of ash-rich by-products from lignite decomposition has demonstrated multi-beneficial outcomes, including performance enhancement and environmentally oriented benefits (Jankovsk *et al.*, 2024; Kumar *et al.*, 2024). However, to date, there remains a lack of systematic understanding of the coupled microstructural and mechanical behaviour of coupled hybrids, which this work will investigate.

This work seeks to fill this knowledge gap by exploring the asphaltic performance of virgin-

virgin. binder modified with lignite, waste polythene and worn-out rubber. Mixture proportions were subsequently identified using the iterative Latin hypercube sampling (LHS) approach to optimise the trade-off between compositional efficiency and performance benefit. The performance of the resulting asphalt mixture was then investigated by means of a suite of mechanical integrity tests [Marshall stability, flow, indirect tensile strength (ITS), and resilient modulus (RM)] complemented by microstructural analysis via X-ray diffractometry (XRD), Energy dispersive spectroscopy (XRF), as well as scanning electron microscopy (SEM). The overall aim of this work is to identify a multi-component hybrid waste composition that yields a convergence of high stiffness, reduced permeability, and cracking resistance.

2 | Materials and Methods

2.1 | Materials

All materials were sourced in Nigeria for cost budgeting and environmental sustainability. The bitumen used was a 60/70 penetration grade binder from Julius Berger Nigeria Plc, Abuja (Bengeril). This type of bitumen has been commonly employed for roads in tropical/democratic climates because of its balance of flexibility/stiffness. The fine and coarse aggregates were procured from Akure, Ondo State, and were found to meet the gradation

requirements of ASTM C136/C136M-19 and BS 812. The aggregate sizes used were 37.5 mm, 25 mm, 19 mm, 9.5 mm, 6.7 mm, and 4.76 mm to ensure load-bearing capacity and interlock with the asphalt. The lignite was obtained from Enugu State, air-dried, and triturated using a Mc Crone mill through a 75 m sieve, and then used as a filler substitute based on its silicate/carbon composition. Waste polythene (WPT) was obtained from used sachet water bags and polyethene carriers at the Federal University of Technology, Akure campus. The polyethene was rinsed, sun-dried and shredded into flakes. Worn-out tyre rubber (WOT) was obtained from vulcaniser waste dumps, shredded into tiny pieces for easy handling, and then snapped into particles smaller than 2 mm for homogeneous dispersion in the bitumen (XPS). These modifiers were used due to their potential to enhance asphalt performance and their waste-recycling capabilities

2.2 | Mix Design and Sample Preparation

The mix design was formulated to investigate the combined influence of lignite, WPT, and WOT. Two binder contents (4 and 7%) were used as control levels. Lignite and WPT were varied from 0% to 8%, and WOT was incorporated at 5%-20%. The mixture proportions were generated using the Latin Hypercube Sampling (LHS) method to ensure that the composition space is represented equally. Thirty (30) different mixes (R1 to R30) were prepared, and their compositions are shown in Table 1.

Table 1 | Sample Preparation

S/N	Sample ID	Bitumen (%)	Lignite (%)	WPT (%)	WOT (%)	Fine Aggregate (%)	Coarse Aggregate (%)
1	BT4FA23CA73	4	0	0	0	23	67
2	BT4L2WOT5WPT2FA25CA69	4	2	2	5	25	69
3	BT4L8WOT15WPT6FA12CA54	4	8	6	15	12	54
4	BT4L4WOT10WPT4FA15CA70	4	4	4	10	15	70
5	BT4L6WOT20WPT2FA10CA66	4	6	2	20	10	66
6	BT4L2WOT10WPT6FA20CA58	4	2	6	10	20	58

7	BT4L8WOT5WPT4FA14CA65	4	8	4	5	14	65
8	BT4L4WOT15WPT2FA18CA57	4	4	2	15	18	57
9	BT4L6WOT5WPT6FA16CA63	4	6	6	5	16	63
10	BT4L2WOT20WPT4FA19CA51	4	2	4	20	19	51
11	BT4L8WOT10WPT2FA17CA59	4	8	2	10	17	59
12	BT4L4WOT20WPT6FA22CA44	4	4	6	20	22	44
13	BT4L6WOT5WPT4FA21CA60	4	6	4	5	21	60
14	BT4L2WOT15WPT2FA23CA54	4	2	2	15	23	54
15	BT4L8WOT5WPT6FA24CA43	4	8	6	5	24	43
16	BT7FA21CA72	7	0	0	0	21	72
17	BT7L2WOT5WPT2FA25CA59	7	2	2	5	25	59
18	BT7L8WOT15WPT6FA12CA52	7	8	6	15	12	52
19	BT7L4WOT10WPT4FA15CA60	7	4	4	10	15	60
20	BT7L6WOT20WPT2FA10CA60	7	6	2	20	10	60
21	BT7L2WOT10WPT6FA20CA45	7	2	6	10	20	45
22	BT7L8WOT5WPT4FA14CA62	7	8	4	5	14	62
23	BT7L4WOT15WPT2FA18CA54	7	4	2	15	18	54
24	BT7L6WOT5WPT6FA16CA40	7	6	6	5	16	40
25	BT7L2WOT20WPT4FA19CA52	7	2	4	20	19	52
26	BT7L8WOT10WPT2FA17CA56	7	8	2	10	17	56
27	BT7L4WOT20WPT6FA22CA41	7	4	6	20	22	41
28	BT7L6WOT5WPT4FA21CA57	7	6	4	5	21	57
29	BT7L2WOT15WPT2FA23CA51	7	2	2	15	23	51
30	BT7L8WOT5WPT6FA24CA50	7	8	6	5	24	50

2.3 | Testing Programme

The asphalt mixtures were subjected to a physical, mechanical and statistical testing programme. Aggregate properties, including gradation, moisture content, impact value, crushing value, and specific gravity, were determined in accordance with BS 812 and ASTM C127-128. The physical testing of the bitumen was carried out in accordance with the standards ASTM D5 (penetration), ASTM D36 (softening point), ASTM D92 (flash and fire points), ASTM D7945 (viscosity), ASTM D113 (ductility), and ASTM D70 (specific gravity). The mechanical testing of the asphalt mixtures was carried out according to the Marshall stability and flow test (ASTM D6927-15), indirect tensile strength (ITS)

(ASTM D4123), and resilient modulus (ASTM D7369). Static loading testing (ASTM D1559; ASTM E139) was also conducted to assess creep and rutting stability. It was necessary to perform the ITS test at a constant temperature of 25 °C, chosen to represent typical variation. Each asphalt mix was tested once due to limitations of materials and facilities. While the results are based on single tests, statistical analysis was performed to identify trends, but no statistical inference was made. Analysis of influence, using Pearson correlation coefficients (R^2 , p-values), was used to explore correlations among performance parameters such as stability, flow, ITS, and resilient modulus. For the purposes of this study, the Pearson correlation coefficient, the coefficient of determination, and the p-value were used to

assess the strength and statistical significance of the correlation between the parameters of the controlled mixture. Normalised indices and comparison indices were used to assess relative performance trends, and effect size analysis (Cohen's *d*) was used to compare the effects of the various additives, relative to the control and hybrid-modified mixture levels, when applied singly or in combination.

2.4 | Microstructural and Chemical Characterisation

Microstructural and mineralogical studies aimed to determine interactions between fillers and binders and the evolution of the asphalt mastic structure. The techniques used included X-ray diffraction analysis (XRD, ASTM C1365), X-ray fluorescence (XRF, ASTM C1254), and scanning electron microscopy coupled with energy-dispersive X-ray analysis (SEM-EDS, ASTM E986). The techniques allowed investigations of crystalline phase distribution, oxide composition and surface morphology/characteristic identifications and elemental mapping, respectively. XRD was used to analyse the distribution of mineral phases in the complex. XRF was used to analyse the chemical composition of the material. SEM-EDS provided analysis of the microstructure, including filler particle dispersion and morphology, the general shape and porosity of the fillers and the distribution of particular elements. The microstructure (via SEM-EDS) and the mineralogy (via XRF, XRD) have been qualitatively related to the test results, with interpretations used with care to avoid unfounded attributions of causality.

3. | Results and Discussion

3.1 | Overall Mix Performance and Group Behaviour

Figure 1 displays the variability of Marshall stability, flow, ITS, and RM for all 30 blends at 4%

and 7% binder content ($n = 15$). Focusing on the performance characteristics at the binder contents alone reveals the Marshall stability varied from 4.60-11.38 kN, translating to group means of 7.95 kN (BT 4%) and 7.20 kN (BT 7%) a 9.4% decrease in stability; in agreement with the increased binder binder, the Marshall stability decreases as a result of reduced aggregate interlock (Li *et al.*, 2022). Flow varied between 2.60-4.70mm, with mean values of 3.55mm (BT 4%) and 3.72mm (BT 7%), a 4.8% increase in deformation with higher binder quantities, consistent with previously described greater mixture ductility at higher binder contents (Sharma *et al.*, 2022). ITS varied between 251 and 418 kN/m, with little difference between group means (331 vs 333 kN/m), implying little sensitivity of mixture performance to binder quantity alone, corroborated by Joohari and Giustozzi (2021). RM varied between ~890 and ~2150 kN/m, with means optimistically dropping from ~1660 kN/m (BT 4%) to ~1550 kN/m (BT 7%) (6.6%), again confirming that higher binder contents reduce mixture stiffness, as found by Sudarsanan and Kim (2022). With the broad range of performance measurements observed both at individual binder contents and overall, it can be inferred that the nature of the modifiers influences performance variation more than the binder content.

3.2 | Comparative Performance of Control and Optimal Mix

Figure 1 (lower panels) depicts the R1 (control), R16 (modified control) and R17 (best hybrid) results. R17 showed 6.60 kN stability, 3.20 flow, 381 kN/m ITS, and 1470 kN/m RM with respect to control (R1) (+20.6% ITS, 17.9% flow). These results are consistent with the increased flexural tensile strength and reduced deformation observed in a hybrid polymer-rubber system, where adequate improvements in binder elasticity and cohesion were achieved (Joohari and Giustozzi, 2021; Sharma *et al.*, 2022). The incremental decreases in stability (7.0% reduction relative to control R1) and RM

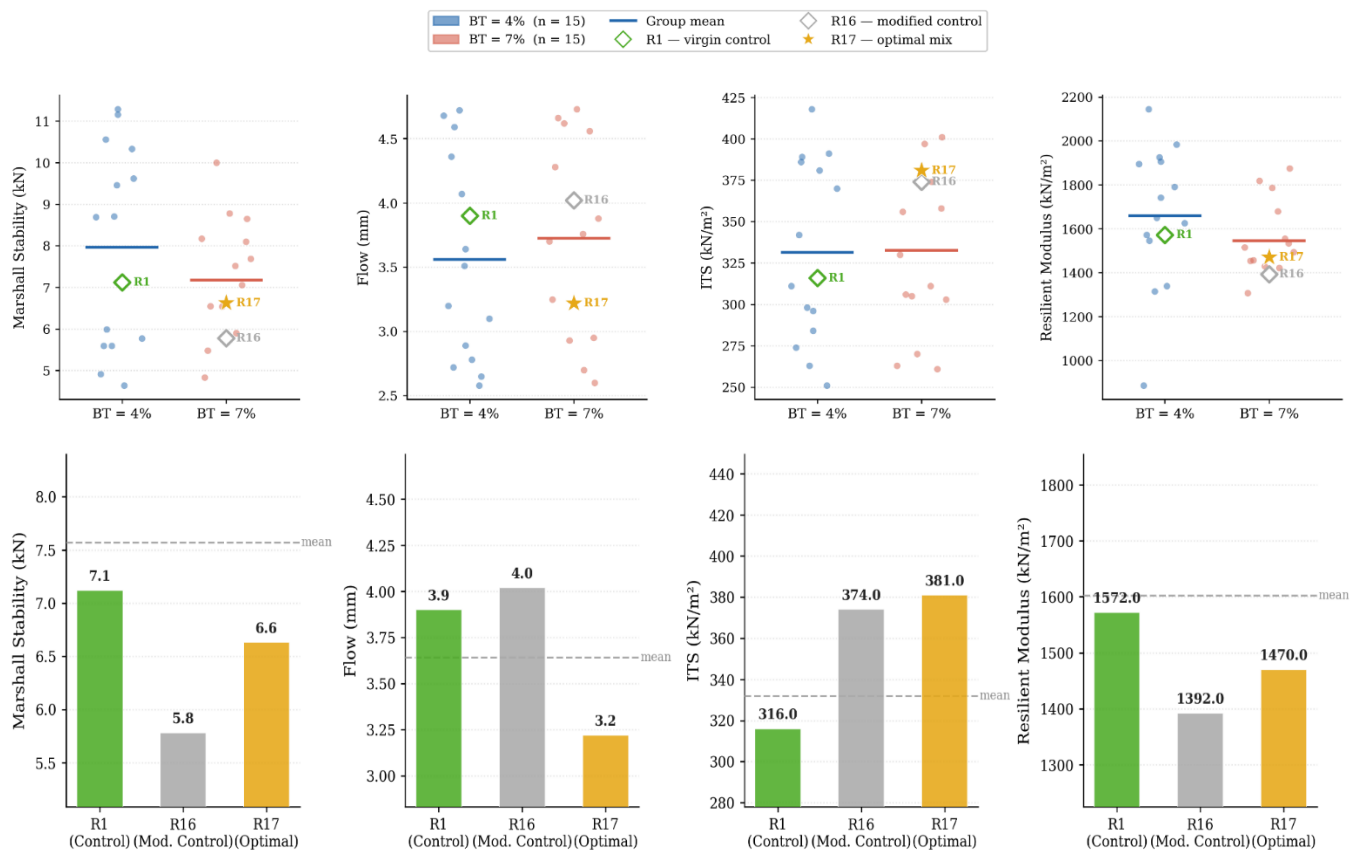


Figure 1 | Individual mix performance across all 30 samples, showing Marshall stability, flow, ITS, and resilient modulus for BT = 4% and 7% (n = 15 each). Horizontal bars denote group means; R1, R16, and R17 are highlighted.

(6.5%) reflect the softening effect of the rubber and thermoplastic modifiers, as previously observed in rubber-modified systems (Juveria *et al.*, 2023). The CoV remains unchanged, indicating good potential for a binder stiffness-flexibility balance, which is necessary to pre-empt fatigue cracking and rutting. Similar balances are established in a hybrid waste-modified system where balancing itself is more important than maximising (Zakerzadeh, 2025).

3.3 | Correlation and Statistical Relationships

Figure 2 revealed a clear trend of a significant positive correlation between stability and RM ($R = 0.827$, $R^2 = 0.683$, $p < 0.001$) as also observed in the literature, where stiffness was directly related to the load-carrying capacity of asphalt mixtures (Li *et al.*, 2022). The unbiased relationship with stability and ITS ($R = 0.013$) further established that tensile behaviour

is binder-controlled and insensitive to aggregate structure, consistent with the determination by Huan *et al.* (2019). Significant negative association with stability and rut depth ($r = -0.86$) as well as RM and rut depth ($r = -0.80$) established the asphalt behaviour that road stiffness is thought to decrease permanent deformation, while the inert relation of flow with stability and rut depth suggested that the deformation behaviour was complex, exhibiting distinct viscoelastic behaviour in polymer-modified/modified mix as seen elsewhere (Sharma *et al.*, 2022).

3.4 | Modifier Interaction and Performance Trends

Figure 3 shows the nonlinear interactions among lignite, WPT, and WOT. Lignite remained stable (~4-6%) and reduced until (~24%), similar to the

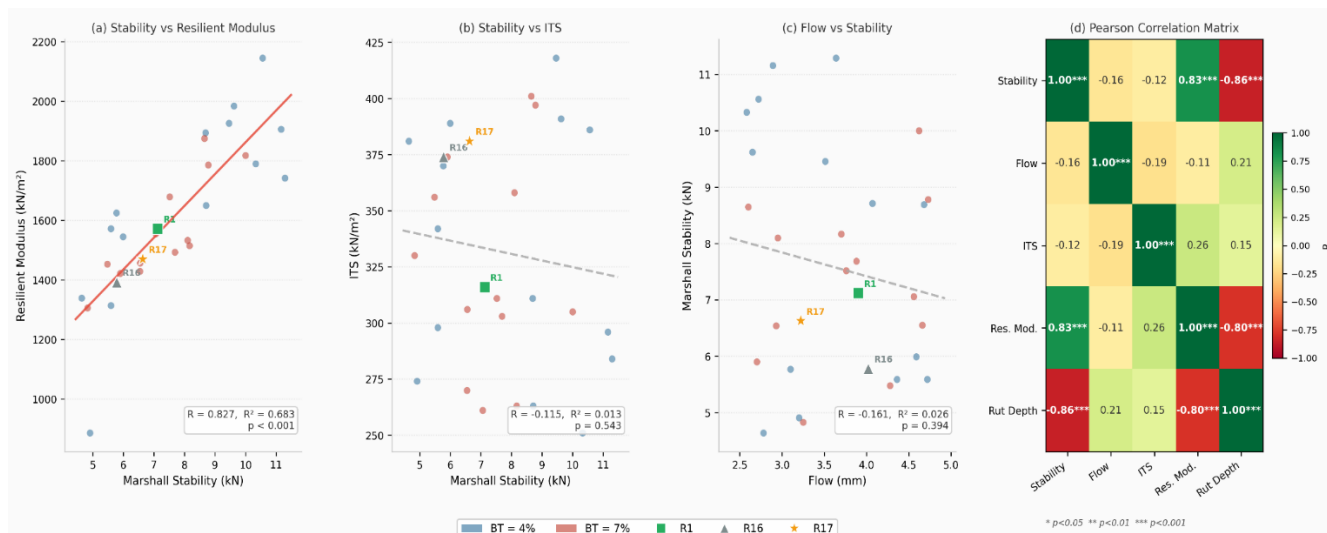


Figure 2 | Bivariate correlations and Pearson matrix (N = 30), showing relationships among stability, flow, ITS, resilient modulus, and rut depth. Significant relationships are indicated by solid regression line

effects of filler agglomeration seen in mineral-modified systems (Safiuddin *et al.*, 2016). WPT improved ITS (+15-20%) by increasing binder cohesion & adhesion similar to the effects of plastic modification on asphalt (Aziz *et al.*, 2015; Huan *et al.*, 2019). WOT improved ductility but reduced stability at higher WOT contents (22-25% reduction at 20% WOT). This was also reported by other researchers, who found that rubber caused a disproportionate softening compared with other modifiers (Juveria *et al.*, 2023; Singaravel *et al.*, 2024). The results indicated that the hybrid modification produced opposing yet co-existing phenomena, evidenced by the increase in stiffness from lignite, which was offset by the ductility of the two elastomers, pointing to multi-material optimisation, as discussed by Nkomo (2022).

3.5 | Performance Optimisation and Effect Size Analysis

Normalised performance and effect size analysis (Figure 3) indicate that R17 shows 93% stability, 83% flow, 121% ITS, and 94% RM compared with the control. The large effect size for ITS ($d > 0.8$) and the moderate-to-large effect size for flow reduction ($d \approx -0.7$ to -1.0) demonstrate significant engineering

gains. Such effect sizes have recently been reported for hybrid waste-modified systems, in which gains in tensile and deformation characteristics have outweighed slight reductions in stiffness (Bastidas-Martínez *et al.*, 2022). The radar plot confirms that R17 exhibits a balanced performance profile, without the extremes apparent when flows are achieved at the expense of other properties. This affirms the potential for performance optimisation through hybridisation, a theme that has been prominent in recent advanced investigations into sustainable asphalt (Wong and Peng, 2022).

3.6 | Engineering Implications

The optimal mix (R17) achieved +20.6% ITS and 17.9% flow, with comparable control stiffness (6-7%). This mixture exhibited good resistance to two different failure mechanisms (cracking and rutting) simultaneously. Similar results were observed in research on other hybrid polymer-rubber systems, which reported increased durability due to the combination of stiffness and flexibility (Joohari and Giustozzi, 2021; Sharma *et al.*, 2022). Overall, the results showed that hybrid waste modification can, in many cases, optimise multiple performance parameters simultaneously, rather than just

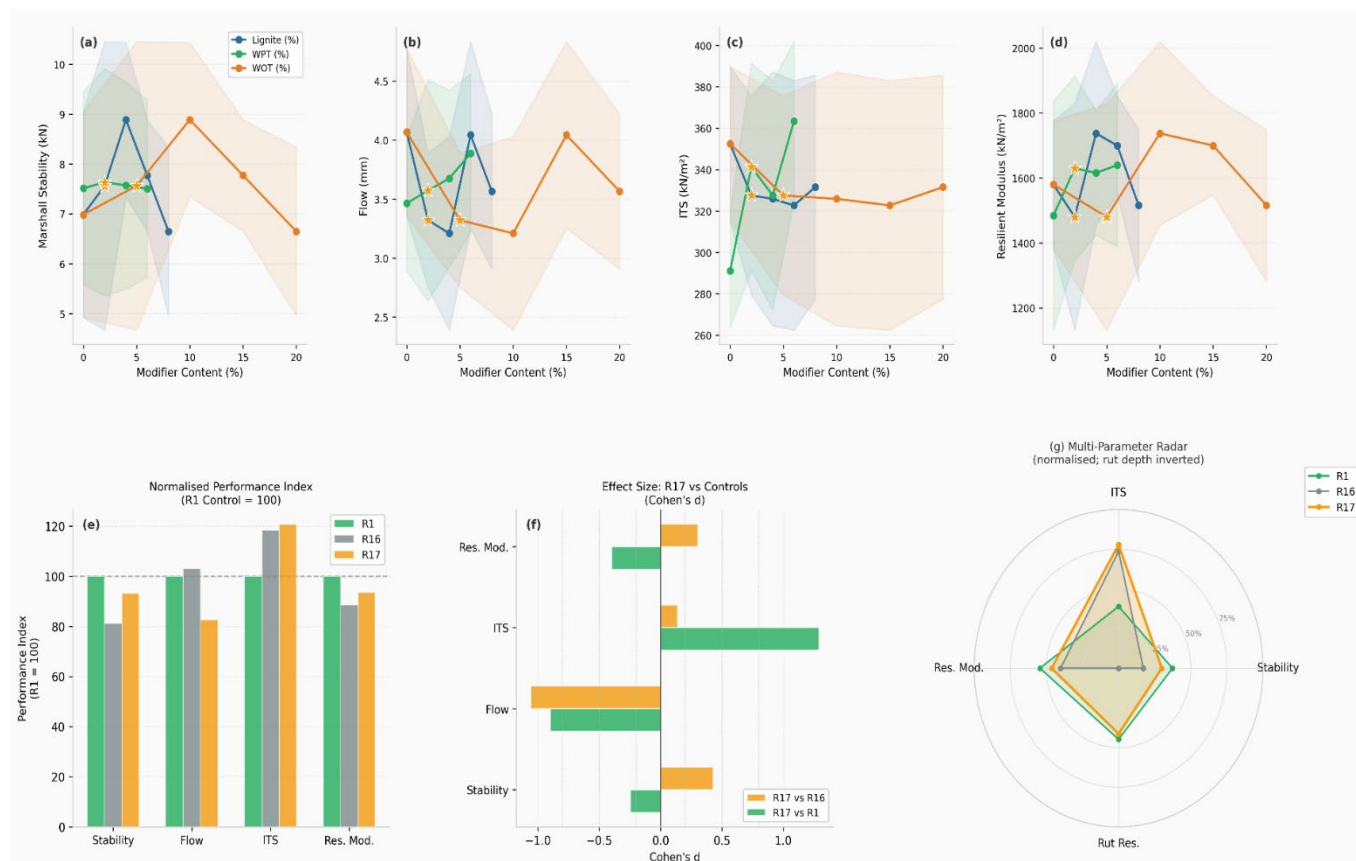


Figure 3 | Modifier interaction profiles, normalised performance indices, effect size (Cohen's d), and radar-based comparison of R1, R16, and R17.

maximising a single property. The increasing focus on multi-parameter optimisation in sustainability experiments aligns with the accelerating pace of circular material use in modern pavement design and engineering (Bamigboye *et al.*, 2021; Wong and Peng, 2022).

3.4 | Microstructural Characterisation

3.4.1 | X-Ray Diffraction (XRD)

XRD data (Table 2 and Figure 4) reveal mostly crystalline SiO_2 quartz, orthoclase (KAlSi_3O_8), muscovite ($\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$) and C graphite phases due to the mineral fillers, whilst the largely amorphous asphalt binder (containing polymer modifiers) shows few or no peaks. The 2 θ range of $2\theta \approx 9\text{--}11^\circ$ and $24\text{--}30^\circ$ correspond to various silicate phases, with relatively similar intensity

quartz peaks ($\approx 24\text{--}30^\circ$) between the modified mixes and control, inferring better mineral dispersion rather than increased binder crystallinity. The presence of weaker calcite and feldspar peaks in the modified mixes is consistent with inferred mineral contributions from lignite and tyre-derived pyrolysis residues. Crystallite sizes in the 263–1387 Å range, with R17 at ~ 963 Å, were recorded for all asphalt-filler systems, indicating moderate structural uniformity. Convergence of the individual crystallite sizes of the modified asphalt mixtures is similar to the stronger structural stability of pyrolysed carbon materials, whilst the high interfacial interaction was evidenced by relatively similar weak peak intensities in the activated carbon filler system and increases in asphalt crystalline mineral phases (Ungureanu *et al.*, 2025; Malpani *et al.*, 2023). The enhanced peak definition coincided with increased resilient moduli (5.6% relative to R16) and a moderately

strong correlation between the mixture stability and modulus ($R^2 = 0.683$), although correlation rather than causality should be emphasised. Therefore, overall, the XRD data showed higher crystalline phase content, a more even distribution

of crystalline mineral phases, and enhanced mineral filler matrix interaction; these phenomena, inferred from the structural information, complement the observed differences in the surface physicochemical properties of the asphalt-binder phases.

Table 2 | XRD Results for Raw Materials and Asphalt Samples

Sample ID	Key Peaks (2 θ)	Dominant Phases	Crystallite Size Range (Å)	Modification Type
WOT	38.05°, 44.25°	Brianyoungite, Graphite, Muscovite	363 – 702	Raw Additive
Lignite	24.07°, 27.89°, 30.84°	Carbonaceous Organics, Quartz, Orthoclase, Sulphur, Muscovite	8.36 – 145.62	Raw Additive
R1	9.37°, 11.05°, 28.62°	Quartz, Anorthite, Orthoclase, Muscovite, Graphite	758	Control (4% BT, No Additives)
R10	9.06°, 24.10°, 27.69°, 28.27°	Quartz, Orthoclase, Anorthite, Muscovite, Graphite	384 – 1387	Modified (WOT)
R16	23.45°, 24.29°, 26.88°, 28.30°	Quartz, Orthoclase, Anorthite, Muscovite	263 – 1086	Control (7% BT, No Additives)
R17	24.34°, 58.94°	Quartz, Graphite, Orthoclase, Anorthite, Muscovite	963	Hybrid (7% BT, 2% L, 5% WOT, 2% WPT)
R25	28.61°, 36.96°	Quartz, Graphite, Orthoclase, Anorthite, Muscovite	807 – 1326	Hybrid (7% BT, 2% L, 4% WPT, 20% WOT)

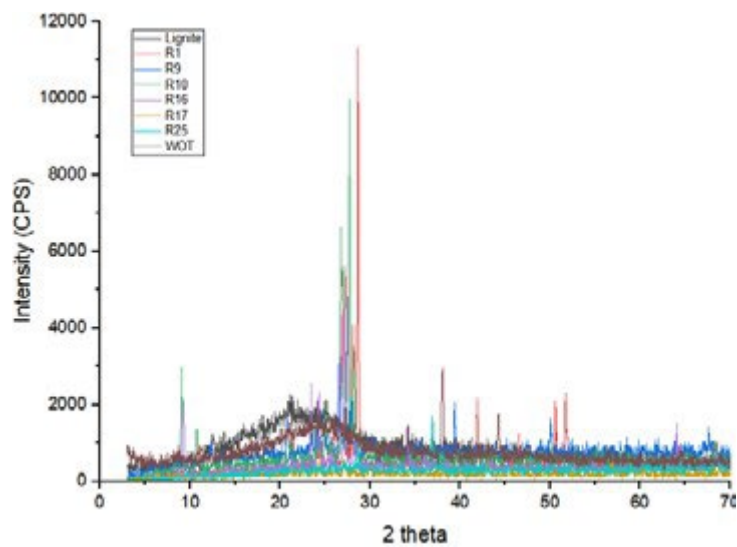


Figure 4 | Combined XRD analysis of selected samples

3.4.2 | X-Ray Fluorescence (XRF)

XRF analysis (Table 3) demonstrates that control and hybrid mixtures are made up of high levels of oxides: SiO₂, Al₂O₃, and Fe₂O₃.. SiO₂ (47.85–62.40%, is particularly high and tends to be slightly higher in modified samples, while Al₂O₃ (~12-14%) and FeO (~7-12%) are also high. CaO shows a significant increase from 5.58% (R1) to 8.11 % (R17) (45% increase). The level of silica is high, which is

indicative of improved stiffness and stability, as demonstrated for lignite-derived materials (Jankovsk *et al.*, 2024). If CaO is found to indicate finer filler-binder interaction, it could be suggested that this may also indicate improved moisture resistance (albeit the ultimate performance is determined by the efficiency of the binder). Given XRF measures mineralogical composition rather than mechanical behaviour, this is merely supporting evidence of the mechanical behaviour demonstrated.

Table 3 | X-ray Fluorescence (XRF) Analysis Results

Oxide (%)	Lignite	R1	R9	R10	R16	R17	R25	WOT
SiO ₂	33.35	59.34	57.31	64.80	64.14	53.40	47.85	37.72
Al ₂ O ₃	13.99	12.65	14.17	12.33	12.80	14.39	12.02	4.23
Fe ₂ O ₃	13.69	9.55	12.54	8.11	7.49	9.67	11.58	2.85
CaO	3.36	5.58	6.60	5.27	7.04	8.11	14.64	5.56
K ₂ O	0.76	6.48	5.86	6.02	5.39	7.61	4.57	0.75
SO ₃	25.61	2.11	0.55	0.66	0.55	1.40	2.65	22.85
TiO ₂	5.15	1.03	1.07	0.92	0.91	1.44	1.25	0.30
Cl	2.16	2.15	0.60	0.73	0.74	3.19	0.59	1.59
ZnO	0.04	0.03	0.03	0.01	0.01	0.02	0.02	22.88
MgO	0.00	0.00	0.00	0.16	0.00	0.00	4.11	0.00
MnO	0.48	0.14	0.21	0.10	0.12	0.19	0.18	0.04
CuO	0.23	0.04	0.04	0.03	0.05	0.06	0.04	0.10
ZrO ₂	0.28	0.17	0.17	0.14	0.17	0.20	0.07	0.03
P ₂ O ₅	0.00	0.15	0.29	0.15	0.18	0.02	0.18	0.09
V ₂ O ₅	0.23	0.04	0.09	0.06	0.08	0.06	0.04	0.01
Cr ₂ O ₃	0.04	0.15	0.08	0.16	0.08	0.03	0.07	0.01
Co ₃ O ₄	0.08	0.04	0.05	0.03	0.03	0.03	0.04	0.19
Ag ₂ O	0.15	0.04	0.03	0.01	0.02	0.03	0.00	0.01
Nb ₂ O ₃	0.09	0.02	0.02	0.01	0.01	0.01	0.01	0.01
Ta ₂ O ₅	0.12	0.03	0.02	0.02	0.05	0.05	0.00	0.23
WO ₃	0.00	0.00	0.00	0.01	0.00	0.00	0.02	0.56

3.4.3 | Scanning Electron Microscopy (SEM-EDS)

SEM images (Figure 5) confirm differences in the morphology of the control and hybrid-modified mixes. The control asphalt exhibits rough binder-aggregate interfaces and large pores (≈5–20 μm),

consistent with weak binder-aggregate adhesion and high porosity. The hybrid modification in R17 has improved homogeneity (a denser matrix) and reduced visible porosity. The micro-fines present in the lignite opened pores and filled voids (contributing to a densified matrix), and WPT and WOT appear to create a more continuous binder phase.

This suggests that a polymer-modified network has formed at the microscopic level, which may be useful for interfacial bonding and stress transfer; this is not measured here. EDS data show a high percentage of carbon in the four tests (~84-94 atom%), with silicon (~3-8 atom%) and aluminium making up the residual. Results from line-scanning microscop-

ic analysis show that the phases present are more evenly dispersed relative to one another in R17 than in the control. The morphology of the hybrid-modified asphalt is consistent with a 20.6% increase in ITS and a 17.9% reduction in flow, which will result in increased resistance to cracking and deformation.

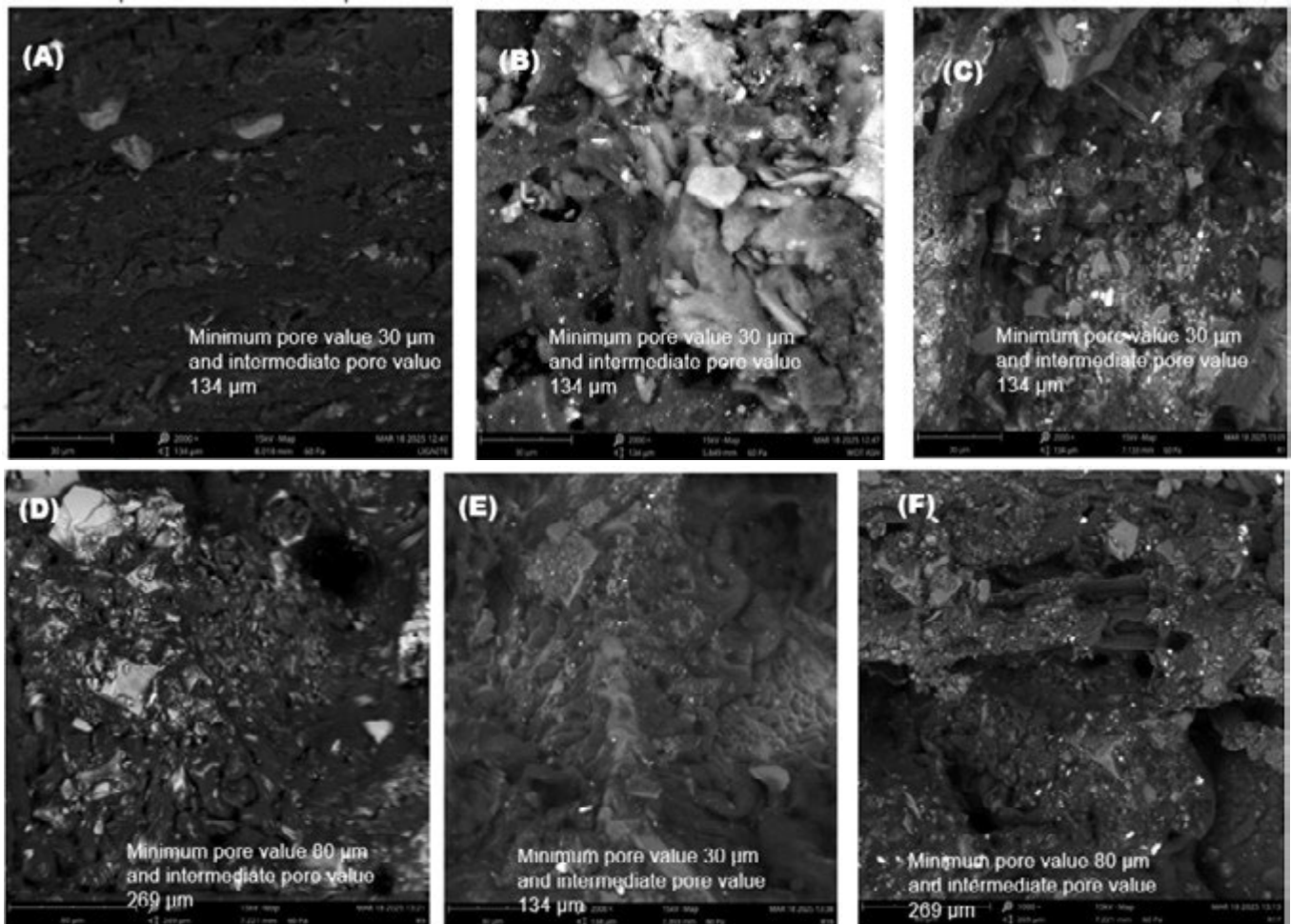


Figure 5 | SEM-EDS micrographs of control and hybrid-modified asphalt samples. (A) Lignite sample; (B) WOT (worn-out tyre) sample; (C) Control mix (Sample R1); (D) Hybrid mix R9; (E) Hybrid mix R10; (F) Hybrid mix R16; (G) Optimum hybrid mix R17; (H) Hybrid mix R2

4 | Conclusion

This research focused on the use of lignite, waste polythene (WPT), and worn-out tyre rubber (WOT) hybrids in asphaltic concrete. Mixture proportions were generated using Latin hypercube sampling (LHS) and tested for mechanical and microstructural responses. It was found that hybrid

modifier optimisation can enhance performance by balancing the influence of WPT and WOT and their interaction. The identified optimum mix ratio of (R17: 7% bitumen, 2% lignite, 2% WPT, 5% WOT) give the most balanced response with the 20.6% increase in indirect tensile strength (ITS) and 17.9% decrease in flow compared to control while maintaining resilient modulus (1470 kN/m) within

6.5% of the control and Marshall stability (6.60 kN) within similar proportions. Correlations between performance parameters suggest a strong correlation for stability and resilient modulus. ($R^2 = 0.683$, $p < 0.001$), but only a weak correlation for stability and ITS ($R^2 = 0.013$), indicating that tensile strength performance is dominated by binder modification. Microstructural results support these findings, with XRD confirming the dominance of silica-rich phases; WPT-modified samples achieving SiO contents ranging from 47.85 to 62.40% according to XRF analysis; and SEM-EDS showing decreased porosity and the presence of more carbonaceous and mineral phases. The control parameters identified as conducive to high performance are characteristics of durable asphalt mixtures with improved resinous properties and enhanced deformation response,

as required for high-stress loading applications. From a sustainability perspective, the application of lignite, WPT and WOT as waste modifiers is encouraging as it could allow less dependency on virgin materials; however this work is limited by the single samples tested per mix and it would be better completed with replicate testing for statistical analysis, suitable ageing studies carried out to assess long term behaviour, moisture susceptibility tests, and field validation trials for performance and life cycle testing. Nonetheless, this work has demonstrated how performance can be optimised through the controlled interaction between mineral and polymer phases via hybrid waste modification in asphalt, providing an alternative means of developing durable, more environmentally friendly pavements.

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