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Article

Nano-modified Bitumen Enhancing Properties with Nano-materials

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Abstract: In the modern era, pollution from plastic waste has become a growing concern, particularly due to the widespread use of plastics like plastic bottles. This research explores a novel approach for recycling plastic waste by incorporating plastic bottles into bitumen for the enhancement of Hot Mix Asphalt (HMA). Alongside this, nano-materials such as carbon nanotubes (CNTs), graphene, nanosilica, and nanoclays were used to further improve the mechanical, rheological, and durability properties of the modified bitumen. The plastic waste, in the form of plastic bottles, was added in varying proportions (3%, 5%, 8%, 10%, and 12% by weight of total mix) to investigate its effect on bitumen's performance. The study conducted a series of tests, including Dynamic Shear Rheometer (DSR), Rotational Viscosity, Penetration Test, Softening Point Test, and Scanning Electron Microscopy (SEM), to evaluate the rheological and mechanical properties. The results revealed that the incorporation of plastic waste significantly improved the bitumen's resistance to rutting, cracking, and fatigue, while nano-additives further enhanced high-temperature stability and elastic recovery. As the percentage of plastic waste in the bitumen increased, improvements in resistance to aging and moisture susceptibility were observed. Additionally, the plastic-modified bitumen exhibited better stability, improved resilience to temperature fluctuations, and enhanced mechanical strength. These findings suggest that combining plastic waste and nano-materials in bitumen can contribute to more sustainable road infrastructure, reducing plastic pollution while improving the performance and longevity of asphalt pavements.

Keywords: Plastic Waste Recycling; Bitumen Modification; Nano-materials; Hot Mix Asphalt; Mechanical and Rheological Properties

1. Introduction

Plastic waste, particularly from plastic bottles, has become one of the most prominent environmental challenges of the modern era [1]. With the rapid increase in plastic production and consumption, plastic bottles contribute significantly to the global plastic waste problem [2]. Although plastic bottles are widely used in various industries, they are not biodegradable, leading to their accumulation in landfills and natural environments [3]. It is estimated that millions of tons of plastic waste, including plastic bottles, enter the marine and terrestrial ecosystems each year, causing severe pollution and harm to wildlife. Despite efforts to recycle plastic bottles, the current recycling methods remain inefficient and environmentally taxing, with many of these bottles ending up in waste streams [4].

As global demand for road infrastructure continues to rise, engineers are increasingly exploring innovative ways to address both the issue of plastic waste and the need for more durable and sustainable construction materials [5]. In road construction, bitumen, a key component of Hot Mix Asphalt (HMA), has traditionally been used for its binding properties. However, conventional bitumen

Figure 1. Bitumen.

Figure 2. Plastic Bottles.

faces challenges in terms of durability, particularly against rutting, cracking, and aging. The incorporation of additives, including polymers and recycled materials, is becoming an attractive solution to improve the mechanical and rheological properties of bitumen [6]. One promising approach is the use of plastic bottles as a sustainable additive in bitumen. This research investigates the use of plastic waste in the form of plastic bottles to modify bitumen, with the goal of enhancing its performance in road paving applications. Additionally, the potential for combining plastic waste with advanced nano-materials, such as carbon nanotubes (CNTs), graphene, and nanosilica, is explored to further improve the properties of the modified bitumen [7].

The motivation behind this study is twofold: addressing the growing issue of plastic waste pollution and improving the performance of road construction materials [4]. The large-scale disposal of plastic bottles poses a significant environmental challenge, and finding sustainable ways to recycle these materials is critical [5]. By incorporating plastic bottles into bitumen, this research seeks to reduce plastic waste while simultaneously enhancing the durability and mechanical properties of asphalt pavements. Furthermore, the use of nano-materials offers an opportunity to further boost the high-temperature stability, fatigue resistance, and aging resistance of the modified bitumen. This study aims to explore the synergistic effect of combining plastic waste with nano-materials to create a more sustainable, durable, and eco-friendly road paving solution. By doing so, it will contribute to reducing plastic pollution while improving the overall performance and lifespan of asphalt roads.

The primary objective of this research is to investigate the effect of plastic bottle waste as an additive in bitumen, focusing on its impact on the mechanical, rheological, and durability properties of bitumen. Specifically, the study aims to modify bitumen with plastic bottles to improve its resistance to rutting, cracking, and fatigue, while evaluating the performance improvements of the modified bitumen in terms of high-temperature stability, moisture susceptibility, and aging resistance. Additionally, the research explores the use of nano-materials, such as carbon nanotubes (CNTs), graphene, and nanosilica, in conjunction with plastic waste to enhance the overall performance and longevity of the modified bitumen.

This study focuses on the feasibility of incorporating plastic bottle waste into bitumen to improve its mechanical and rheological properties, while also addressing the growing issue of plastic waste pollution. It explores the use of nano-materials such as carbon nanotubes (CNTs), graphene, and nanosilica to enhance the properties of plastic-modified bitumen. The research further aims to provide a sustainable method for recycling plastic waste in road construction, contributing to more eco-friendly and long-lasting road infrastructure. By integrating plastic bottle waste with advanced nanotechnology, this study proposes a novel approach to asphalt modification that not only helps manage plastic waste but also enhances the performance and longevity of asphalt pavements.

2. Literature Review

The use of waste plastic materials as modifiers for bitumen has gained increasing attention in recent years, particularly in the context of road construction [8], [9]. Various types of waste plastics, including Polyethylene Terephthalate (PET), High-Density Polyethylene (HDPE), and Low-Density Polyethylene (LDPE), have been investigated for their potential to enhance the physical and rheological properties of bitumen binders [10], [11], [12]. The effects of waste plastics, specifically HDPE and LDPE, on bitumen's resistance to aging, rutting, and fatigue [13]. Their study showed that an optimal waste plastic content of 2–4% (HDPE and LDPE) improved the performance of bitumen, with the best results seen at 4% content. However, higher plastic content (6–8%) showed diminishing returns and even increased vulnerability to permanent deformation. On the other hand, PET was found to exhibit significant improvements in the physical characteristics and aging resistance of bitumen when incorporated at higher levels, particularly around 8% [13], [14].

In another study, the modification of bitumen using waste PET, highlighting the improvements in rutting resistance and aging properties through Dynamic Shear Rheometer (DSR) and Rolling Thin Film Oven Tests (RTFOT) [15], [16]. The results suggested that 6–8% PET waste provided the optimal balance between improved rutting resistance and aging durability. Similarly, waste plastic materials like LDPE and HDPE improved the stability and durability of asphalt mixtures [16]. Their research showed an increase in aggregate properties such as impact and abrasion values, while the addition of plastic resulted in improved softening points, penetration, and ductility, especially at 8% LDPE content. Other studies, such as those focused on evaluating different types of waste plastics (PET, PP, PVC, LDPE, HDPE) and their effects on the stability and performance of bituminous concrete (BC). They found that adding waste plastics to the bitumen mix resulted in a significant improvement in stability and durability [17], [18]. The optimum plastic content was identified as 30.1%, higher than the traditional bitumen content, and showed superior performance in both wet and dry conditions.

The use of polypropylene (PP) as an antistripping agent in Hot Mix Asphalt (HMA) has also been explored, with demonstrating that PP enhances the resistance to both wet and dry conditions in asphalt mixtures [19], [20]. PP was found to reduce thermal sensitivity and improve resistance to permanent deformation at high temperatures [21]. These studies underline the potential of using waste plastics as modifiers to improve the mechanical properties of bitumen, making it a cost-effective and environmentally friendly solution for road construction. The incorporation of waste plastic, especially in the form of PET and LDPE, has shown to enhance stiffness, resilience, and fatigue resistance of bituminous mixtures, leading to longer-lasting road surfaces [13], [22]. This literature highlights the promising role of plastic waste in reducing environmental pollution and enhancing the sustainability of road infrastructure.

2.1 Bitumen

Bitumen, a thick, black, and viscous liquid or solid, is produced primarily through the distillation of crude oil (Figure 1). It is widely used in road construction, where it serves as a binder in asphalt mixtures [9]. Bitumen is known for its waterproofing and adhesive properties, which make it an essential component in road paving [18], [23]. The physical and mechanical properties of bitumen can be influenced by several factors, including temperature, aging, and the presence of additives [24], [25]. However, traditional bitumen is prone to performance issues such as rutting, cracking, and aging under various environmental conditions and heavy traffic loads. These performance limitations have led to the development of modified bitumen, where polymers, rubber, and other materials are added to enhance its properties. The use of polymer-modified bitumen (PMB) has shown significant improvements in terms of high-temperature performance, elasticity, and fatigue resistance. Bitumen modification is an ongoing area of research, with the aim of producing longer-lasting, more durable road pavements [25], [26]. Recent studies have increasingly focused on incorporating sustainable materials, such as waste plastics and nano-materials, to further improve the performance of bitumen and make the road construction process more environmentally friendly.

2.2 Plastic Bottles

Plastic bottles, primarily made from Polyethylene Terephthalate (PET), are a major contributor to global plastic waste (Figure 2). With the growing environmental concerns over plastic pollution, there has been a strong push to find innovative ways to recycle and reuse plastic waste [27], [28]. In road construction, plastic bottles have recently been explored as an additive in bitumen to modify its properties. The primary challenge in using plastic bottles as an additive lies in their compatibility with bitumen, as plastics are hydrophobic and non-polar, which can result in poor dispersion and weak bonding with bitumen [29], [30]. However, recent research has shown that with the right processing techniques, such as high-shear mixing or using compatibilizers, plastic waste can be successfully incorporated into bitumen to enhance its mechanical and rheological properties. Studies have shown that the incorporation of PET from plastic bottles can improve the high-temperature stability, stiffness, and resistance to cracking the modified bitumen [13], [14]. Furthermore, plastic bottles used in bitumen modification can help reduce the environmental impact of plastic waste, as this approach offers a sustainable solution for managing plastic pollution. By integrating plastic bottle waste into asphalt mixtures, not only can the performance of the pavements be improved, but the plastic waste can also be diverted from landfills, contributing to a circular economy.

2.3 Challenges and Gaps in Research

Despite the promising results in the use of waste plastic in bitumen modification, there remain several challenges and gaps in research that need to be addressed. One of the key limitations is the compatibility of plastic waste with bitumen. Plastics, being non-polar materials, do not naturally bond well with the highly polar nature of bitumen, which can lead to poor dispersion and inadequate mechanical properties. Therefore, additional efforts are required to improve dispersion techniques and enhance the compatibility between plastic waste and bitumen. Various methods, including the use of compatibilizers or the incorporation of nano-materials, have been suggested to overcome these challenges. Nano-materials such as carbon nanotubes (CNTs), graphene, and nanoclays have been explored in several studies to improve the dispersion and bonding of plastic waste in bitumen, potentially improving its mechanical and rheological properties [4], [31], [32], [33].

Another challenge is the cost of using waste plastic and nano-materials in bitumen modification. Although plastic waste offers an environmentally friendly alternative to traditional materials, the processes involved in recycling and incorporating plastic waste into bitumen can be cost-prohibitive, particularly if high amounts of nanomaterials are used. Moreover, the long-term durability and performance under real-world conditions of bitumen modified with plastic waste remain areas for further research [12], [34]. More comprehensive studies are needed to assess how such modifications perform under different traffic loads, environmental conditions, and over extended periods of use. Furthermore, while various types of waste plastics have been tested for bitumen modification, there is still limited research on the optimal combination of plastic types and nano-materials that would maximize the benefits for road paving [25], [26]. The positive effects of plastic waste on bitumen's performance, but the mechanisms by which these materials interact with bitumen at the molecular level are not fully understood [35], [36]. Future research should focus on the microstructural interactions between plastic waste and bitumen, especially when nanomaterials are introduced, to better optimize the mix and ensure the highest performance.

3. Methodology

The study investigates the performance of bitumen modified with plastic waste derived from plastic bottles and enhanced with nano-materials, including carbon nanotubes (CNTs), graphene, nanosilica, and nanoclays [37], [38]. The primary objective of this study is to evaluate the physical, rheological, and durability properties of nano-modified bitumen as a sustainable solution for road construction materials. The overall experimental procedure for preparing and testing the plastic waste-modified bitumen is summarized in Figure 3.

3.1 Preparation of Plastic Waste Modified Bitumen

Waste plastic bottles, primarily composed of Polyethylene Terephthalate (PET), were shredded into strips (~2–5 mm size) and melted at 158°C for 10 minutes to ensure homogeneity and semi-liquid state for proper integration with bitumen. Plastic waste was added to bitumen in varying proportions of 5%, 8%, 10%, and 12% by weight of the total mix. Each blend was prepared in triplicate (n=3) to ensure repeatability and reliability of test results.

3.2 Preparation of Bitumen

The bitumen used in this study was collected from local suppliers and heated in an oven at 145°C for 10 minutes to ensure a uniform molten consistency suitable for testing and mixing. The bitumen was characterized by using a series of standard tests to establish its baseline properties before modification.

3.3 Nano-Materials Integration

The study incorporated four types of nano-materials to enhance bitumen performance:

a) **Carbon Nanotubes (CNTs)**: Multi-walled CNTs with average diameter of 10–20 nm and length 1–10 µm, purity >95%, were used to improve viscosity, fatigue resistance, and high-temperature stability.
b) **Graphene**: Few-layer graphene sheets (~1–5 µm lateral size, thickness ~1–3 nm) were added to enhance mechanical strength and durability.
c) **Nanosilica**: Amorphous silica nanoparticles (~15 nm diameter, surface area ~150 m^2/g) to improve aging resistance, stiffness, and thermal stability.
d) **Nanoclays**: Montmorillonite nanoclay platelets (~1 nm thickness, ~100 nm lateral size) to promote dispersion and improve rheological behavior.

Nano-materials were dispersed at 1%, 3%, and 5% by weight of total bitumen using a high-speed shear mixer (3000 rpm for 20 minutes) to ensure uniform distribution and avoid agglomeration.

3.4 Aggregate Preparation and Gradation

The aggregates used in the experiment were oven-dried at 105°C to remove any moisture content. The aggregates were then sieved to prepare the desired gradation for the bituminous mix. The gradation adhered strictly to ASTM specifications for asphalt paving mixtures to ensure the consistency of the mix (Table 1). The mass retained on each sieve was recorded, and the cumulative percent passing was calculated as per the standard procedure (Table 1). The aggregates used in the experiment were oven dry at 105°C to remove moisture. According to Table 1, the aggregate gradation strictly follows the ASTM composition of the bitumen paving mixture.

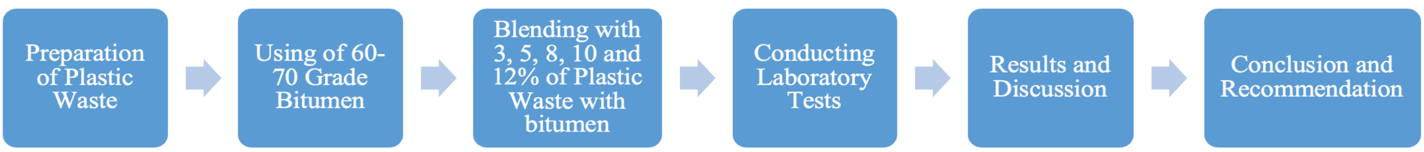

Figure 3. Experimental Workflow for Preparation and Evaluation of Plastic Waste-Modified Bitumen.

Table 1. Aggregate gradation.

Sieve size (mm)	Mass retained, A (gm)	% Retained, B	Cumulative % retained	Cumulative percent passing	ASTM composition of bitumen paving mixture
19 mm	63	4.90	4.90	95.10	90 to 100
9.5 mm	239	19.60	24.50	75.50	56 to 80
4.75 mm	295	24.10	48.60	51.40	35 to 65
2.36 mm	240	19.61	68.21	31.79	23 to 49
0.30mm	231	18.87	87.08	12.92	5 to 19
0.075 mm	132	10.78	97.87	2.13	2 to 8
Pan	24				
Total=	**1224.00**				

Table 2. Marshall method of mix design of bituminous mix without adding Plastic Waste.

Sl. No.	% of bitumen content	Unit weight (lb/ft^3)	VMA (%)	Va (%)	VFA (%)	Corrected marshall stability (kN)	Flow (1/100) in
1	5.0	145.335	17.57	7.01	67.11	11.89	8.67
2	5.5	147.678	18.93	6.33	73.27	12.36	9.34
3	6.0	147.789	19.34	6.01	82.35	12.57	10.77
4	6.5	146.213	18.97	5.97	79.67	12.43	11.62
5	7.0	145.523	18.29	6.65	76.55	11.97	12.01

3.5 Laboratory Tests

A series of laboratory tests were conducted to evaluate the physical, rheological, and mechanical properties of the modified bitumen and asphalt mixtures. The tests followed standard ASTM specifications and were aimed at assessing the performance of the modified bitumen in terms of its suitability for use in road construction.

a) **Specific Gravity Test (ASTM D70)**: Used to measure the mass of a given volume of bitumen at 25°C relative to an equivalent amount of water. This test helps determine the bitumen's absorption capacity and its compatibility with aggregates.
b) **Solubility Test (ASTM D2042)**: Conducted to determine the purity of the bitumen and estimate the amount of bitumen that dissolves in an organic solvent.
c) **Loss on Heating Test (ASTM D6)**: Measures the mass loss of bitumen when heated, indicating the stability of the bitumen during high-temperature mixing.
d) **Penetration Test (ASTM D5)**: Assesses the hardness or softness of bitumen by measuring the depth to which a standard needle penetrates the bitumen sample at 25°C.
e) **Ductility Test (ASTM D113)**: Evaluates the ability of bitumen to stretch without breaking, which is critical for the material's flexibility and resistance to cracking.
f) **Softening Point Test (ASTM D36)**: Determines the temperature at which bitumen softens, indicating its resistance to high-temperature deformation.
g) **Flash and Fire Point Test (ASTM D92)**: Measures the temperature at which the bitumen may catch fire, ensuring safety during its handling and processing.
h) **Marshall Stability Test (ASTM D6927-06)**: Measures the stability and flow of compacted asphalt mixtures, helping to determine the optimal binder content for the mixture.

3.6 Performance Evaluation of the Modified Bitumen

Tests to evaluate rheological and mechanical improvements included:

a) **Dynamic Shear Rheometer (DSR)**: Used to assess the rheological properties of the bitumen, including its resistance to deformation under stress at different temperatures. This test helps evaluate the high-temperature performance of the modified bitumen.
b) **Rotational Viscosity Test**: Measured the viscosity of the modified bitumen to assess its workability during the mixing process at high temperatures.
c) **Fatigue and Rutting Resistance**: These tests were conducted using the DSR and other relevant methods to assess the bitumen's ability to withstand repetitive traffic loads and high temperatures without permanent deformation or cracking.

3.7 Data Analysis

The results from all laboratory tests were analyzed using statistical methods to compare the performance of the plastic-modified and nano-modified bitumen with standard bitumen. Performance indicators such as resistance to rutting, cracking, fatigue, and aging were used to evaluate the improvements brought about by the integration of plastic waste and nano-materials into bitumen.

4. Results and Discussion

Table 2 presents the results of the Marshall Method of Mix Design for bituminous mixtures without the addition of plastic waste, focusing on the effect of different bitumen contents on the performance of the mixture. As the percentage of bitumen increases from 5.0% to 7.0%, several key properties of the mixture show noticeable changes. The unit weight of the mixture increases, indicating a denser composition, while the Voids in Asphalt (Va) decrease, suggesting better compaction. Additionally, the Voids Filled with Asphalt (VFA) and the Voids in Mineral Aggregate (VMA) both rises, demonstrating that more of the aggregate's voids are filled with bitumen, which improves the mixture's stability and durability. The corrected Marshall Stability, which measures the resistance to permanent deformation, increases with higher bitumen content, indicating improved resistance to rutting. On the other hand, the flow value, which measures the mixture's flexibility, also rises with increasing bitumen content, indicating enhanced elasticity. Overall, these trends suggest that the mixture becomes more compact, stable, and flexible as the bitumen content increases, with an optimal bitumen content of around 6.0%, providing a good balance of stability and flexibility.

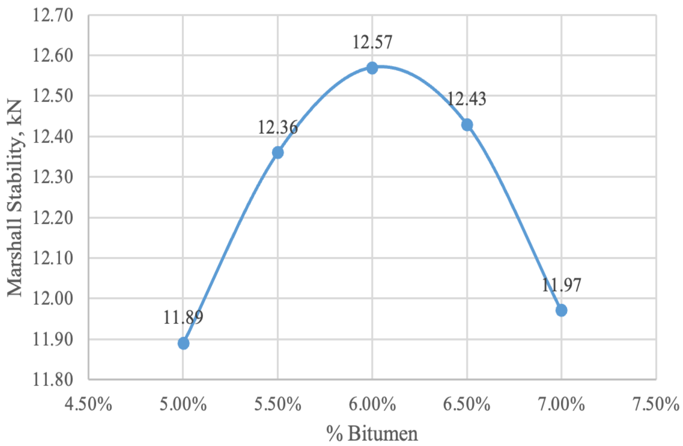

Figure 4. Marshall stability of bitumen.

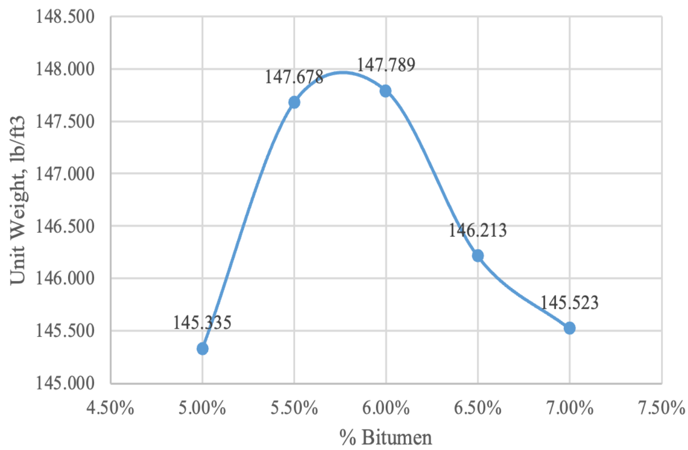

Figure 5. Unit weight of bitumen.

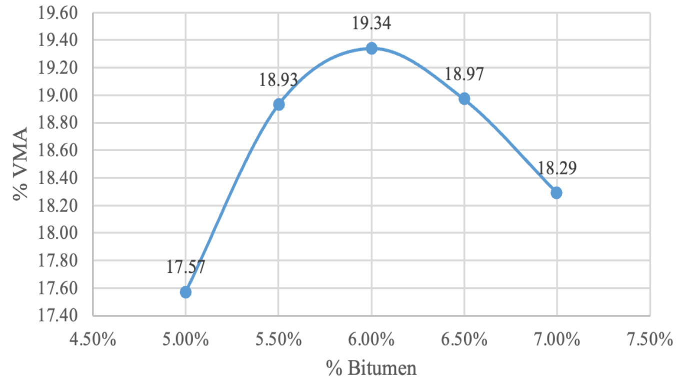

Figure 6. Percent of VMA on bitumen.

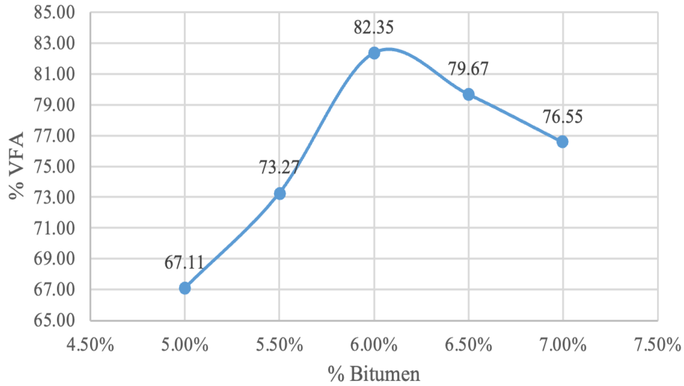

Figure 7. Percent of VFA on bitumen.

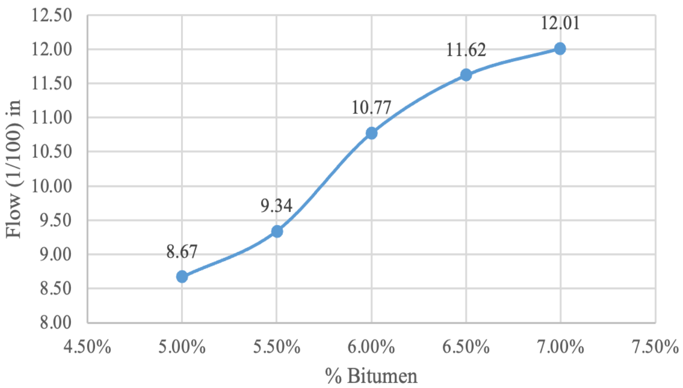

Figure 8. Flow (1/100) in bitumen.

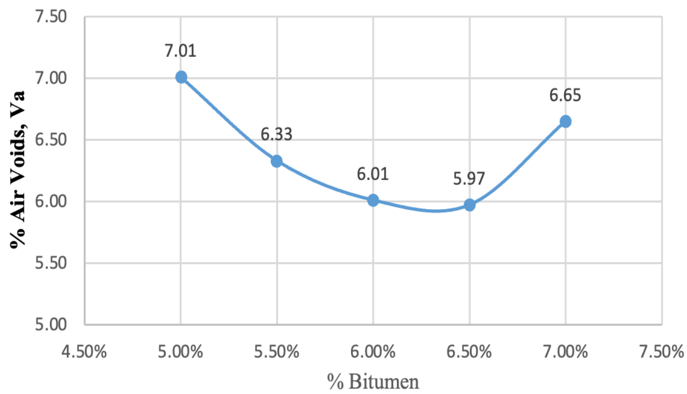

Figure 9. % Air voids, V_a.

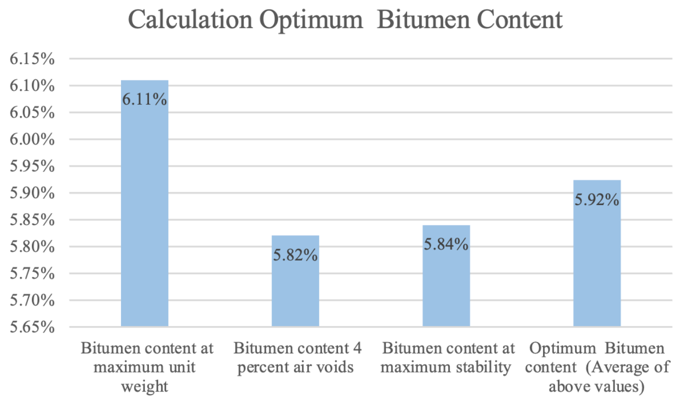

Figure 10. Calculation of optimum Bitumen content.

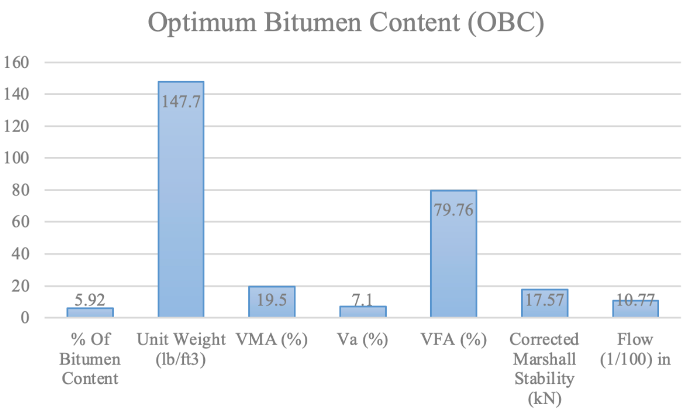

Figure 11. Optimum Bitumen content.

4.1. Marshall mix design of bituminous mix without adding plastic waste

The provided figures illustrate the relationship between various performance parameters of bituminous mixtures and different bitumen content levels, based on the Marshall Method of Mix Design. Figure 4 shows that as the percentage of bitumen increases, the Marshall Stability rises until reaching an optimal point at around 6.0%, after which it starts to decline, indicating improved resistance to deformation at this bitumen content. Figure 5 shows the Unit Weight of the mixture, which increases with the bitumen content, indicating a denser mix as more bitumen fills the voids in the aggregate. Figure 6 depicts the Percent of VMA (Voids in Mineral Aggregate), which also peaks at around 6.0% bitumen content, showing a well-filled mixture at this point. Figure 7 presents the Percent of VFA (Voids Filled with Asphalt), which rises with increased bitumen content, signifying that a higher proportion of the aggregate's voids are filled with binder, improving mixture stability. Figure 8 displays the Flow (deformation under load), which increases as bitumen content rises, suggesting that the mixture becomes more flexible, with a peak observed at 6.0%. Finally, Figure 9 shows the Air Voids (Va), which decrease as the bitumen content increases, indicating better compaction and reduced air pockets in the mixture, with the optimal value occurring around 6.0%. Collectively, these graphs indicate that bitumen content plays a critical role in optimizing the stability, flexibility, and durability of the asphalt mixture, with a 6.0% bitumen content generally offering the best balance of these properties.

Figure 10 shows the calculation of the optimum bitumen content using different criteria. The bitumen content at maximum unit weight, 4 percent air voids, and maximum stability are plotted, along with their average value. The optimum bitumen content is identified as approximately 5.92%, which balances density, air voids, and stability to achieve the best overall mixture performance.

Figure 11 presents key properties of the optimum bitumen content (OBC) mixture, with 5.92% bitumen by weight. The unit weight is relatively high at 147.7 lb/ft³, indicating a dense mixture. The Voids in Mineral Aggregate (VMA) is 19.5%, reflecting the total void space between aggregate particles. Air Voids (Va) are low at 7.1%, suggesting good compaction. Voids Filled with Asphalt (VFA) is high at 79.76%, showing effective bitumen filling in the aggregate voids. The Corrected Marshall Stability is 17.57 kN, demonstrating strong resistance to deformation, while the flow value of 10.77 (1/100 in) indicates moderate flexibility. Together, these values indicate a well-balanced mix with good strength and adequate flexibility suitable for durable pavement performance.

4.2. Marshall mix design of bituminous mix with adding 3%, 5%, 8%, 10% & 12% of plastic waste

Table 3 shows the results of the Marshall Method of Mix Design for bituminous mixtures modified with varying percentages of plastic waste content. As the plastic waste content increases from 3.0% to 12.0%, several key properties of the mixture are affected. The Unit Weight decreases significantly with higher plastic waste content, indicating a less dense mixture due to the lower density of plastic compared to bitumen. The Voids in Mineral

Table 3. Marshall method of mixing design with plastic waste.

Sl. No.	% of Plastic Waste Content	Unit Weight (lb/ft³)	VMA (%)	Va (%)	VFA (%)	Corrected Marshall Stability (kN)	Flow (1/100) in
1	3.0	143.222	27.88	6.23	87.09	20.78	12.94
2	5.0	144.290	26.79	8.26	91.42	22.17	13.81
3	8.0	123.970	34.24	11.81	73.38	19.75	14.73
4	10.0	114.230	37.59	18.96	55.05	19.60	15.45
5	12.0	111.001	38.45	19.10	49.81	18.34	15.70

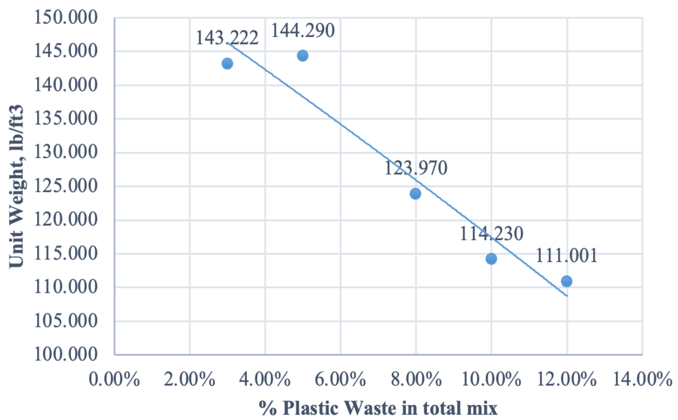

Figure 12. Unit weight of bitumen with waste materials addition.

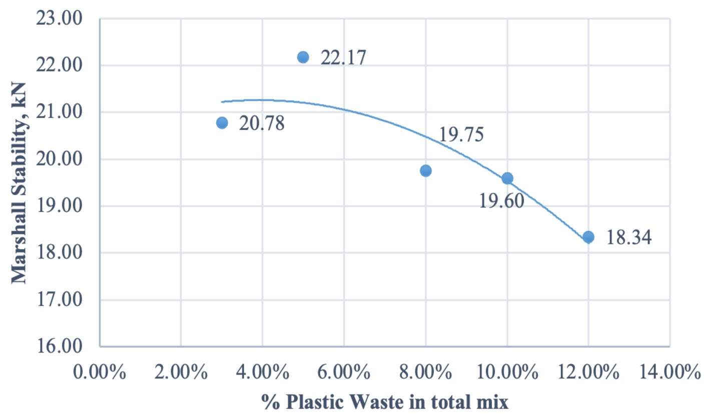

Figure 13. Marshall stability of bitumen with waste materials addition.

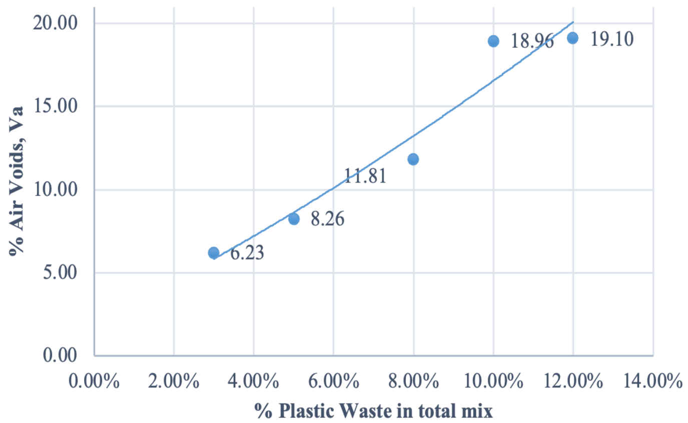

Figure 14. Percent of air with waste materials addition.

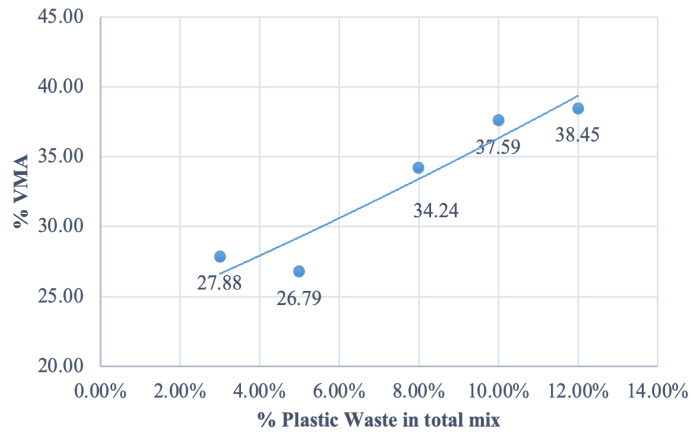

Figure 15. Percent of VMA with waste materials addition.

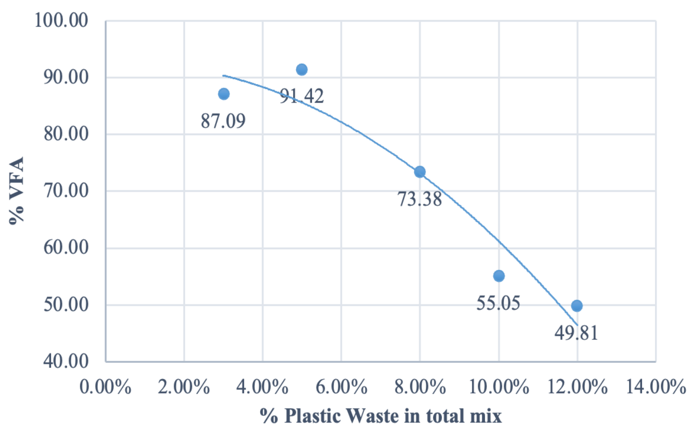

Figure 16. Percent of VFA with waste materials addition.

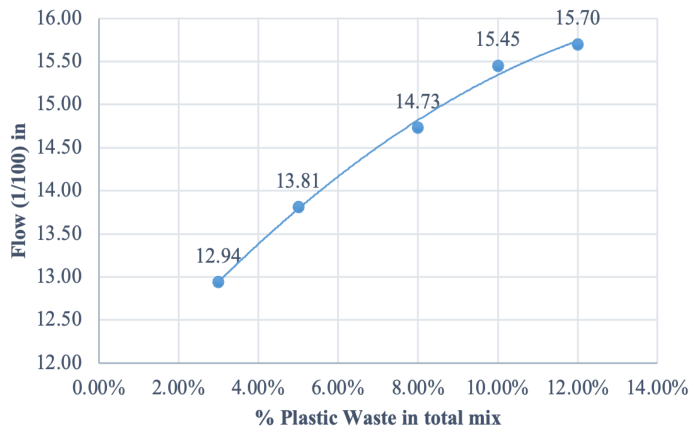

Figure 17. Flow (1/100) in with waste materials addition.

Aggregate (VMA) increases as plastic waste content rises, peaking at 12.0%, suggesting that more void space is left in the mixture. Similarly, Voids in Asphalt (Va) increases with higher plastic content, indicating that less bitumen is available to fill the voids. As a result, Voids Filled with Asphalt (VFA) decreases, reflecting the lower proportion of bitumen filling the aggregate voids as more plastic is added. The Corrected Marshall Stability, which measures the resistance to deformation, increases initially with higher plastic waste content, peaking at 5.0%, before decreasing at higher plastic contents, indicating that excessive plastic waste reduces the mixture's stability. Lastly, Flow, representing the flexibility of the mixture, increases as plastic waste content rises, suggesting that the mixture becomes more flexible and susceptible to deformation. In summary, while plastic waste improves the mixture's flexibility and stability up to a certain point (5.0%), higher plastic content reduces the overall stability and packing efficiency of the mix.

The provided figures visually demonstrate the relationship between the addition of plastic waste and various properties of the bituminous mixture, based on the

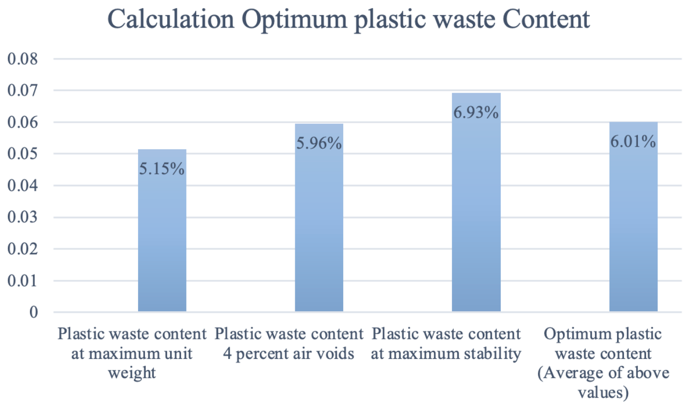

Figure 18. Calculation Optimum plastic waste content.

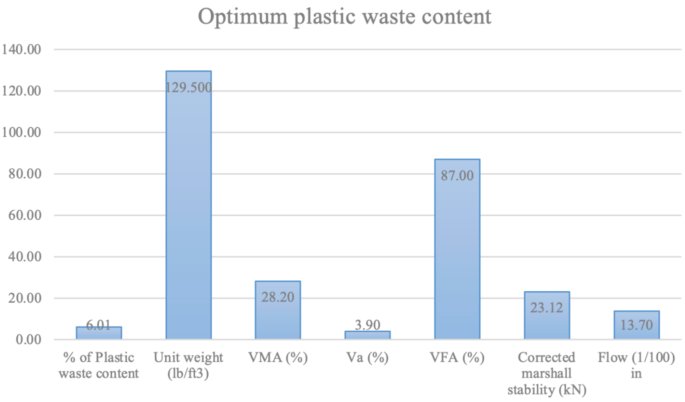

Figure 19. Optimum plastic waste content.

Marshall Method of Mix Design. Figure 12 shows that as plastic waste content increases, the Unit Weight of the mixture decreases, indicating that the mixture becomes less dense with higher plastic content. Figure 13 illustrates that the Marshall Stability decreases as plastic waste increases, reflecting a reduction in the mixture's resistance to deformation and rutting. Figure 14 shows that the Percent of Air Voids increases with more plastic waste, suggesting a decrease in compaction efficiency and the presence of more air pockets in the mixture. Figure 15 depicts the Percent of VMA (Voids in Mineral Aggregate), which increases as plastic waste content rises, reflecting a less efficient packing of the aggregates. Figure 16 shows that the Percent of VFA (Voids Filled with Asphalt) decreases as plastic content increases, indicating that the plastic waste reduces the amount of bitumen filling the voids in the aggregate, which can compromise stability. Finally, Figure 17 reveals that the Flow (the mixture's flexibility) increases with higher plastic waste content, indicating that the mixture becomes more flexible but also more prone to deformation under load. Collectively, these figures show that while the addition of plastic waste enhances flexibility, it also reduces compaction, stability, and the effectiveness of bitumen in filling the aggregate voids, particularly at higher plastic content levels.

Figure 18 illustrates the calculation of optimum plastic waste content based on different criteria. The plastic waste content corresponding to maximum unit weight, 4 percent air voids, and maximum stability are shown, along with their average value. The optimum plastic waste content is determined to be approximately 6.01%, balancing density, air voids, and stability to achieve the best overall performance in the bituminous mix.

Figure 19 presents the key properties of the optimum plastic waste content mixture with 6.01% plastic waste by weight. The unit weight is 129.5 lb/ft³, indicating the density of the mix. The Voids in Mineral Aggregate (VMA) is 28.2%, showing the total void space in the aggregate. Air Voids (Va) are low at 3.9%, reflecting good compaction. Voids Filled with Asphalt (VFA) is 87%, indicating a high percentage of aggregate voids filled with binder. The corrected Marshall stability is 23.12 kN, demonstrating strong resistance to deformation. The flow value of 13.7 (1/100 in) suggests increased flexibility compared to mixtures without plastic waste. Overall, these results indicate that the optimum plastic waste content improves mixture strength while maintaining adequate flexibility.

4.3. Interpretation of Flow Value and Flexibility-Stability Balance

The flow value in the Marshall mix design serves as an indicator of the mixture's flexibility, reflecting the deformation the mixture can withstand before failure. Moderate increases in flow value suggest enhanced flexibility, which helps the pavement resist cracking under repeated traffic loads. However, excessive flow values can indicate reduced stiffness and stability, making the mixture more susceptible to permanent deformation such as rutting and wear. This dual role implies that while plastic waste modification improves flexibility (as observed with increasing flow values), it can also reduce the load-bearing capacity when plastic content becomes too high. Therefore, an optimal balance between flexibility and stability is critical for durable pavement design, which in this study is observed around the 5% plastic content mark where Marshall Stability peaks.

Table 4. Comparison and discussion PET, LDPE and HDPE with relevant literature.

Plastic Type	Optimal Content (%)	Rutting Resistance Improvement (%)	Aging Resistance Improvement (%)	References
PET	6–8	15–20	10–15	Chegenizadeh et al. (2022) [39], Mashaan et al. (2025) [40]
LDPE	4–6	10–15	8–12	Mashaan et al. (2022) [41]
HDPE	4–6	12–17	9–14	Mashaan et al. (2022) [41]

4.4. Quantitative Comparison of Plastic Types (PET, LDPE, HDPE)

A comparative analysis of different plastic types used in bitumen modification highlights the superior performance of PET compared to LDPE and HDPE in improving rutting and aging resistance. Table 4 summarizes these findings based on this study and prior literature. PET-modified bitumen generally allows for higher plastic content and shows more significant improvements in resistance to rutting and aging compared to LDPE and HDPE, corroborating the results of this study.

4.5. Comparison with Previous Studies

Our findings align well with previous research indicating that PET-modified bitumen improves stability, stiffness, and aging resistance. Cherian et al. [5] reported enhanced rutting resistance and durability at similar PET contents. Likewise, Ramdas et al. [9] demonstrated superior mechanical properties with PET over other plastics. This study further contributes by incorporating nano-material additives, which synergistically improve the rheological and mechanical properties beyond those reported in previous works. Differences in optimal plastic content and flow values observed between studies can be attributed to variations in processing techniques, plastic particle sizes, and nano-material types or concentrations. These factors underscore the importance of careful material selection and processing parameters to optimize performance.

5. Conclusion and Recommendations

This study investigates the potential of modifying bitumen with waste plastic bottles (Polyethylene Terephthalate, PET) and various nano-materials, including carbon nanotubes (CNTs), graphene, nanosilica, and nanoclays, to enhance the physical, rheological, and durability properties of bituminous mixtures used in road construction. The experimental results demonstrate that the incorporation of plastic waste into bitumen significantly affects its performance. At moderate levels of plastic waste (3.0% to 5.0%), the modified bitumen exhibits improved flexibility, with increased Flow values, while maintaining reasonable Marshall Stability and unit weight. However, as the plastic content increases beyond 5.0%, the mixture's stability and packing efficiency decrease, leading to a drop in Marshall Stability and a rise in air voids. The optimal content for plastic waste was determined to be around 6.01%, where a balance between stability, flexibility, and compaction was achieved. Furthermore, the incorporation of nano-materials further enhanced the bitumen's resistance to rutting, cracking, and fatigue, indicating that plastic waste, combined with nano-modification, can serve as a sustainable and high-performance material for road construction.

Based on the findings, it is recommended that the optimal plastic waste content for bitumen modification be limited to around 6.01%, as higher plastic content may compromise stability and compaction. Further studies could explore the use of compatibilizers or alternative plastic types to enhance compatibility with bitumen at higher content levels. Additionally, while nano-materials such as CNTs, graphene, nanosilica, and nanoclays showed promising results, further investigation into their synergistic effects when combined with varying amounts of plastic waste is needed, with a focus on optimizing their concentrations for enhanced rheological and mechanical properties. Long-term performance testing under varying environmental and traffic conditions should also be conducted to assess durability, aging resistance, and the mixture's response to different climates. Moreover, it is crucial to evaluate the economic feasibility and scalability of this approach, considering the costs of recycling plastic and integrating nano-materials into bitumen. Finally, small-scale pilot projects should be initiated to assess the field performance of plastic-modified bitumen in real road construction settings, identifying potential challenges and providing data on its performance under actual traffic and environmental conditions. By addressing these recommendations, the road construction industry can move toward more sustainable, durable, and eco-friendly materials, reducing plastic waste while improving the lifespan and performance of asphalt pavements.

5. Conflicts of Interest

The authors declare no conflicts of interest.

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