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# From Bin to Binder: Unleashing Waste Butter's Potential as a Pioneering Bio-Modifier for Sustainable Asphalt Engineering

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**Abstract:** Exploring the interface of environmental sustainability and civil infrastructure development, this study introduces waste butter (WB), a byproduct of animal fat processing, as a novel bio-modifier in asphalt production. This approach not only recycles animal waste but also charts a course for sustainable infrastructural development, contributing to a reduced environmental impact and promoting circular economy practices. The experiments incorporated varying WB concentrations (e.g., 3%, 6%, and 9% by weight of binder) into standard AP-5 asphalt, employing advanced analytical tools for comprehensive characterization. These included thin-layer chromatography–flame ionization detection (TLC-FID), Fourier-transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and Differential Scanning Calorimetry (DSC). The critical properties of the asphalt blends, such as penetration, softening point, viscosity, ductility, rutting factor (Dynamic Shear Rheometer), and thermal susceptibility (Penetration Index, Penetration–Viscosity Number), were assessed. FT-IR analysis indicated negligible chemical alteration with WB addition, suggesting predominantly physical interactions. TLC-FID showed a decrease in aromatic and asphaltene components but an increase in resin content, highlighting the influence of WB's fatty acids on the asphalt's chemical balance. The colloidal instability index ($I_C$) confirmed enhanced stability due to WB's high resin concentration. Meanwhile, SEM analysis revealed microstructural improvements with WB, enhancing binder compatibility. TGA demonstrated that even a minimal 3 wt. % WB addition significantly improved thermal stability, while the DSC results pointed to improved low-temperature performance, reducing brittleness in cold conditions. Rheologically, WB incorporation resulted in increased penetration and ductility, balanced by decreased viscosity and softening point, thereby demonstrating its multi-faceted utility. Thermal susceptibility tests emphasized WB's effectiveness in cold environments, with further evaluation needed at higher temperatures. The DSR findings necessitate careful WB calibration to meet Superpave rutting standards. In conclusion, this research positions waste butter as a superior, environmentally aligned bio-additive for asphalt blends, contributing significantly to eco-friendly civil engineering practices by repurposing animal-derived waste.

**Keywords:** waste butter; bio-modifier; AP-5 asphalt; TLC-FID; FT-IR; SEM; thermodynamic stability; rheological properties

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## 1. Introduction

In contemporary environmental discourse, there is an escalating emphasis on sustainable waste management. This imperative compels industries across the spectrum to re-evaluate their operational modalities, technological methodologies, and material selections. A sector that stands prominently within this discourse is the agro-food industry, which, together with domestic households and gastronomic establishments, grapples with the sizable production and subsequent disposal of oils and fats [1,2]. Butter, a ubiquitous culinary component, is frequently relegated to waste due to factors ranging from overproduction, limited shelf-life, and storage mismanagement to non-compliance with rigorous quality or the aesthetic benchmarks stipulated by retailers [3,4]. To quantify this issue, global statistics indicate that annually, several hundred thousand metric tons of butter are either discarded or rendered obsolete [5,6].

The environmental consequences of ill-managed waste oils, encompassing factors ranging from waste butter to used cooking oils, are multifaceted [7]. These encompass aquatic ecosystem degradation from waterborne contamination, the diminution of soil fertility, and a noteworthy augmentation to the inventory of global greenhouse gas emissions [8]. Reflecting upon the magnitude of this dilemma, the daily discarding of fats and oils is not solely an attribute of large-scale industrial activities but a byproduct of household and commercial culinary undertakings [9]. Beyond the evident ecological repercussions, this scenario elucidates a pronounced inefficiency in resource optimization. Yet, within this quandary lies an innovative prospect that remains underexplored: the potential integration of waste butter in road pavement compositions.

The domain of road construction has historically been a hotbed for investigating additives aimed at amplifying both the physiochemical and mechanical attributes of pavements. Notable among these are polymers [10,11], crumb rubber [12], and specific industrial byproducts, each evaluated for their capacity to fortify asphalt's resistance to challenges, such as wear, temperature variance, and mechanical stress. Amidst this gamut of additives, waste butter, characterized by its abundance of fatty acids or, eventually, resins, emerges as a compelling contender [13,14]. The innate constitution of butter, encompassing both saturated and unsaturated fatty acids, as well as additional constituents like water, milk solids, and occasionally salt [15], possesses properties suggestive of its potential efficacy as an asphalt additive. Specifically, its lipidic profile, predominantly saturated fats, can potentiate viscoelasticity [16]. Further, the distinct physiochemical attributes of butter—its melting point, textural consistency, and resistance to oxidative aging [17]—underscore its potential to augment asphalt's overall elasticity and resilience against common roadway deformities.

The historical surveying of bio-additive applications in road construction revealed many materials that have garnered investigative interest. Waste vegetable oils have previously been the focus of rejuvenation studies to restore aged bitumen's malleability [18,19]. Concurrently, tall oil, a byproduct of the pulp and paper sector, has been evaluated for its prospective merits in enhancing the adhesive properties of road binders [20,21]. These investigative endeavors collectively pave the way for a holistic examination of waste butter in asphaltic compositions—a frontier that, as of the present, remains comparatively embryonic in both academic and industrial arenas.

In this manuscript, our ambition is to meticulously navigate this relatively unexplored domain. Our inquiry is bifurcated: to discern whether waste butter can serve as an efficacious and sustainable substitute to traditional asphalt additives and to highlight the potential ecological benefits inherent in the reconversion of a prevalent waste material into a purposeful application. Through scrupulous analyses, we aspire to elucidate waste butter's potentiality as both an environmentally astute selection and a potential linchpin in pioneering sustainable road construction methodologies.

## 2. Materials and Methods

### *2.1. Asphalt Bio-Modifier Preparation from Waste Butter (WB)*

The transformation of waste butter (WB), primarily derived from dairy industries, into a promising bio-modifier for asphalt involves a systematic sequence of controlled thermal processing, meticulous purification, and filtration procedures [22,23]. The streamlined methodology, vividly depicted in Figure 1, has been designed precisely to preserve the modifier's intrinsic qualities and efficacy. Table 1 provides a comprehensive analysis of the nutritional, elemental, fractional, and physical characteristics of WB for an in-depth perspective.


**Figure 1.** Streamlined conversion of waste butter (WB) from dairy industries into a bio-modifier for asphalt formulations: a detailed procedural insight. (**A**,**B**) Collection phase: partnering with local dairy producers to procure an ideal starting volume of 1 kg for extraction and assay initiatives. (**a**,**b**) Thermal treatment: methodically melting the waste butter within a 60–70 °C bracket for a targeted span of 5–10 min, ensuring the intactness of fatty acids. (**c**) Settling interval: allocating a 20–30-min duration post-melting for the natural separation of milk foam and sedimented particulates. (**d**) Precision filtration: implementing an ultra-fine nylon mesh strainer initially, succeeded by filter paper sieving; multiple iterations may be pursued for optimal clarity. (**e**,**f**) Culmination: yielding a refined bio-modifier, expertly prepared for integration into asphalt formulations and subsequent performance evaluation.

The first critical stage is the collection of waste butter. Engaging with a consortium of local diary industries offers an optimal strategy, ensuring a consistent and ample supply of this primary material. For the purposes of this research, we targeted an initial collection of approximately 1 kg. This quantity not only facilitates the extraction process but also ensures that there is a sufficient amount for subsequent testing phases.

Following the collection, the raw waste butter undergoes a heating phase to transition it into a liquid state. Maintaining a temperature range of 60–70 °C is crucial. This specific range ensures the effective melting of the butter while preventing any undesired thermal degradation of its inherent fatty components. Depending on the initial consistency and quantity of the waste butter, a heating duration of 5 to 10 min is typically adequate.

Once in its liquid state, the next challenge is removing impurities. Waste butter, especially from dairy industry sources, often contains remnants of milk foam and other minor contaminants. To address this, the heated butter is allowed to settle for 20 to 30 min. During this time, most of the milk foam and heavier particulates naturally rise to the surface or settle at the bottom, making the subsequent purification phase more effective.

The final step in this preparatory journey is filtration. Using an ultra-fine nylon mesh strainer or a similar filtration apparatus, the liquid butter is carefully filtered to remove any lingering impurities. The process might require multiple passes through a filter paper to ensure a pure, uncontaminated bio-modifier.

Upon completion of these stages, what remains is a clarified bio-modifier, derived from waste butter and poised for integration and testing in asphalt formulations.

**Table 1.** Comprehensive nutritional, elemental, fractional, and physical profile of waste butter (WB).

| Nutrition Facts (per 100 g WB) | Mean ± SD |
|---|---|
| Total fat | 85.00 g |
| Saturated fat | 6.50 |
| *Trans* fat | 70.00 g |
| Cholesterol | 210.00 mg |
| Sodium | 10.00 mg |
| Total carbohydrate (traces of lactose) | ≤1.00 g |
| Total sugars | ≤1.00 g |
| Protein (e.g., casein) | ≤1.00 g |
| Calories | 760 Cal |
| **Elemental Analysis** | |
| C (Carbon) | 56.85 ± 0.67 wt. % |
| H (Hydrogen) | 8.80 ± 0.56 wt. % |
| N (Nitrogen) | 0.05 ± 0.00 wt. % |
| S (Sulfur) | 0.00 ± 0.00 wt. % |
| O (Oxygen) | 8.48 ± 0.48 wt. % |
| **SARA Generic Fractions** | |
| Saturates | 0.00 ± 0.00 wt. % |
| Aromatics | 0.00 ± 0.00 wt. % |
| Resins | 100.00 ± 0.00 wt. % |
| Asphaltenes-like components | 0.00 ± 0.00 wt. % |
| **Physical Properties** | |
| Softening point | 34.20 ± 0.23 °C |
| Rotational viscosity at 35 °C | 100.50 ± 5.73 cP |
| Rotational viscosity at 135 °C | 0.00 ± 0.00 cP |
| Melting point | 34 °C |
| Solubility | Insoluble in water |
| Density at 25 °C | 0.9110 g $cm^{-3}$ |
| Color | Golden hue |
| Flavor | Rich–creamy |
| Odor | Milk–creamy |

### *2.2. X-ray Diffraction Analysis of Waste Butter Peaks*

X-ray diffraction (XRD) was deployed to elucidate the crystalline structures inherent within the waste butter. For this, the Bruker AXS D8 Advance Diffractometer (Bruker AXS GmbH D8 Advance, Karlsruhe, Germany) was utilized, operating with a Cu-Kα radiation source (λ = 1.54005 Å) at a voltage of 40 kV and a current of 40 mA. The resultant diffraction scans spanned a 2θ range from 10° to 90° and advanced at 1° $min^{-1}$.

In the spectrum displayed in Figure 2, distinctive peaks can be identified, indicative of the complex crystalline nature of butter fats [24–26]. Notably, the recorded peaks were pinpointed at 2θ values of 19.55°, 20.22°, 20.84°, 21.59°, and 23.23°. Their clustering and relative proximities hint at the multifaceted polymorphic configurations present in the sample. For instance, the peak at 19.55° could be attributed to the α form, which is often characterized by its less-ordered structure, especially observed in fats that have recently crystallized. The subsequent peaks at 20.22° and 20.84° might signify various transitional stages or nuances within the β′ (prime) phase, known for its semi-ordered crystalline association with medium-chain saturated fats. This phase gives butter its characteristic softer texture.

Furthermore, the 21.59° peak can be perceived as another variant or orientation of the β′ phase, underlining the diversity of this polymorphic form within butter samples. Lastly, the emergence of the peak at 23.23° alludes to the presence of the β form. This crystalline structure is typically linked to long-chain saturated fats and is distinguishable by its organized structure, resulting in a more rigid texture.

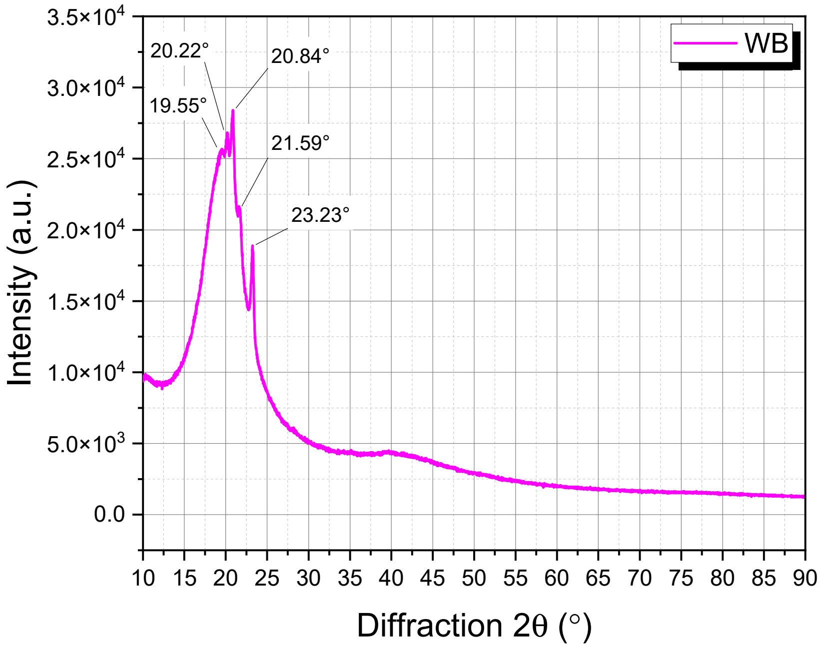


**Figure 2.** XRD profile of waste butter (WB).

In essence, understanding these crystalline nuances through XRD offers an invaluable perspective on waste butter's potential as an asphalt additive. Such insights can help predict its thermal stability, binding prowess, and overall performance within innovative asphalt formulations.

### *2.3. Materials, Preparation, and Formulation of Waste-Butter-Infused Asphalt Blends*

The accredited South Korean Federation of Ascon Industry Cooperative R&D Center, situated in Osan-si, Gyeonggi-do, graciously provided the base AP-5 asphalt (PG 70–10) for our research endeavors. A comprehensive overview of its quintessential physicochemical attributes can be found in Table 2. On a parallel note, waste butter (WB), denoted as such in subsequent references and visually represented in Figure 1A,B, was collected from a local dairy industry in Cheonan City, South Korea. Table 1 furnishes detailed insights into the nutritional–chemical–physical specifications and the fundamental composition of WB, respectively.

To formulate WB–asphalt blends, we employed an L5M-A mixer, courtesy of Silverson Machines Inc., East Longmeadow, MA, USA. This mixer boasts a robust 3000 rpm high-shear speed. Alongside this, a heating mantle of model GLHMD-B100 by Global Lab Co., Ltd., located in Siheung-si, Gyeonggi-do, Republic of Korea, maintained an operational temperature of 180 °C, mirroring the standard production temperature for Hot-Mix Asphalt (HMA). To achieve optimal fluid dynamics during the mixing phase, the control binder was pre-heated in an oven for a duration of 2 h at a stable 140 °C. This meticulous step ensured that the blend retained ideal liquidity while minimizing oxidation effects.

The blends were prepared using 1000 mL cylindrical aluminum containers, each loaded with roughly 600 g of molten bitumen and systematically heated from 140 to 175 °C [27–29]. Various proportions of waste butter (for instance, 3 wt. %, 6 wt. %, and 9 wt. % WB by the blend's total weight) were systematically integrated into the control bitumen at the aforementioned temperature [30–32]. The rationale behind choosing these varying concentrations was due to the initial unknowns surrounding the base formulation of WB and its reactive behavior with the binder and thermal conditioning. Additionally, the considerable variance in concentration was strategically selected to furnish a comprehensive evaluation of the blend's performance in terms of parameters, like physicochemistry, microstructure, thermo-morphology, and rheology.

**Table 2.** Physicochemical properties of the base AP-5 asphalt cement.

| Elemental Analysis | Mean ± SD |
|---|---|
| C (Carbon) | 86.62 ± 1.62 wt. % |
| H (Hydrogen) | 10.78 ± 0.19 wt. % |
| N (Nitrogen) | 0.51 ± 0.02 wt. % |
| S (Sulfur) | 5.65 ± 0.05 wt. % |
| O (Oxygen) | 0.90 ± 0.03 wt. % |
| **SARA Generic Fractions** | |
| Saturates | 3.35 ± 0.28 wt. % |
| Aromatics | 59.62 ± 1.88 wt. % |
| Resins | 16.77 ± 2.00 wt. % |
| Asphaltenes | 20.22 ± 1.79 wt. % |
| Colloidal Instability Index ($I_C$) ‡ | 0.3085 |
| **Physical Properties** | |
| Penetration at 25 °C | 69.83 ± 0.68 dmm |
| Softening point | 47.90 ± 0.11 °C |
| Rotational viscosity at 135 °C | 71.00 ± 1.06 cP |
| Ductility at 25 °C | 130.50 ± 0.00 cm |
| Density at 25 °C | 1.00 ± 00 g $cm^{-3}$ |

‡ $I_C$ = ([Saturates] + [Asphaltenes])/([Aromatics] + [Resins]).

Given the culmination of insights from this study, a judicious determination will be made regarding the ideal quantity that promises the most commendable engineering properties. Conclusively, both the untreated and WB-infused asphalt samples underwent a 2 h mixing process at 180 °C to ensure blend homogeneity [27–29]. The two-hour mixing duration for untreated asphalt was implemented to maintain consistent experimental conditions, which is crucial for ensuring the reliability of comparative assessments. This approach was applied universally across all sample types to eliminate procedural biases, facilitating a clear and unbiased evaluation of the waste butter's influence on asphalt properties. The resultant bituminous samples were decanted into multiple cans, subsequently distributed into sealed metal tins, and stored at room temperature (approximately 25 °C), positioning them for future evaluations and assorted tests post-preparation.

### *2.4. Analytical Evaluation of Waste Butter Proportions on Base AP-5 Asphalt Using TLC-FID Analysis*

The influence of varied proportions of waste butter (e.g., 3, 6, and 9 wt. % WB) on the chemical composition of base AP-5 asphalt was comprehensively analyzed using the thin-layer chromatography with flame ionization detection (TLC-FID) analyzer from Iatron Laboratories, Tokyo, Japan. This device was fitted with a metallic rack holding silica rods (Type Chromarod-S5) sourced from LSI Medience Corporation, Tokyo, Japan.

These chromarods, having a length of 15.2 cm, particle size of 5 μm, and pore diameter of 60 Å, underwent a rigorous activation and cleansing process using the hydrogen ($H_2$) flame from the FID to eradicate potential residues and impurities [33]. This meticulous step was crucial for ensuring consistency and precision in the outcomes. The system operated on high-purity hydrogen and air, supplied at flow rates of 160 mL $min^{-1}$ and 2 L $min^{-1}$, respectively, via an integrated pump [33].

A sample solution was prepared at a 2% (*w*/*v*) concentration using either bitumen/dichloromethane or waste butter/dichloromethane. Using a 5 μL microdispenser from Drummond Scientific, Broomall, PA, USA, a 1 μL aliquot of this solution was deposited onto the chromarod, serving as the stationary phase [33].

Subsequently, the chromarod underwent a thorough immersion in three separate development tanks filled with *n*-hexane (70 mL, 45 min), toluene (70 mL, 17 min), and a methanol/dichloromethane mix (3.5/66.5 mL, 5 min) [33]. This immersion process was essential for the sequential extraction of saturates, aromatics, and resins, while asphaltenes remained unaffected at the initial application site on the chromarod. Upon exposing the constituents on the chromarod to the $H_2$ flame, distinct organic ions materialize, which are then transformed into identifiable current intensities by the FID. The peaks representing these compounds were meticulously recorded and integrated, enabling subsequent determination of the saturates, aromatics, resins, and asphaltenes (SARA wt. %) concentrations [33].

Following each immersion, the rod rack was placed in a drying chamber at 85 °C for about 2 min to remove residual solvents. For every asphalt sample modified with waste butter, five replicates were analyzed, each with a scan duration of 30 s. The entire Iatroscan assessments were conducted four times for accuracy [33].

### *2.5. FT-IR Spectroscopic Profiling of Base AP-5 Asphalt with Waste Butter Concentrations*

The Hyperion (3000 FT-IR) Spectrometer from Bruker Optics, Ettlinger, Germany, was used to analyze samples with a spectral resolution of 1 $cm^{-1}$, spanning wavenumbers from 4000 to 650 $cm^{-1}$, averaging 30 scans per sample. Combined with KBr, a thin disc of the sample was prepared to chemically profile both the waste butter and the waste-butter-modified asphalt samples. The FT-IR analysis was conducted to understand the effects of integrating various concentrations of waste butter (e.g., 3, 6, and 9 wt. % WB) on the chemical composition and structure of the base AP-5 asphalt.

### *2.6. Scanning Electron Microscopy (SEM) and EDXS Evaluation of Waste-Butter-Integrated AP-5 Asphalt Nanostructures and Elemental Composition*

To elucidate the nuanced effects of incorporating specific concentrations of waste butter (e.g., 3, 6, and 9 wt. % WB) on the elemental framework and surface nanostructure of the pristine base AP-5 bitumen, the advanced JSM-6010LA SEM (JEOL Ltd., Tokyo, Japan), integrated with energy-dispersive X-ray spectroscopy (EDXS), was employed. For optimal electron microscopy visualization, each of the diverse asphalt specimens, inclusive of those enriched with waste butter, was first immersed in liquid nitrogen ($LN_2$, −80 °C). Subsequently, an ultra-fine layer of gold, approximately 10 nm in thickness, was deposited to enhance electrical conductivity. During SEM data acquisition, meticulous parameters were maintained: a magnification set at ×3000, an electron beam current calibrated to 5 nA, a precise working distance of 10 nm, and an accelerating voltage at 5 kV.

### *2.7. Thermogravimetric Analysis (TGA) of Base AP-5 Asphalt with Varied Waste Butter Concentrations: Assessing Thermal Characteristics*

Utilizing a Thermogravimetric Analyzer (TGA Q500, TA Instruments, New Castle, DE, USA), the nuanced thermal characteristics of base AP-5 bitumen were systematically assessed upon the integration of waste butter at various concentrations (e.g., 3, 6, and 9 wt. % WB). During the TGA analysis, approximately 10–15 mg of the binder or waste butter sample underwent a controlled thermal ramp from 25 to 1000 °C at an increment rate of 20 °C $min^{-1}$, all within an inert nitrogen atmosphere (flow rate of 150 mL $min^{-1}$). To ascertain methodological precision and repeatability, this analytical protocol was iteratively performed thrice.

### *2.8. Calorimetric Investigations of Base AP-5 Bitumen Modified with Waste Butter via Differential Scanning Calorimetry (DSC)*

Leveraging the advanced technology of TA Instruments' Differential Scanning Calorimeter (DSC Q20 V24.11 Build 124, New Castle, DE, USA), a comprehensive calorimetric study was conducted to explore the thermal behavior alterations in base AP-5 bitumen when modified with various concentrations of waste butter (WB). Precise quantities of waste butter, at concentrations of 3, 6, and 9 wt. % WB, were integrated into the bitumen to assess their thermal impact.

In preparation for the DSC analysis, a carefully weighed sample of approximately 1–2 mg, comprising the WB and modified asphalt blend, was sealed in an aluminum crucible. This setup was then placed in a nitrogen-rich atmosphere to avoid any oxidative interference. A crucible without any sample was used as a control to ensure accuracy in the measurements.

The experimental procedure commenced with a gradual increase in temperature from ambient to +50 °C at a controlled rate of 10 °C min$^{-1}$. This initial heating phase, spanning 10 min, established a reliable baseline for the thermal behavior of the sample. Following this, the temperature was lowered to −90 °C at the same rate and then raised again to a peak of +150 °C. These carefully controlled heating and cooling cycles were critical for eliminating any previous thermal history in the asphalt specimens.

After the first thermal cycle, the sample underwent rapid cooling from +150 °C to −90 °C, where it was held for 10 min. It was then reheated to +150 °C at 10 °C min$^{-1}$. This second heating phase was crucial for obtaining key calorimetric data, particularly the changes in enthalpy (ΔH) and critical phase transition temperatures, such as the glass transition ($T_g$) and melting temperatures ($T_m$).

To ensure the robustness and reproducibility of the results, the entire DSC experimental protocol was performed in triplicate. This rigorous approach underscores our commitment to delivering empirical insights into how waste butter concentrations influence the thermal properties of AP-5 asphalt cement.

### *2.9. Rheological Characterization of Base AP-5 Asphalt Cement Modified with Waste Butter: Comprehensive Evaluation of the Penetration, Softening Point, Rotational Viscosity, and Ductility*

Utilizing advanced rheological techniques, a suite of ASTM-conforming assessments was meticulously undertaken to elucidate the nuanced effects of waste butter incorporation (at concentrations of 3, 6, and 9 wt. % WB) on the physico-rheological behaviors of the AP-5 asphalt cement.

#### 2.9.1. Penetration

The needle penetration, a pivotal metric elucidating binder hardness or malleability, was meticulously quantified via a Humboldt Mfg Electric Penetrometer (Humboldt Mfg. Co., Elgin, IL, USA), strictly adhering to the ASTM D5 protocol [34]. Ensuring optimal reproducibility, the binder's response to a calibrated 100 g loaded needle, under a tightly regulated environment of 25 °C, was recorded for a 5 s interval, with precision to a tenth of a millimeter. The penetration test was conducted on five separate occasions to guarantee consistent reproducibility, exactitude, and precision.

#### 2.9.2. Softening Point

Transitioning to the softening point—a critical determinant of asphalt's thermal susceptibility—the Ring & Ball Test Apparatus RKA 5 (Anton Paar GmbH, Ashland, VA, USA) was employed, as dictated by ASTM D36 [35]. This phase involved tracking the temperature threshold at which the binder transits to a pre-defined viscosity level, substantiated by the binder's 2.5 cm flow under a standardized 1 cm diameter steel ball. The ring-ball test was executed on four distinct occasions to ensure its reliability, meticulous accuracy, and pinpoint precision.

#### 2.9.3. Rotational Viscosity

Delving into the rotational viscosity (RV)—a quantifier of internal shear resistance—the Brookfield DV III Rheometer (Brookfield, Middleboro, MA, USA) was engaged, strictly adhering to the procedural intricacies of ASTM D4402 [36]. With precision temperature control set at 135 °C, the torque essential for rotating an SC4-27 spindle at a fixed 20 rpm whilst immersed in 10 ± 0.50 g of the binder was methodically computed. The viscosity assessment was performed on four separate instances to ascertain its consistent reproducibility, stringent accuracy, and unwavering precision.

#### 2.9.4. Ductility

Concluding with the ductility assessment—indicative of a binder's cohesive and elongational capacity—parameters from ASTM D113 [37] were stringently observed. Under controlled conditions at 25 °C, a binder briquette was elongated in a calibrated water bath environment, progressing at 5 cm per minute. The ultimate elongation prior to fracture offered invaluable insights into the binder's tensile resilience, rendered in precise centimeters. The ductility examination was undertaken four times to secure reliable reproducibility, unerring accuracy, and meticulous precision.

### *2.10. Nuanced Evaluation of Waste Butter's Influence on Base AP-5 Bitumen's Temperature Susceptibility (TS) Metrics*

In the realm of foundational AP-5 bitumen characterization, paramount metrics such as the Penetration Index (PI) and the Penetration–Viscosity Number (PVN) have been judiciously employed to decipher the nuanced impact of distinct concentrations of waste butter (WB), delineated as 3, 6, and 9 wt. %. Elevated manifestations of PVN and PI inherently suggest a binder's diminished predisposition toward temperature-induced variances. In juxtaposition, lesser metrics delineate a heightened temperature receptivity of said binder [38]. In scenarios characterized by elevated service temperatures, binders characterized by mitigated temperature sensitivities are observed to epitomize superior high-viscosity attributes. Analogously, under the spectrum of reduced service temperatures, these binders manifest commendable flexibility [38]. It warrants emphasis that such methodological approaches, albeit invaluable, are fundamentally broad-stroke representations pertaining to a binder's rheological comportment within tangible hot-mix paving contexts. As such, to cultivate a profound and empirically robust linkage with tangible road paving outcomes, the incorporation and meticulous scrutiny of Dynamic Shear Rheometer (DSR) data for the binders under consideration are indispensable.

The Penetration Index's (PI) derivation is encapsulated by Equation (1) [38]:

$$\mathrm{PI} = \frac{1952 - 500\log \mathrm{P} \ - 20 \times \mathrm{SP}}{50\log \mathrm{P} - \mathrm{SP} - 120} \tag{1}$$

P: penetration, quantified in units of 0.1 mm (dmm) and ascertained under conditions of 25 °C, 100 g, and 5 s.

SP: the softening point, gauged in degrees Celsius (°C).

The Penetration–Viscosity Number (PVN) was subsequently computed via Equation (2) [38]:

$$\mathrm{PVN} = -1.5\left(\frac{4.2580 - 0.7967\log\mathrm{P} - \log\mathrm{V}}{0.7591 - 0.1858\log\mathrm{P}}\right) \tag{2}$$

P: penetration, quantified in units of 0.1 mm (dmm) and determined under stipulated conditions of 25 °C, 100 g, and 5 s.

V: kinematic viscosity, ascertained at 135 °C, and conveyed in units of centistokes (cSt).

### *2.11. Rigorous Examination of Base AP-5 Asphalt Cement's Viscoelastic Response via Dynamic Shear Rheometry: Emphasis on the Rutting Potential*

In this systematic evaluation, a Dynamic Shear Rheometer (DSR)—the Thermo Scientific™ HAAKE™ MARS™ Rheometer, crafted by ThermoFisher, situated in Newington, NH, USA—was the instrument of choice. The principal objective was to delve rigorously into the ramifications of distinct concentrations of waste butter (e.g., 3, 6, and 9 wt. % WB) on the viscoelastic characteristics of the foundational AP-5 asphalt cement.

The DSR examination centered exclusively on rutting potential, especially at elevated temperatures ranging from 46 to 82 °C. This testing approach was meticulously aligned with the procedures delineated in ASTM D7175 [39]. Under unaged conditions, the rheological nuances of both the untreated and the WB-infused bitumen samples were discerned at a loading frequency of 10 rad $s^{-1}$ (1.59 Hz). This frequency was judiciously selected to emulate the shearing dynamics congruent with vehicular movement at approximately 55 mph (90 km $h^{-1}$).

Crucial to ascertaining the longevity and robustness of the roadway infrastructure, parameters such as the rutting factor were derived by leveraging the complex shear modulus |G*|, commonly termed stiffness, along with the phase angle (δ), which encapsulates potential plastic deformation. In the context of elevated temperature assessments (46–82 °C), the bitumen specimens adhered to an exacting thickness of 1 mm coupled with a diameter of 25 mm, dimensions meticulously optimized for analytical precision.

## 3. Results and Discussion

### *3.1. Impact of Waste Butter on the SARA Fraction Distribution in AP-5 Asphalt: A TLC-FID Analysis*

To comprehensively elucidate the intricate mechanisms underlying the influence of waste butter (WB) on the chemical constitution of the base AP-5 asphalt, a methodological application of thin-layer chromatography complemented by flame ionization detection (TLC-FID) was undertaken. This analytical approach was systematically executed on pristine AP-5 asphalt, renowned for its PG 70–10 classification, in conjunction with specimens rejuvenated by varying quantifiable incorporations of WB, spanning concentrations of 3, 6, and 9 wt. %.

The empirical results, elucidated in Figure 3, delineate a quintessential Iatroscan histogram pertinent to the unadulterated AP-5 asphalt. Within this spectrum, four salient bands emerge, meticulously categorized as follows: (1) saturates, representing saturated hydrocarbons, which encapsulate 3.35 ± 0.28 wt. % of the composition; (2) aromatics, encompassing aromatic hydrocarbons, accounting for 59.63 ± 1.88 wt. %; (3) resins, contributing 16.78 ± 2.00 wt. %; and (4) asphaltenes, which are predominant, amassing 20.23 ± 1.79 wt. %. This compartmentalized assemblage is ubiquitously acknowledged within petrochemical domains as the SARA fractions.

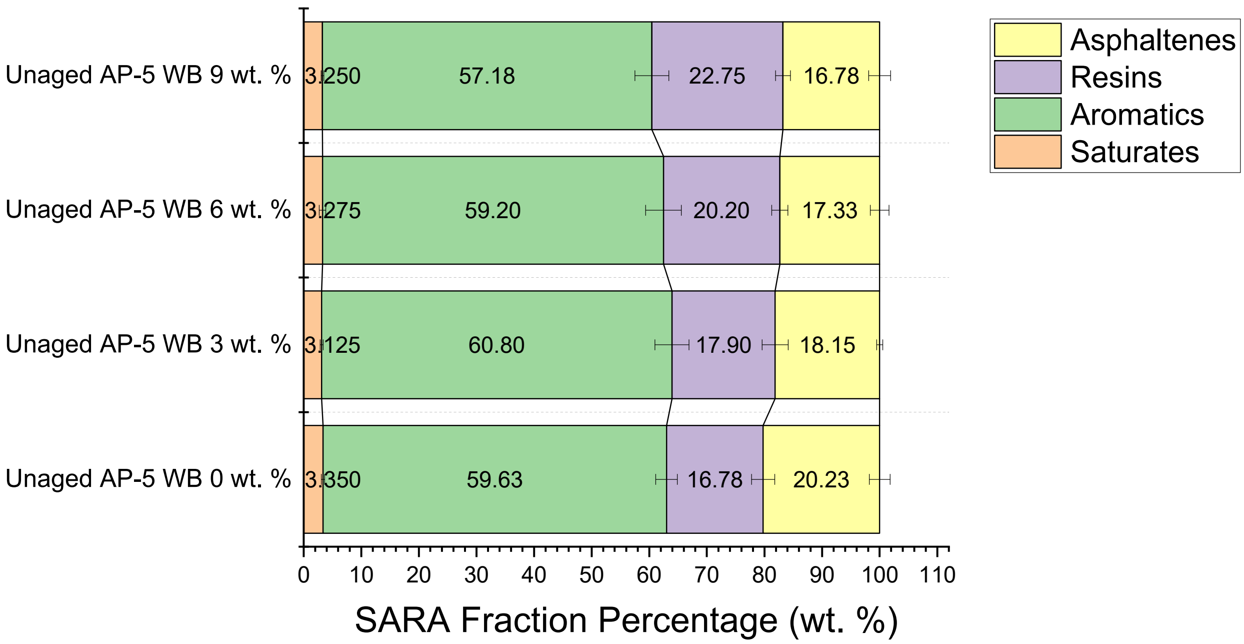


**Figure 3.** Influence of incremental waste butter proportions (e.g., 3, 6, 9 wt. % WB) on the SARA components of pristine base AP-5 bitumen.

Upon rigorous examination of Figure 3, intriguing revelations manifest. Consequent to the progressive infusion of WB—assertively characterized as 100 wt. % resins—into the foundational AP-5 asphalt matrix, there is an unequivocal attenuation in the content of aromatics and asphaltenes. Simultaneously, a proportional augmentation is discerned within the resin segment of the maltene phase. However, the saturate fraction exhibits remarkable stasis, alluding to its inherent resistance to chemical perturbations.

The observed changes in the aromatic fraction with the addition of waste butter (WB) to AP-5 asphalt are influenced by both dilution effects and chemical interactions between WB and the aromatic hydrocarbons. Initially, with the addition of 3 wt. % WB, a slight increase in aromatics is observed. This may be due to the partial solubilization and dispersion of WB components within the asphalt matrix, temporarily increasing the aromatic fraction.

However, with further increases in WB content to 6 wt. % and 9 wt. %, a decrease in the aromatic fraction is evident. This reduction is likely due to more pronounced chemical interactions where the resinous compounds in WB facilitate the conversion or association of aromatic hydrocarbons into more polar fractions, such as resins. This hypothesis is supported by the concurrent increase in the resin fraction observed in the TLC-FID analysis. These interactions suggest that WB's resinous compounds integrate with aromatic hydrocarbons, transforming them into resins and, thus, reducing the aromatic content.

Further detailed studies are required to confirm these interactions and fully understand the underlying mechanisms responsible for these observed changes.

The increase in the resin fraction is primarily due to the composition of the waste butter itself, which is entirely resinous. As WB is incorporated into the asphalt, the inherent resins from the WB directly contribute to the resin fraction of the asphalt. Additionally, the interactions between WB resins and the other components of the asphalt, such as the conversion of aromatics to resins and the solubilization of asphaltenes, further augment the resin fraction. This dual contribution underscores the notable rise in the resin content observed in the TLC-FID analysis.

The decrease in asphaltene content can be attributed to the solubilizing effect of the added WB resins. Asphaltenes, high-molecular-weight and polar molecules, tend to agglomerate and form micelles. The infusion of WB resins may disrupt these micelles, leading to the dispersion of asphaltenes into smaller molecules, which then integrate into the resin fraction. This solubilization effect explains the observed reduction in asphaltenes and the concomitant increase in the resin content.

Nevertheless, further investigations are needed to validate the proposed mechanisms and fully comprehend the long-term effects of incorporating waste butter into asphalt.

### *3.2. Understanding Colloidal Dynamics: The Role of Waste Butter in Base AP-5 Asphalt*

Bitumen's microstructure is intrinsically colloidal, where a delicate balance exists between stabilizing and destabilizing forces. The inclusion of external agents, such as waste butter (WB), can tip this balance, affecting the overall stability and rheological properties of the asphalt. To elucidate these intricate interactions, researchers often turn to the Colloidal Instability Index ($I_C$) or the Gaestel Index [40–42].

This index serves as a barometer for the compatibility dynamics between WB and base AP-5 asphalt. It provides a ratio between the dispersed phase, comprising flocculants and asphaltenes, and the dispersing phase, which includes peptizers (resins in the case of WB) and solvents [43,44]. It is essential to understand that a lower $I_C$ value implies a more stable system where asphaltenes remain well-dispersed. In contrast, higher $I_C$ values warn of potential asphaltene destabilization, leading to problematic aggregations in the system [43,44].

The data displayed in Figure 4 offer intriguing insights. The base AP-5, devoid of any WB, has an $I_C$ of 0.3086, suggesting a fairly stable system. However, as WB is introduced, the $I_C$ values consistently decline, hinting at enhanced stability. For instance, with a 3 wt. % WB inclusion, the $I_C$ drops to 0.2703, and with 9 wt. % WB, it plummets to an impressive 0.2505. This consistent trend underscores the stabilizing influence of WB on the AP-5 asphalt, possibly due to its 100 wt. % resin content.

Colloidal chemistry offers another perspective. In this realm, bitumen is visualized as a sea of micelles. Each micelle features an asphaltenic core shielded by a resinous layer, all floating in a continuous maltenic medium [40]. The equilibrium of SARA components allows us to categorize these binders further. Binders with $I_C$ values below 0.7, referred to as sol bitumina, have a high resin content and minimal asphaltenes. Such binders possess reduced viscosity and elevated fluidity [45]. Interestingly, all AP-5 asphalt samples, whether pure or mixed with varying WB percentages, fall under this category. On the other end of the spectrum, we have gel bitumina, with $I_C$ values above 1.2, characterized by their high viscosity and reduced fluidity [45]. Between these two extremes lie the sol–gel bitumina, with a balanced rheological profile [45].

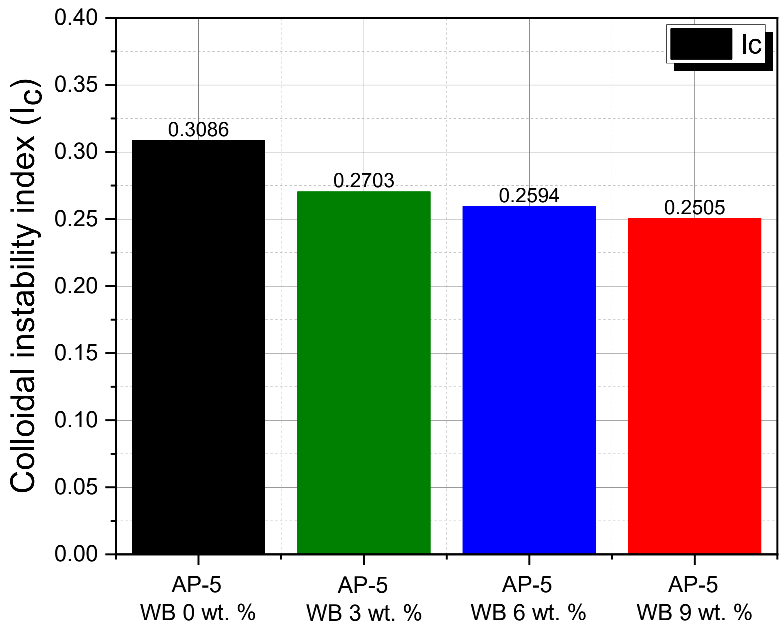


**Figure 4.** Influence of varying concentrations of waste butter (e.g., 3, 6, and 9 wt. % WB) on the colloidal instability index ($I_C$) of unaged base AP-5 asphalt.

In summation, the infusion of WB into AP-5 asphalt significantly enhances its colloidal stability. This positive influence is primarily attributed to WB's rich resin content. However, while these preliminary observations are promising, the true potential and real-world applicability of this modified asphalt warrant further investigation.

### *3.3. In-depth Analysis of Waste Butter's Molecular Impact on Base AP-5 Asphalt via FT-IR Spectroscopy*

FT-IR spectroscopy enabled a rigorous examination of how waste butter (WB) impacts the molecular structures and chemical composition of base AP-5 asphalt. As displayed in Figure 5, the FT-IR spectra elucidate not only the distinctive characteristics of WB but also the properties of unmodified base AP-5 asphalt and those modified with varying proportions of WB. Specifically, these modifications span proportions of 3, 6, and 9 wt. % WB, all observed within a frequency range of 4000–650 $cm^{-1}$.

The Fourier-transform infrared (FT-IR) spectrum of waste butter (WB) unveils a myriad of peaks, emblematic of its primary constituents: triglycerides derived from fatty acids [46,47]. At the higher end of the wavenumber spectrum, a peak at 3842 $cm^{-1}$ suggests the presence of O–H stretching, which could potentially be attributed to bound water or hydroxyl groups, shedding light on butter's minor constituents apart from fats. A corresponding O–H stretching vibration, often linked to hydrogen-bonded hydroxyl groups, emerges prominently at 3472 $cm^{-1}$. This reaffirms that butter, despite its primary fatty makeup, contains other components, possibly derivatives from its parent milk.

Diving deeper into the intricacies of unsaturation within butter's fatty acids, the =C–H stretching vibrations materialize at 3008 $cm^{-1}$, underscoring the presence of unsaturated bonds. This is flanked by the aliphatic chains' characteristic signals, represented by the C–H asymmetric and symmetric stretching at 2919 $cm^{-1}$ and 2851 $cm^{-1}$, respectively.

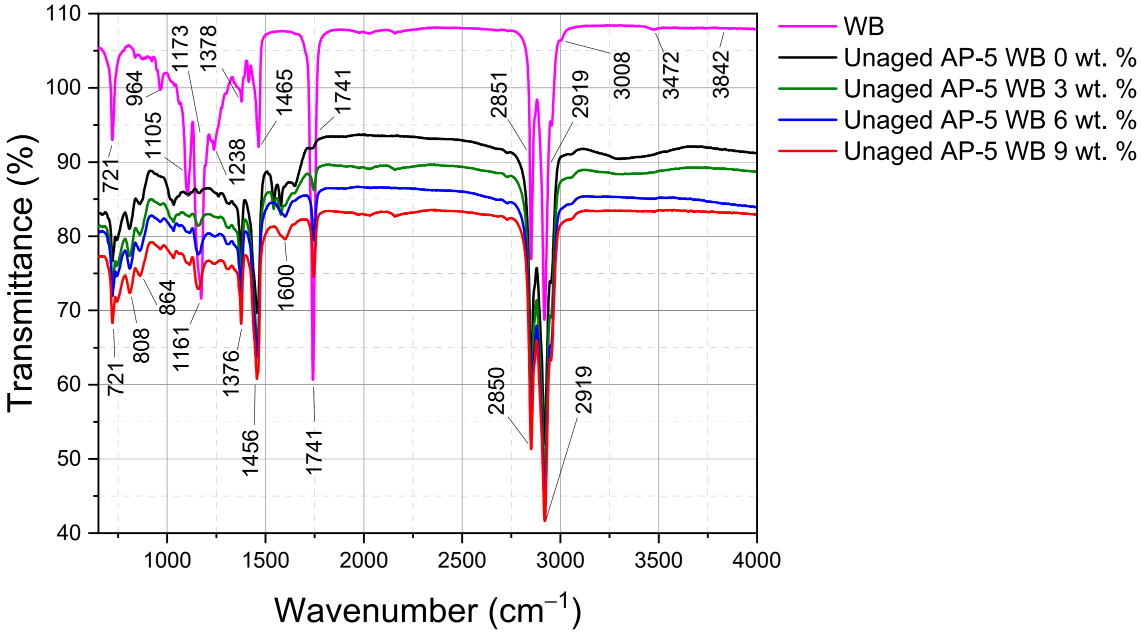


**Figure 5.** FT-IR spectra of waste butter (WB) and base AP-5 bitumen: a comparative analysis across different WB loadings (e.g., 3, 6, and 9 wt. %) in unaged conditions.

One of the most tell-tale signs of butter's molecular structure is the strong C=O stretching vibration at 1741 cm$^{-1}$, eloquently echoing the ester linkages where fatty acids conjugate with glycerol in triglycerides. Further substantiating the rich aliphatic nature of butter, the C–H bending vibration resonates at 1465 cm$^{-1}$.

The spectrum's mid-region provides intriguing insights, with peaks around 1238 cm$^{-1}$ and 1173 cm$^{-1}$, likely indicative of C–O stretching and O–H bending vibrations, further illuminating the ester components and other potential oxygen-bearing constituents. A similar resonance, potentially for C–O stretching, is discernible at 1105 cm$^{-1}$.

As we venture toward the lower wavenumbers, the 964 cm$^{-1}$ peak emerges, shedding light on the *trans* configuration of unsaturated fatty acids via the trans =C–H out-of-plane bending. Lastly, the rhythm of long aliphatic chains in butter is encapsulated by the C–H rocking vibration observed at 721 cm$^{-1}$.

In summation, the FT-IR spectrum of butter offers a comprehensive tableau of its molecular and chemical architecture. However, it is pivotal to note that the spectrum's nuances may vary based on the butter's specific fatty acid composition and any other ancillary components.

In the spectra of base AP-5 asphalt [48,49] treated with WB, a correlation emerges between the WB content and the intensities of associated peaks; a progressive rise in WB content is mirrored by a similar increase in peak intensities. Such observations underscore the infusion of WB's molecular properties into the asphalt matrix. For instance, the spectral region between 2850 and 2919 cm$^{-1}$, known for its association with C–H stretching vibrations in aliphatic chains, exhibits pronounced peaks. Meanwhile, the C–H deformations in $CH_2$ and $CH_3$—both asymmetric and symmetric—are distinctly evident at wavenumbers 1462–1458 cm$^{-1}$ and 1375–1377 cm$^{-1}$, respectively. A particularly salient absorption peak in the vicinity of 1579–1598 cm$^{-1}$ corresponds to C=C stretching vibrations in aromatic rings. While this band is conspicuously absent in the bio-modifier spectrum, a diminishing trend in its intensity is observed upon incremental WB addition to the base AP-5 asphalt, suggesting a concomitant reduction in asphaltene content. Concurrently, the spectral band spanning 1743 to 1746 cm$^{-1}$, attributed to C=O stretching vibrations, gains prominence, hinting at the presence of ketones, anhydrides, and carboxylic acids.

Remarkably, the inherent carbonyl peak in the base AP-5 asphalt spectrum, attributed to mild environmental oxidation, presents a nuanced shoulder. This feature experiences a linear amplification with successive WB incorporation, underscoring a meticulous alteration in the molecular structure. Such a trend underscores a concurrent augmentation in maltenes content, a crucial determinant of binder softness.

To summarize, introducing the bio-based additive, WB precipitates substantial alterations in the chemical structure and composition of the fresh binder.

### *3.4. Micro-Morphological and Elemental Evolution of Base AP-5 Asphalt following Waste Butter (WB) Integration: A Comprehensive SEM-EDX Investigation*

To gain a comprehensive understanding of the micro-morphological, micro-mechanical, and elemental attributes of the base AP-5 asphalt, a methodical examination was undertaken using scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDX). This seminal investigation aimed to elucidate the effects and compatibility of waste butter (WB) when integrated with the base AP-5 asphalt.

A series of illustrative SEM images (Figure 6A–D) are provided to visually represent the nuanced changes in the asphalt's fracture surfaces. In Figure 6A, the base AP-5 asphalt in its pristine form can be observed, showcasing a sleek, undisturbed fracture surface devoid of any layered or gully formations. However, a contrasting narrative emerges when WB is introduced. The subsequent images, particularly Figure 6C,D, highlight hilly contours interspersed with gully-like depressions, a marked deviation from the untreated asphalt's morphology.

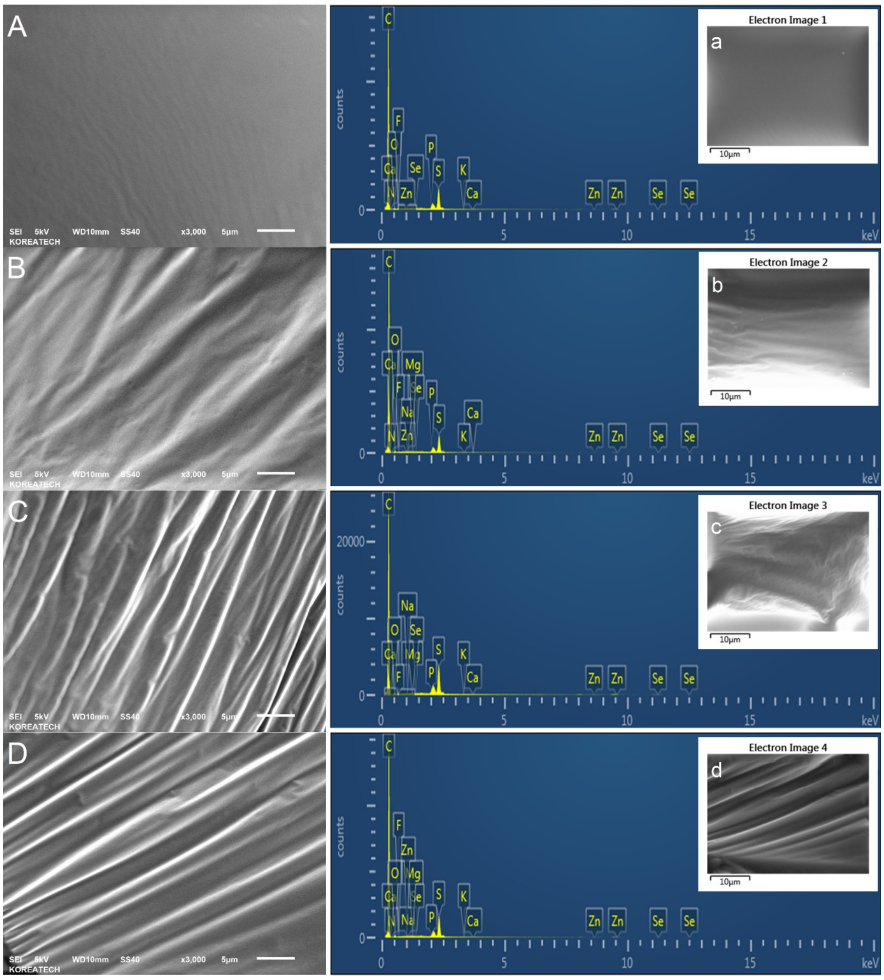


**Figure 6.** SEM photomicrographs and corresponding EDXS spectra at ×3000 magnification: comparing untreated and WB-treated asphalt samples. Designations are as follows: (**A**,**a**) Fresh–pure base AP-5 asphalt; (**B**,**b**) AP-5 integrated with 3 wt. % WB; (**C**,**c**) AP-5 fortified with 6 wt. % WB; and (**D**,**d**) AP-5 enriched with 9 wt. % WB.

Intriguingly, Figure 6 offers no discernible presence of WB or any distinguishable interfaces between the bio-modifier and the asphalt. This omission underscores the remarkable compatibility between the base AP-5 asphalt and WB. Notably, the asphalt sample infused with a 9 wt. % concentration of WB (as depicted in Figure 6D) reveals a textured, multi-layered morphological structure. Yet, even with this pronounced integration of WB, the absence of phase separation becomes evident, alluding to the exceptional miscibility of the bio-modifier with bitumen.

In a rigorous SEM-EDX analysis of the AP-5 asphalt, following its integration with WB, certain elemental shifts become palpable. At a modest incorporation of WB, the carbon (C) constituents exhibited a slight attenuation, whereas the oxygen (O) component revealed a more pronounced elevation. Sulfur (S), in contrast, showed a diminishing trend. As the WB concentration was further intensified, the carbon content continued to decrease, while both oxygen and sulfur values exhibited notable fluctuations. When the integration reached its zenith with the highest observed WB concentration, the carbon content demonstrated a partial rebound, oxygen levels stabilized, and sulfur approached its initial concentration. These data underscore the dynamic interplay of elemental constituents in AP-5 asphalt in response to incremental WB incorporations.

It is also of paramount significance to highlight that the mineral elements characteristically inherent in butter [50]—fluorine (F), sodium (Na), magnesium (Mg), phosphorus (P), potassium (K), calcium (Ca), zinc (Zn), and selenium (Se)—remained conspicuously absent or fell below the detectability threshold in the asphalt–butter composite using the SEM-EDX technique. Their minuscule concentrations insinuate the indispensability of deploying more sophisticated analytical modalities for their unequivocal detection.

Probing the elemental perturbations more profoundly, the attenuation in the carbon content can be ascribed to the intrinsic chemical constitution of the WB. It is postulated that the fatty acids and glycerides within WB potentially engage in chemical interactions with the carbonaceous entities within the asphalt [51]. The augmented oxygen content delineates potential esterification or oxidation processes, thus engendering a more oxygen-prevalent milieu. The nuanced trajectory of sulfur—an initial decrement followed by an increment—illuminates the multifaceted chemical interplays at hand, suggesting the presence of sulfur-rich moieties in the WB exhibiting differential reactivity contingent upon their concentration.

### *3.5. The Thermal Characterization and Resilience of Waste-Butter-Amalgamated Bituminous Blends via Thermogravimetric Analysis (TGA)*

Thermogravimetric analysis (TGA) is a quintessential thermal analytical method extensively utilized to discern the temperature-dependent behavior of bio-modifier/bitumen composites. At its core, TGA quantitatively measures the weight diminution of a binder substance, which might originate from rudimentary procedures like desiccation or intricate chemical transformations that yield gaseous products and induce structural disintegration. Within this study, a sophisticated thermal gravimetric analyzer was deployed to systematically monitor the weight variations in bituminous samples due to temperature modulations.

Figure 7 offers a comprehensive and quantitative examination of the TGA trajectories corresponding to WB, the pristine base AP-5 asphalt, and an assortment of bituminous compounds integrated with differentiated WB concentrations (e.g., 3, 6, and 9 wt. %).

For the unadulterated base AP-5 asphalt (i.e., Unaged AP-5 WB 0 wt. %) under an atmospheric nitrogen ($N_2$) milieu, the TGA analysis was delineated across three distinct phases [52,53]. The initial weight-diminution phase (phase 1), extending from temperatures of 35.69 °C to 391.50 °C, encapsulates the expulsion of primary volatile constituents, inclusive of saturates and aromatics. These emanate predominantly from the disjunction of fragile molecular bonds facilitated by molecular chain fragmentations and cycloreversion reactions. Progressing to the subsequent phase (phase 2), circumscribed by temperatures ranging from 391.50 °C to 470.03 °C, there is a substantial 76.76 wt. % weight abatement due to the exhalation of secondary volatiles and the concomitant combustion of resins.

Herein, molecular detachments of hydrogen and methyl transpire, leading to the synthesis of asphaltenes and char, which predominantly stem from the free radical molecule condensation processes. The final phase (phase 3), manifested on the TGA curve, represents a weight decrement of approximately 23.24 wt. % within the temperature spectrum of 470.03–999.95 °C, marking the degradation of asphaltenes and subsequent char formation (5.06 wt. %).

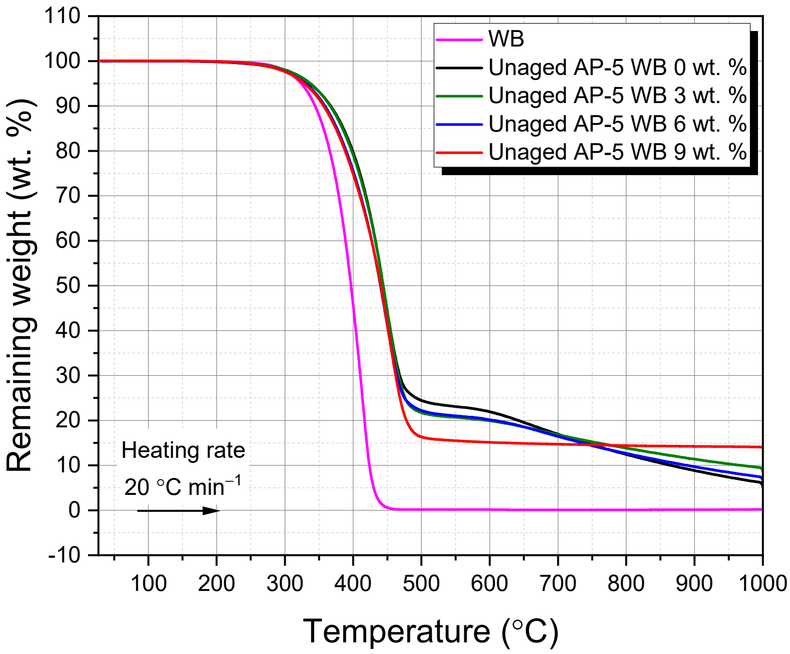


**Figure 7.** Thermogravimetric (TGA) curves: comparing waste butter (WB), unadulterated base AP-5 asphalt, and AP-5 variants fortified with specified WB concentrations (e.g., 3, 6, and 9 wt. %).

Furthermore, the WB sample's thermal stability was rigorously assessed under a nitrogen atmosphere via TGA. As expounded in Figure 7, a pronounced plateau emerges, serving as a testament to the robust thermal endurance of this lipidic component, persisting up to an approximate 366.84 °C. The decomposition terminus is pinpointed at roughly 429.13 °C. This thermal degradation manifests in two pivotal stages, ostensibly demarcating the decomposition of unsaturated fatty acids (encompassing the 26.68–366.84 °C range) and their saturated counterparts (within the 366.84–429.13 °C domain) [54].

Table 3 provides a synthesized overview of the empirical data gleaned from the TGA/DTGA plots and elucidated in Figures 7 and 8. A discerning analysis reveals that the thermal robustness of the asphalt amalgamations witnessed a nuanced alteration after incorporating 6 and 9 wt. % WB. Contrarily, an infusion of 3 wt. % of butter subtly augmented its stability. This nuanced thermal modulation is feasibly attributed to the pronounced concentration of unsaturated fatty acids [33,55], imbuing the asphalt binder with a modicum of susceptibility to incipient thermal degradation.

Conclusively, upon a holistic appraisal, it is cogently posited that the bio-modifier in question—waste butter—imparts no adverse perturbations upon the thermal characteristics of bituminous blends. Its competence in revitalizing the seasoned binder in base AP-5 asphalt, particularly within standardized operational temperature thresholds (ensuring the $T_{max}$ of HMA amalgamation and production remains below 200 °C), is underscored.

**Table 3.** Comparative thermogram metrics from TGA/DTGA: evaluating waste butter (WB), pristine base AP-5 asphalt, and WB–AP-5 formulations at loadings of 3, 6, and 9 wt. % WB at a steady heating rate of 20 °C min$^{-1}$.

| Specimen | TGA/DTGA (°C) | | | | | | −ΔW (wt. %) |
|---|---|---|---|---|---|---|---|
| | Phase 1 | Phase 2 | Phase 3 | $T_{onset}$ | $T_{offset}$ | $T_{max}$ | |
| WB | 26.68–366.84 | 366.84–429.13 | 429.13–998.86 | 366.84 | 429.13 | 413.64 | 0.23 |
| AP-5 WB 0 wt. % | 35.69–391.50 | 391.50–470.03 | 470.03–999.95 | 391.50 | 470.03 | 447.03 | 5.06 |
| AP-5 WB 3 wt. % | 31.17–392.07 | 392.07–472.91 | 472.91–999.96 | 392.07 | 472.91 | 449.68 | 8.62 |
| AP-5 WB 6 wt. % | 32.04–384.23 | 384.23–469.61 | 469.61–999.97 | 384.23 | 469.61 | 446.93 | 6.39 |
| AP-5 WB 9 wt. % | 25.95–388.19 | 388.19–477.01 | 477.01–999.95 | 388.19 | 477.01 | 453.25 | 13.84 |

$T_{onset}$: Initiation of thermal degradation (°C); $T_{offset}$: Conclusive temperature for thermal loss (°C); $T_{max}$: Peak decomposition temperature (°C); ΔW: Carbonaceous residual content at 1000 °C (wt. %).

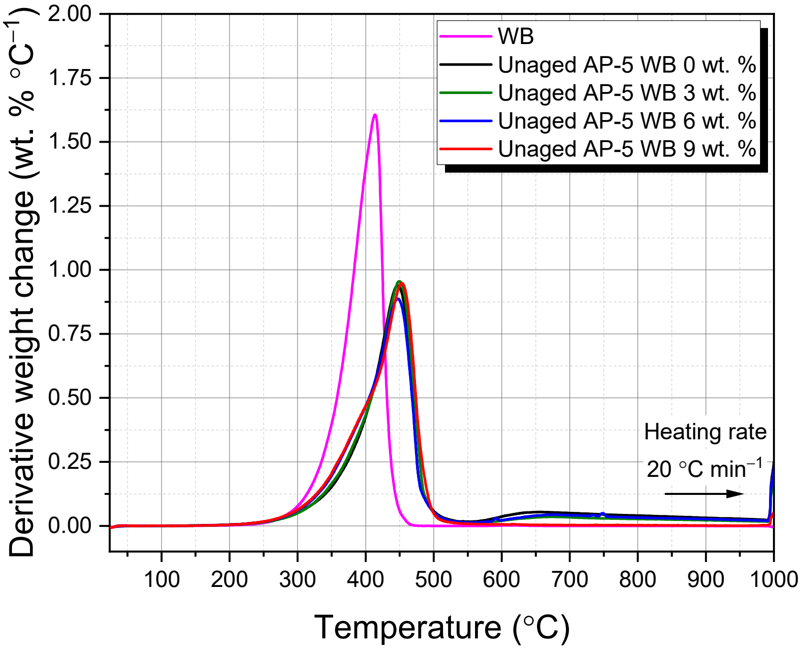


**Figure 8.** Differential thermogravimetric (DTGA) trajectories: evaluating waste butter (WB), unmodified base AP-5 Asphalt, and variants augmented with selected WB concentrations (e.g., 3, 6, and 9 wt. %).

### *3.6. Catalyzing Sustainable Infrastructure: A Comprehensive Analysis of Waste Butter's Influence on AP-5 Asphalt Cement Thermal Properties Determined via Differential Scanning Calorimetry (DSC)*

In our quest for more sustainable construction materials, a detailed investigation was conducted to assess the impact of waste butter's (WB) integration on the thermal characteristics of AP-5 asphalt cement. This was achieved through the application of Differential Scanning Calorimetry (DSC), a critical technique in delineating the effects of varied WB proportions (e.g., 3, 6, and 9 wt. %) on the thermal transitions and phase behaviors within the asphalt matrix.

The DSC thermograms, illustrated in Figure 9, juxtapose the thermal profiles of pristine AP-5 asphalt with those modified by WB incorporation. A thorough analysis of these thermograms indicates that unmodified AP-5 asphalt exhibits dual glass transition temperatures ($T_g$), specifically at $T_{g1}$ = −15.89 °C and $T_{g2}$ = +12.08 °C.

Regarding the DSC analysis of waste butter (WB), as shown in Figure 9, its thermogram reveals a melting range from 0 °C to 40 °C (ΔH = 69.07 J g$^{-1}$). This range is predominantly due to the variable saturation levels of triacylglycerols (TAGs) in the butter fats, with higher-saturation TAGs melting at elevated temperatures and vice versa [56–58].

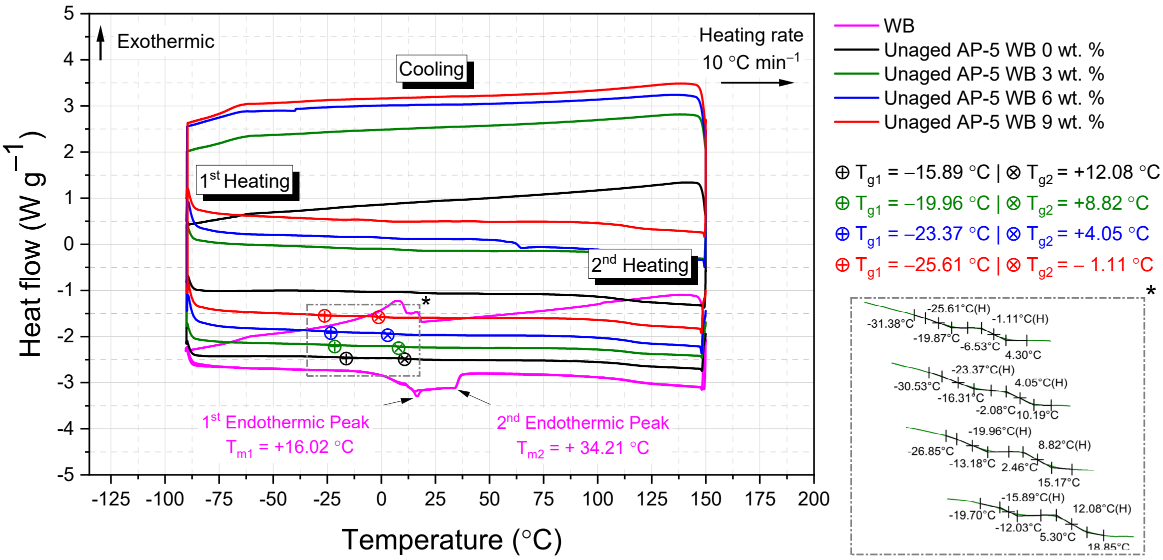


**Figure 9.** Differential Scanning Calorimetry (DSC) trajectories: assessing the thermal behavior of waste butter (WB), unmodified base AP-5 asphalt, and variants augmented with selected WB concentrations (e.g., 3, 6, and 9 wt. %).

The WB thermogram exhibits two overlapping endothermic peaks corresponding to the melting behaviors of different fatty acid (FA) types. The first, more pronounced peak around $T_{m1}$ = +16.02 °C is primarily due to the melting of unsaturated FAs and TAGs [56–58]. This is influenced by compositional changes such as the length of the fatty acid chains and the degree of unsaturation, which are critical factors in the phase transitions of fats and oils [59]. Following this is a secondary peak around $T_{m2}$ = +34.21 °C, reflective of saturated FA and TAG melting [56–58]. The nature of the distribution of fatty acids in triacylglycerol (TAG) species also plays a role in these phase transitions, further illustrating the complex lipid architecture within WB [56–58]. The dominance of the first peak over the second in the thermogram implies a greater concentration of unsaturated over saturated fatty acids in WB [56–58], a crucial factor in its solid-to-liquid transition and its interaction with the asphalt matrix across varied temperatures. The two-phase melting process of WB is vital in determining its performance in different industrial applications [56–58].

It is noteworthy that the glass transition temperatures of neat AP-5 asphalt may deviate from literature values due to factors like the source, production process, and composition [60–63]. The geological origin and the specific diagenetic processes, along with refining methodologies (e.g., distillation, fractionation, storage), significantly influence the asphalt's Tg values [60–63].

In the AP-5 asphalt matrix, phase transitions are attributed to various amorphous structures. For example, linear alkanes exhibit a $T_{g\,Sat}$ of between −88 °C and −60 °C. The maltene phase (i.e., saturates, aromatics, resins) has a $T_{g\,Mal}$ of around −20 °C. The interfacial region between maltenes and asphaltenes shows a $T_{g\,Mal\text{–}As}$ near −10 °C, and the asphaltenes themselves exhibit a $T_{g\,As}$ of around +70 °C [64–67].

The strategic integration of waste butter (WB) into AP-5 asphalt cement yields a nuanced alteration in its thermal characteristics, as evidenced by Differential Scanning Calorimetry (DSC) analysis. This incorporation notably reduces the glass transition temperature ($T_g$) of the asphalt matrix, a phenomenon that is reflective of the molecular interplay between the maltene phase of the asphalt and the resinous fatty constituents of WB. This molecular synergy results in a refined thermal profile for the WB-enriched asphalt, distinct from its unmodified counterpart.

Such modifications in thermal behavior are not just confined to shifts in $T_g$. They extend to a broader spectrum of the asphalt's performance attributes. The lowered $T_g$, as a consequence of WB integration, augments the asphalt's performance in colder environments, enhancing its resistance to low-temperature-induced cracking [68,69]. This is a pivotal improvement, particularly in regions subjected to severe winter conditions.

### *3.7. Physical and Rheological Transformation of Base AP-5 Asphalt following Waste Butter Incorporation: A Multi-Faceted Conventional Binder Test Analysis*

The results derived from the conventional binder tests, namely, penetration, softening point, viscosity, and ductility, were employed to assess the influence of varying dosages of waste butter (WB)—specifically, 3, 6, and 9 wt. %—on the physical and rheological properties of the base AP-5 asphalt. The data from these tests are depicted in Figure 10, Figure 11, Figure 12, and Figure 13 regarding the penetration, softening point, viscosity, and ductility values, respectively.

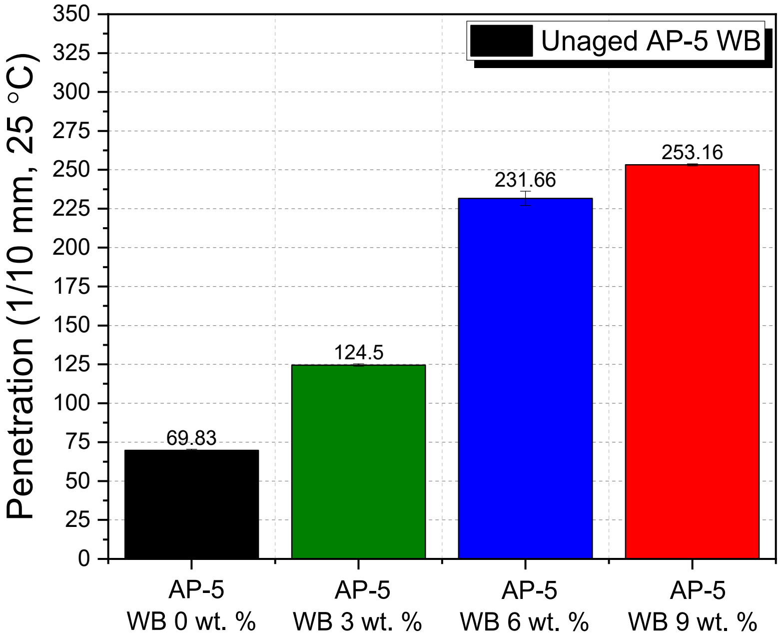


**Figure 10.** Influence of diverse waste butter fractions (e.g., 3, 6, and 9 wt. % WB) on the penetration characteristics of unaged base AP-5 bitumen.

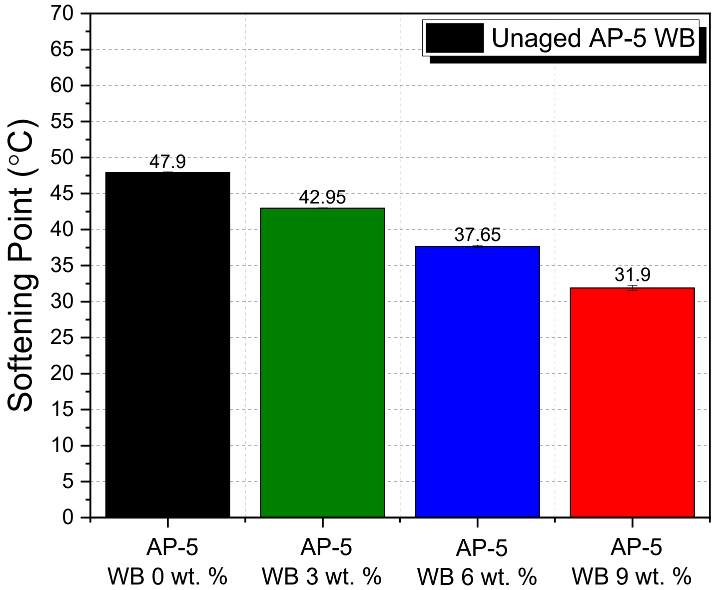


**Figure 11.** Impact of different waste butter proportions (e.g., 3, 6, and 9 wt. % WB) on the softening point of unaged base AP-5 bitumen.

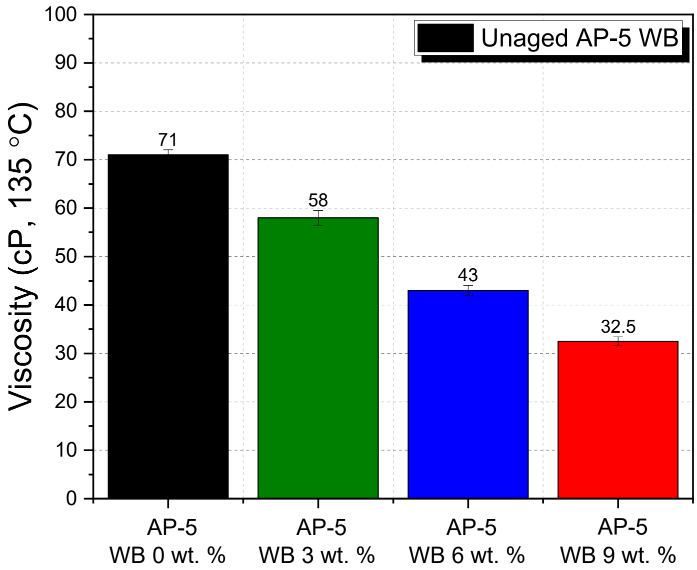


**Figure 12.** Influence of varied waste butter concentrations (e.g., 3, 6, and 9 wt. % WB) on the viscosity of unaged base AP-5 bitumen.

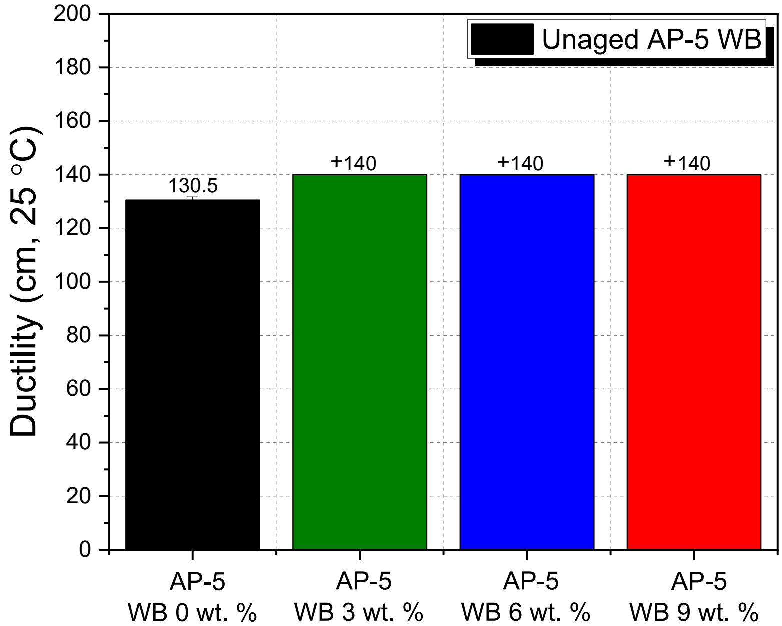


**Figure 13.** Impact of distinct waste butter proportions (e.g., 3, 6, and 9 wt. % WB) on the ductility of unaged base AP-5 bitumen.

### 3.7.1. Penetration Analysis: Waste Butter's Influence on Base AP-5 Asphalt Malleability and Performance

In the sophisticated domain of asphalt rheology, the base AP-5 asphalt, categorized under the Superpave performance-graded binder (PG 70–10), stands out as a foundational reference material. Conforming to the ASTM D946 standards [70], the intrinsic rheological behavior of unmodified AP-5 asphalt was comprehensively charted.

Figure 10 showcases the transformative influence of waste butter (WB) on the rheological attributes of AP-5 asphalt. As the data show, even a marginal inclusion of WB creates a pronounced shift in the asphalt's penetration metrics. This implies a palpable decrease in its inherent hardness, which can be primarily attributed to the rich biochemical profile of waste butter.

#### 3.7.2. Softening Point Analysis: Unraveling the Thermo-Mechanical Impact of Waste Butter on Base AP-5 Asphalt

Figure 11 offers an intricate insight into the transformational influence of varied waste butter (WB) concentrations on the softening point of pristine base AP-5 asphalt. This metric, intrinsically linked to the binder's thermo-mechanical endurance, is paramount in road construction considerations.

With each progressive increment in WB, there is an observed systematic decline in the softening point of the asphalt binder. The baseline, represented by the unadulterated AP-5 asphalt, is a commendable 47.90 °C. However, as WB concentrations rise to 3, 6, and 9 wt.%, there are pronounced reductions to 42.95 °C, 37.65 °C, and a significant 31.90 °C, respectively. This pattern not only underscores a palpable decrement in the material's thermal rigidity but also resonates profoundly with findings from previous penetration tests, as elucidated in Figure 10.

Piercing through the superficial implications, a lowered softening point potentially translates to the binder becoming less resistant to deformations at elevated temperatures. In lay terms, in the scorching summer heat, the road might become more susceptible to rutting and other high-temperature deformities, especially in areas grappling with intense vehicular traffic. On the contrary side, an intriguing advantage emerges. A diminished softening point could confer upon the asphalt enhanced flexibility during colder periods, making it a robust candidate for regions plagued by harsh winters [71]. Here, the modified binder might effectively stave off fatigue cracking, prevalent distress induced by thermal fluctuations in colder regions [71].

#### 3.7.3. Viscosity Analysis: Deciphering the Impact of Waste Butter on Base AP-5 Asphalt's Rheological Behavior and Molecular Interactions at Compaction Temperatures

Figure 12 showcases an intricate analysis of viscosity—a cardinal metric elucidating a binder's inherent resistance to deformation under externally applied shear forces. Conducted at a judiciously chosen temperature of 135 °C, emblematic of the compaction stage for asphalt binders, the findings illuminate a progressive viscosity diminution, correlating with escalating concentrations of waste butter (WB). An analytical dissection of the data reveals a compelling decrement in viscosity, plummeting from an outset value of 71.00 cP (with 0 wt. % WB) to a mere 32.50 cP at the highest tested WB concentration of 9 wt. %.

At the molecular echelon, the fatty acid moieties resident within WB instigate perturbations in the intricate web of intermolecular cohesion characteristics of AP-5 asphalt [72,73]. This molecular interplay grows increasingly conspicuous with escalating WB incorporation, accentuating the substantive viscosity reductions, particularly palpable at 6 and 9 wt. % WB [72,73].

#### 3.7.4. Waste Butter Augmentation in Base AP-5 Asphalt: A Deeper Dive into Ductility Dynamics

Figure 13 distinctly showcases the measurable influence of waste butter (WB) on the ductility characteristics of base AP-5 asphalt. In the unaltered control sample, ductility stands at 130.50 cm. However, introducing WB at 3 wt. % creates a significant surge in this measure, crossing the 140.00 cm threshold. Notably, this enhanced ductility remains consistent, even when the WB content is raised to 9 wt. %.

From a mechanics standpoint, bettered ductility can bolster defense against fatigue cracking. The fatty acids in WB could act as reactive hubs or shields, slowing the binder's oxidation, a prime precursor to fatigue crack inception [74,75]. This delay in the oxidation process might preserve the binder's viscoelastic traits over time, reducing early micro-cracking and their eventual evolution into larger cracks [74,75].

### *3.8. Deciphering the Rheological Transformations: Integrating PI and PVN Metrics to Assess Temperature Susceptibility (TS) in Asphalt Binders Enriched with Waste Butter Derivatives*

In the realm of pavement engineering, bitumen's thermoplastic behavior plays a crucial role, particularly its propensity to undergo rheological transformations in response to thermal gradients. The quantification of temperature susceptibility (TS) stands as a linchpin in asphalt pavement design. Essentially, TS offers an incisive metric, delineating the rate at which a binder's consistency—whether represented through penetration or through viscosity metrics—responds to variances in temperature. For a holistic and precise elucidation of TS, especially in unaged asphalt specimens fortified with distinct proportions of waste butter (e.g., 3, 6, and 9 wt. % WB), the amalgamation of metrics such as the Penetration Index (PI) and the Penetration–Viscosity Number (PVN) is imperative.

#### 3.8.1. Rheological Intricacies of Base AP-5 Asphalt: Dissecting the Thermo-Mechanical Impact of Waste Butter Resinous Additives on Penetration Index (PI) Dynamics

Data extrapolated from Figure 14 illuminate the PI value spectrum, spanning from −3.54 to −0.41. In the ambit of advanced road infrastructural engineering, optimal PI metrics are predominantly nestled between the −1 and +1 benchmarks [38]. A deviation beyond these boundaries, particularly submerging beneath −2, raises concerns regarding enhanced thermal susceptibility [38]. Such a proclivity suggests that the bituminous construct might be at the mercy of brittle fractures, especially when assailed by capricious climatic variables [38]. Conversely, asphalt specimens with an index surpassing +1 manifest attributes of brittleness, often symptomatic of attenuated thermal adaptability juxtaposed with amplified elastic characteristics [38].

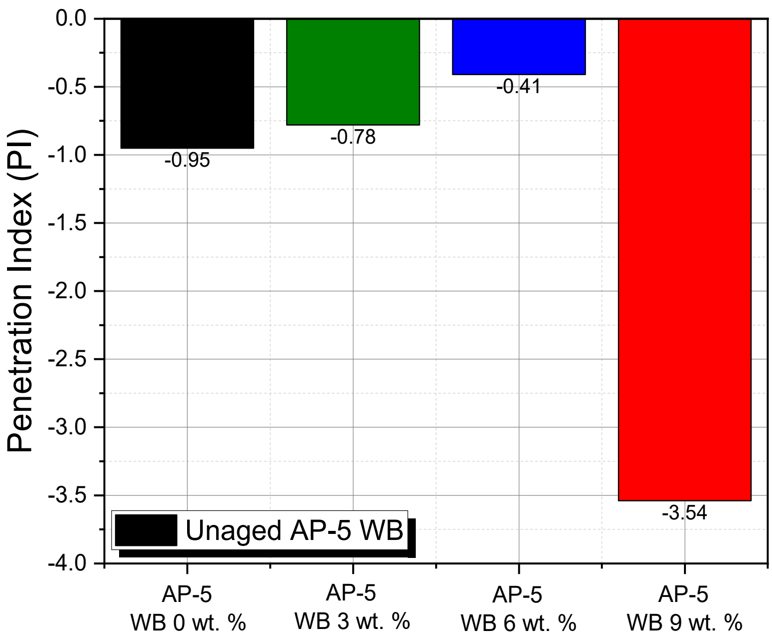


**Figure 14.** The influence of graduated waste butter concentrations (e.g., 3, 6, and 9 wt. % WB) on the Penetration Index (PI) of pristine base AP-5 asphalt under unaged conditions.

Our investigation orbits around the strategic integration of waste butter, a derivative teeming with free fatty acids, which intriguingly account for staggering 100 wt. % resins. These resins, by virtue of their inherent chemical and physical properties, are poised to modify the binder's thermo-rheological response. Initial observations from the unamalgamated AP-5 asphalt (i.e., AP-5 WB 0 wt. %) evince a PI of −0.95. A systematic augmentation of waste butter concentrations yields intriguing modulations: −0.78 at 3 wt. % and −0.41 at 6 wt. %, culminating in a pronounced decline to −3.54 at 9 wt. %. This precipitous decrement at elevated concentrations suggests a potential inflection point, wherein the resinous attributes of the free fatty acids might engender counterintuitive rheological ramifications, leading to pronounced brittleness.

3.8.2. Thermal Susceptibility of Base AP-5 Asphalt: Evaluating the Rheological Impact of Waste Butter Derivatives on Penetration–Viscosity Numbers (PVNs)

Within the scientific ambit of Figure 15, a rigorous quantitative articulation of Penetration–Viscosity Numbers (PVNs) is presented, contrasting the physicochemical properties of unmodified AP-5 asphalt with its counterparts, systematically enriched with waste butter (WB) derivatives, under strictly unaged conditions. The documented PVN parameters are delineated within a range of −3.96 to −3.36. According to established rheological benchmarks and corroborated by comprehensive literature [38], paving-grade asphalts typically exhibit PVN values spanning from −2.0 to +0.5. An elevated PVN index generally indicates an asphalt binder's diminished susceptibility to thermal variations.

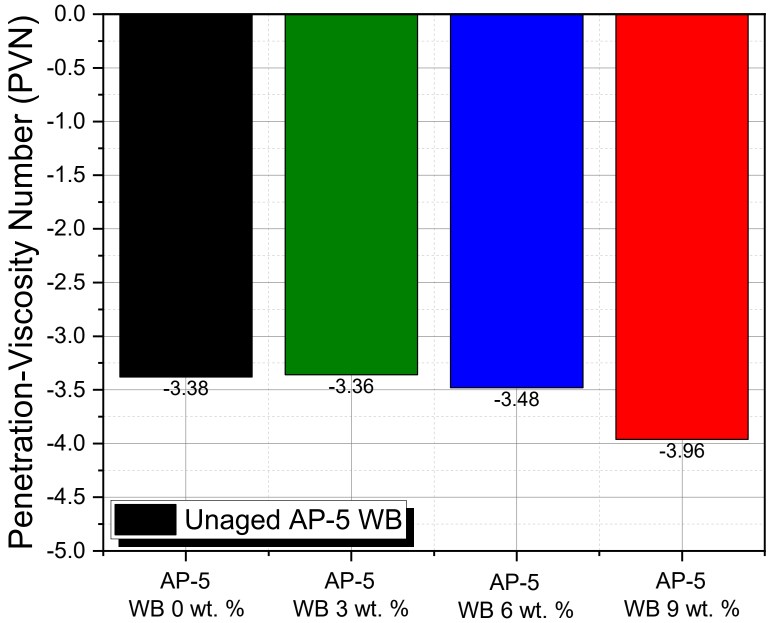


**Figure 15.** The influence of graduated waste butter concentrations (e.g., 3, 6, and 9 wt. % WB) on the Penetration–Viscosity Number (PVN) of pristine base AP-5 asphalt under unaged conditions.

Upon in-depth evaluation, the pristine AP-5 asphalt formulation registered a PVN metric of −3.38. This specific datum not only underscores the binder's inherent resilience to temperature fluctuations but also exemplifies its potential to mitigate the phenomena of transverse cracking, even under notably reduced thermal conditions.

Meticulous scrutiny of Figure 15 elucidates that the incorporation of waste butter, specifically enriched in 100 wt. % free fatty acid resin constituents, engenders notable alterations in the bituminous matrix's thermal susceptibility characteristics. These resinous constituents, characterized by their unique physicochemical signature, interact synergistically with the bituminous matrix, effecting nuanced modulations in its rheological and thermal response. Consequently, while this resin-rich integration fortifies the binder's resistance to low-temperature deformations, it intimates potential challenges pertaining to its rutting resistance under high-thermal-load scenarios.

In summation, the methodical integration of waste butter's resinous constituents into the asphaltic formulation offers promising rheological advantages, particularly under sub-zero conditions. However, it mandates a comprehensive and judicious evaluation, particularly under high-temperature scenarios, to ensure optimal pavement performance.

*3.9. DSR Analysis of Waste Butter Concentrations on Rutting Resilience in Unaged Base AP-5 Asphalt: Insights into the Modulus Behavior and Superpave Compliance*

The intricate evaluation via the Dynamic Shear Rheometer (DSR) was strategically employed to deconstruct the intricate nuances of varying waste butter concentrations (e.g., 3, 6, and 9 wt. % WB) on the rutting behavior of base AP-5 asphalt, specifically under unaged conditions.

At the crux of the DSR's analytical methodology lies the rutting resistance metric, denoted as G*/sin δ. Within this framework, |G*| epitomizes the complex shear modulus, underscoring the binder's intrinsic resilience against shear deformations, while "δ" represents the phase angle, a pivotal indicator of the viscoelastic balance within the asphaltic binder. Elevated G*/sin δ values are synonymous with a binder that leans toward elasticity, a trait crucial for resistance against continuous traffic-induced shear stresses. Contextualizing within the practical realm of asphalt application, fresh asphalt, when subjected to minimal or no aging during the laying process, should exhibit a stiffness quotient G*/sin δ that exceeds 1.0 kPa at a uniform operational temperature [76]. This threshold not only mitigates potential vulnerabilities during crucial phases such as mixing and compaction but also heralds longevity and structural coherence of the paved infrastructure [76].

Delving into the nuanced outcomes furnished by DSR, as elucidated in Figure 16, differential behavior between the unmodified AP-5 asphalt (i.e., Unaged AP-5 WB 0 wt. %) and its counterparts enriched with waste butter is manifested. The pristine AP-5, under unaged conditions, emerged as the benchmark, showcasing minimal propensity toward rut-induced deformations. Tracing the progression of waste butter integration reveals a multifaceted interplay: as waste butter concentrations escalate, coupled with a systematic modulation in operational temperatures, the stiffness parameter G*/sin δ exhibits a decelerating trajectory. This trend can be epistemologically tethered to the physicochemical behavior of waste butter within the bituminous matrix. Waste butter, rich in fatty acid derivatives, potentially softens the binder matrix, enhancing penetration depth–ductility metrics and concomitant reductions in key attributes like the softening point and viscosity. Notwithstanding this softening effect, it is worth noting that at sub-critical temperatures (below 52 °C), all binder formulations under scrutiny met the rigorous standards (i.e., G*/sin δ $\geq$ 1 kPa), as stipulated by the Superior Performing Asphalt Paving (Superpave) criteria [76]. Specifically, across temperature spectra of 70, 64, 58, and 52 °C, the foundational AP-5 bitumen, juxtaposed with its variants integrated with incremental waste butter concentrations (e.g., 3, 6, and 9 wt. % WB), adhered to these stipulations with impeccable fidelity.

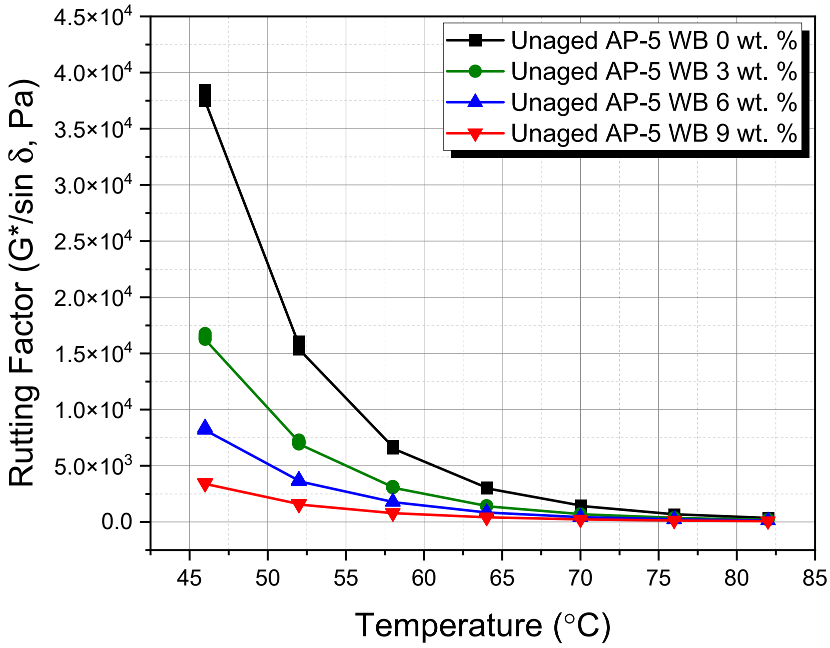


**Figure 16.** The rutting factor (G*/sin δ) for unaged base AP-5 asphalt across varied waste butter concentrations (e.g., 3, 6, and 9 wt. % WB) vs. temperature.

### *3.10. Enhancing Asphalt Performance via Waste Butter Integration: Insights and Future Directions*

The exploration of waste butter as a bio-modifier for AP-5 asphalt cement reveals promising paths to enhance both the performance and sustainability of asphalt blends. By repurposing an organic waste product, this initiative not only advances waste reduction but also augments the mechanical properties of traditional asphalt.

#### 3.10.1. Experimental Methodology and Study Limitations

The methodology applied in this study emphasizes detailed assessments of the chemical, physical, and rheological alterations in asphalt due to waste butter modification. Notable improvements, especially in low-temperature performance, were observed, though this study was conducted within the confines of a controlled laboratory environment. Thus, translating these results to practical, real-world scenarios remains uncertain. Future research should extend these findings across different asphalt grades and environmental conditions to validate the broader application potential of waste butter in asphalt modification.

#### 3.10.2. Integration within the Existing Body of Knowledge

This research aligns with existing studies on using bio-modifiers in asphalt, enhancing the current understanding by investigating higher concentrations of modifiers than commonly studied. The observed improvements in thermal and mechanical properties underscore the viability of organic waste materials in advancing asphalt technology, particularly for enhancing thermal crack resistance and flexibility at lower temperatures.

#### 3.10.3. Conclusive Insights for Practical Implementation

Harnessing the comprehensive data from Dynamic Shear Rheometer (DSR) assessments, it is evident that incorporating specific concentrations of waste butter (e.g., 3%, 6%, and 9% by weight of binder) into standard AP-5 asphalt necessitates precise calibration for real-world application. The sustainability offered by integrating waste butter needs to be balanced against its impact on rutting susceptibility, necessitating a tailored approach to pavement design and construction, especially in thermally volatile environments. Continuous monitoring during pavement installation is crucial to ensure the long-term durability and consistent performance of the infrastructure. Integrating waste butter into the asphalt mix highlights a convergence of sustainability and performance optimization, requiring an informed, flexible, and adaptive implementation strategy.

#### 3.10.4. Future Research Trajectories

The encouraging outcomes from including waste butter in the asphalt binder call for further investigation into the long-term durability and performance of these modified blends under actual pavement conditions. Assessing the environmental impacts throughout the lifecycle of bio-modified asphalt is essential to fully ascertain its sustainability. Future studies could also explore the economic ramifications of scaling up waste butter use within the asphalt industry to ensure environmental and economic feasibility.

The expanded insights from this study promote ongoing research and development of sustainable materials in civil engineering, advocating for the broader adoption of innovative practices that enhance the construction of more robust and sustainable infrastructure.

## 4. Conclusions

Pursuing sustainable innovations in civil infrastructure, our research elucidates the promising potential of using waste butter (WB)—a predominantly overlooked animal fat byproduct—in reshaping AP-5 asphalt formulations due to its bio-enhancing properties. This pioneering approach not only underscores the environmentally conscious utilization of animal-derived residues but also signifies a critical nexus between ecological responsibility and infrastructural progression.

In examining the intricate rheology of asphalt, we meticulously assessed the ramifications of incorporating diverse waste butter (WB) concentrations, delineated as 3, 6, and

9 wt. %. Using advanced analytical techniques, like TLC-FID, FT-IR, SEM, TGA, and DSC, the chemical aspects of asphalt were thoroughly investigated. Concurrently, vital physical properties, such as the penetration, softening point, viscosity, and ductility, were closely scrutinized. Our deployment of the DSR (Dynamic Shear Rheometer) was instrumental in gauging the rutting factor, granting a comprehensive insight into the interplay between thermal susceptibility, as defined by the Penetration Index (PI) and Penetration–Viscosity Number (PVN), and the bituminous composition fortified with WB derivatives.

The key findings of our exploration encompass the following:

- Following WB addition, the FT-IR spectrum exhibited no emergent spectral peaks, indicating predominantly physical rather than chemical interplay.
- TLC-FID analyses portrayed a reduction in aromatic and asphaltene contents offset by an increased resin presence, emphasizing WB's central role in balancing asphalt's inherent chemical milieu.
- The colloidal instability index ($I_C$) emphasized enhanced stability, a testament to WB's pronounced 100 wt. % resin composition.
- SEM evaluations unveiled nuanced micromorphological shifts with WB inclusions, affirming the harmonious amalgamation within the binder structure.
- Significantly, TGA findings spotlighted that a modest 3 wt. % WB introduction considerably elevates the asphalt binder's thermal endurance.
- DSC analysis highlighted a notable improvement in the asphalt's performance at lower temperatures upon integrating WB, signifying a leap in the material's cold weather adaptability.
- From a rheological standpoint, WB's infusion led to heightened penetration and ductility, albeit with reduced viscosity and softening point metrics, underscoring its multifarious enhancement attributes.
- Thermal susceptibility (TS) evaluations revealed that integrating resin-rich WB derivatives enhances the thermo-rheological characteristics of AP-5 asphalt, reinforcing its resilience in colder climates and accentuating the necessity for meticulous evaluations under elevated thermal conditions.
- DSR assessments concluded that the judicious introduction of WB into fresh AP-5 asphalt inherently adjusts its viscoelastic properties. Still, accurate calibrations are imperative for achieving peak rutting resistance, consistent with Superpave criteria.
- A pivotal observation propounds that a 3 wt. % WB concentration might optimize the asphalt performance. However, this premise mandates further empirical validation.

To fully fathom and authenticate WB's rejuvenating impact on asphalt, forthcoming studies should focus on aging evaluations using methods such as an RTFO (rolling thin-film oven) and PAV (pressure-aging vessel). Probing into enduring performance indicators, cost implications, and extended environmental repercussions remains paramount to steer this narrative conclusively.

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