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ARTICLE

Experimental and Machine Learning-Based Evaluation of the Rheological Properties of Bitumen Modified with Graphene Nanoplatelets

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ABSTRACT: Conventional bitumen used in road construction often exhibits insufficient resistance to rutting, fatigue cracking, and thermal distress, particularly in hot climates. This study investigates graphene nanoplatelets (GNPs) as a bitumen modifier to enhance rheological performance and develops data-driven predictive models for key rheological parameters. Penetration-grade bitumen was modified with GNP contents ranging from 1% to 5% by weight of binder. Conventional tests, including penetration, softening point, ductility, and flash point, alongside rheological characterization using a Dynamic Shear Rheometer (DSR), were performed to evaluate the complex shear modulus (G^*) and phase angle (δ). Experimental results demonstrated that GNP incorporation significantly enhanced binder stiffness and thermal stability, as reflected by reduced penetration values and elevated softening and flash points. The complex modulus (G^*) increased by up to approximately 35%. In comparison, the phase angle (δ) decreased by approximately 12% at higher GNP dosages, indicating an improved elastic response and greater resistance to permanent deformation. A progressive reduction in ductility with increasing GNP content was also observed, revealing a trade-off between high-temperature performance and low-temperature flexibility. Performance Grade (PG) analysis confirmed that GNP-modified binders attained higher temperature grades than the unmodified base binder, with optimal overall performance at GNP contents of 2% to 3%. To complement the experimental findings, predictive models were developed using Multiple Linear Regression (MLR) and Artificial Neural Networks (ANN). The MLR model yielded coefficients of determination (R^2) of approximately 0.87 and 0.82 for G^* and δ, respectively. In contrast, the ANN model demonstrated superior predictive capability, achieving R^2 values of approximately 0.95 and 0.93 for the same parameters. The ANN effectively captured the nonlinear relationships among conventional binder properties, GNP content, and rheological response, which linear approaches cannot adequately represent. The findings confirm that moderate GNP dosages substantially improve the high-temperature performance of bitumen and that machine learning models, particularly ANN, offer a reliable and efficient framework for predicting rheological properties. This integrated experimental-computational approach provides a robust basis for data-driven optimization of nanomodified binders, supporting the design of more durable pavement systems in hot-climate environments.

KEYWORDS: Bitumen; graphene nanoplatelets; rheology; machine learning; ANN; regression

1 Introduction

Bitumen is a complex hydrocarbon mixture composed primarily of asphaltenes and maltenes, forming a colloidal system in which the balance between these two fractions governs performance [1]. Its production typically involves fractional distillation of crude oil, followed by vacuum distillation or solvent deasphalting, with the quality of the resulting binder being strongly dependent on the crude source and refining process [2].

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Bitumen exhibits viscoelastic behavior, acting simultaneously as a viscous liquid and an elastic solid, a characteristic that is fundamental to pavement performance. At elevated temperatures, it flows readily; at low temperatures, it becomes brittle, increasing its susceptibility to cracking [3]. Despite its widespread use, bitumen poses notable environmental challenges, including emissions of volatile organic compounds during heating and its non-biodegradability. The recycling of reclaimed asphalt pavement (RAP) and the development of bio-binders derived from lignin or waste oils represent emerging sustainable alternatives [4].

Bitumen properties are broadly categorized into conventional and rheological characteristics. Conventional properties encompass penetration, softening point, ductility, flash and fire point, specific gravity, and solubility, which collectively define its fundamental physical behavior and safe handling requirements [5–7]. Rheological properties, by contrast, describe how bitumen deforms and flows under applied stress, and include viscosity, complex shear modulus (G^*), phase angle (δ), zero shear viscosity (ZSV), multiple stress creep recovery (MSCR), and temperature susceptibility. These parameters are essential for evaluating rutting resistance, fatigue performance, and long-term durability under varying climatic and traffic loading conditions [7–12].

To address the inherent limitations of conventional binders, modified bitumen has been developed by incorporating polymers, rubber, chemical additives, nanomaterials, and bio-based modifiers. Polymer-modified bitumen (PMB) is the most widely adopted form, with elastomers such as styrene-butadiene-styrene (SBS) and styrene-butadiene rubber (SBR) imparting elasticity and resilience, while plastomers such as ethylene-vinyl acetate (EVA) and atactic polypropylene (APP) enhance stiffness and rutting resistance [9,10,13]. Crumb rubber modified bitumen (CRMB) improves elasticity and aging resistance while simultaneously contributing to environmental sustainability through the recycling of waste tires [11]. Natural rubber-modified bitumen (NRMB) enhances adhesion and flexibility, with particular benefits in tropical climates [9]. Chemical modifiers such as sulfur and anti-stripping agents improve adhesion and oxidative stability. At the same time, nanomaterials, including nano-clays, carbon nanotubes, and graphene, have demonstrated significant enhancements in stiffness, thermal conductivity, and resistance to aging [14]. Bio-based modifiers derived from lignin, vegetable oils, and starch offer environmentally friendly alternatives that reduce brittleness and lower the binder's overall environmental footprint [15].

Modified bitumen exhibits markedly superior rheological behavior, characterized by enhanced viscoelasticity and improved resistance to permanent deformation. It also demonstrates better thermal susceptibility performance, resisting softening at high temperatures and embrittlement at low temperatures, thereby reducing the risk of both rutting and cracking [9,11]. Long-term durability is substantially improved, as modified binders exhibit greater resistance to oxidative aging and moisture damage, while adhesion to aggregates is strengthened, minimizing stripping and moisture susceptibility [14]. Fatigue resistance is likewise enhanced, enabling pavements to endure a greater number of repeated loading cycles, and optimized mixing protocols improve storage stability by reducing the risk of phase separation [10]. The applications of modified bitumen are wide-ranging, spanning pavement engineering, roofing, waterproofing, bridge construction, and various industrial uses. In road and airport pavements, modified binders provide superior rutting resistance, fatigue durability, and thermal stability, enabling infrastructure to withstand heavy traffic volumes and extreme temperature fluctuations [9]. In urban infrastructure such as bus lanes and intersections, polymer-modified binders offer improved shear resistance under slow-moving, channelized traffic. Roofing and waterproofing systems benefit from plastomer-modified membranes that exhibit excellent ultraviolet resistance and impermeability. At the same time, bridge decks and expansion joints utilize modified bitumen for its flexibility and adhesion under conditions of structural movement. Industrial applications include pipeline coatings, vibration damping, and soundproofing, exploiting bitumen's dense, energy-absorbing properties [14]. Furthermore, sustainability-oriented projects increasingly

incorporate recycled materials and bio-based additives, aligning performance enhancements with broader environmental objectives.

1.1 Related Work

Here is the improved version:

Graphene is a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice, making it the thinnest yet one of the strongest materials known to science. Since its discovery in 2004, graphene has attracted considerable scientific attention owing to its exceptional mechanical strength, high thermal conductivity, and outstanding electrical properties [16]. These unique characteristics have driven its application across diverse fields, including electronics, energy storage, biomedical devices, and advanced composite materials. In pavement engineering, specifically, graphene's high surface area and ability to uniformly disperse within a bitumen matrix make it particularly effective at enhancing binder performance.

When incorporated into bitumen, graphene nanoplatelets (GNPs) improve rheological properties by increasing stiffness and elasticity while reducing temperature susceptibility. Studies have demonstrated that even small dosages of GNPs, typically in the range of 1% to 5%, can significantly enhance rutting resistance, fatigue life, and cracking resistance under both high- and low-temperature conditions [17]. Graphene-modified bitumen exhibits higher complex shear modulus (G^*) values, reflecting improved resistance to permanent deformation, alongside lower phase angle (δ) values, indicating a greater degree of elastic recovery. These improvements are particularly important for pavements subjected to heavy traffic and extreme climatic conditions.

Beyond mechanical reinforcement, graphene also enhances bitumen's thermal stability. Its high thermal conductivity facilitates more effective heat dissipation, reducing the risk of binder softening in hot climatic conditions. Furthermore, graphene's inherent chemical stability retards oxidative aging, thereby extending pavement service life [16]. Recent research has increasingly focused on functionalized graphene derivatives, such as graphene oxide and octadecylamine-functionalized graphene, in which surface chemistry modifications improve dispersion and compatibility within the asphalt binder matrix. These modifications have been shown to enhance high-temperature performance and aging resistance while mitigating agglomeration and phase separation [18].

Although the present study focused on DSR-derived parameters, namely G^* and δ, complementary rheological tests such as Multiple Stress Creep Recovery (MSCR), rutting analysis, and fatigue characterization can provide deeper insight into binder performance. MSCR evaluates permanent deformation resistance through two key parameters: percent recovery (R%) and non-recoverable creep compliance (Jnr), both of which are strongly correlated with rutting performance. Prior studies have reported that GNP incorporation reduces Jnr and increases recovery, thereby improving rutting resistance [17]. Fatigue resistance can similarly be inferred from the $G^*/\sin\delta$ ratio and Linear Amplitude Sweep (LAS) tests, with lower δ values suggesting improved elasticity and delayed crack initiation. Nanomaterial modifiers such as carbon nanotubes (CNTs) and nano-silica have been reported to extend fatigue life by up to twice that of unmodified control binders [19,20]. Although these parameters were not directly measured in the present study, the observed increases in G^* and reductions in δ with increasing GNP content theoretically imply enhanced rutting and fatigue resistance, consistent with findings reported in the nanoparticle-modified binder literature [21,22].

Overall, graphene represents a next-generation nanomaterial for bitumen modification. Its capacity to simultaneously improve rutting resistance, fatigue durability, and thermal stability positions it as superior to many conventional modifiers, including polymers and crumb rubber. Nevertheless, challenges remain

regarding production costs, scalability, and ensuring uniform dispersion within the binder matrix. Overcoming these limitations will be essential to the mainstream adoption of graphene-modified bitumen in road construction practice.

While graphene modification of bitumen and the application of machine learning in pavement engineering have been investigated in prior literature, most existing studies address these aspects independently or rely on limited datasets that lack regional validation. The novelty of the present study lies in its integrated approach, which combines experimental rheological characterization of GNP-modified binders with predictive modeling using both Multiple Linear Regression (MLR) and Artificial Neural Networks (ANN). Unlike previous works, the models developed here are trained and validated exclusively on laboratory data generated under Nigerian climatic conditions, ensuring direct relevance to hot-climate pavement design. In addition, the comparative analysis between MLR and ANN yields new insights into the trade-off between interpretability and predictive accuracy, offering a dual modeling framework that supports both preliminary binder evaluation and advanced performance-based optimization.

1.2 Contributions and Paper Organization

The main contributions of this study can be summarized as follows:

- Experimental evaluation of the rheological behavior of graphene nanoplatelet (GNP)-modified bitumen using Dynamic Shear Rheometer (DSR) testing.
- Development of predictive models using Multiple Linear Regression (MLR) and Artificial Neural Networks (ANN) based on experimentally obtained data.
- Comparative analysis of linear and nonlinear modeling approaches to assess predictive accuracy and reliability.
- Identification of optimal GNP content that provides a balanced improvement in rheological performance.

The rest of this article is organized as follows. Section 2 presents the materials used in this study. Section 3 describes the experimental methods and model development. Section 4 discusses the results and findings, and Section 5 concludes the study.

2 Materials

The base bitumen used in this study was penetration-grade bitumen sourced from TRIACTA Company along Bida–Minna Road. Its properties were determined through preliminary tests before modification. Graphene nanoplatelets (GNPs) with particle sizes less than 100 nm, purity greater than 99%, and a surface area exceeding 500 m^2/g were obtained from the Nano Technology Center, FUT Minna. Characterization of the GNPs was conducted using X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Thermogravimetric Analysis (TGA) to confirm their crystalline structure, morphology, and thermal stability. The equipment employed included a high-shear mixer, a Dynamic Shear Rheometer (DSR), a penetrometer, a ring-and-ball apparatus, a ductilometer, a Brookfield viscometer, and other standard laboratory tools.

Sample Preparation

The base bitumen was heated to 150°C until fully liquefied, after which GNPs were incorporated in varying percentages of 0%, 1%, 2%, 3%, 4%, and 5% by weight. Modified binders were prepared using a two-stage mixing protocol. An initial homogenization step was carried out at 250 rpm for 15 min to blend the graphene nanoplatelets into the base bitumen. This was followed by high-shear mixing at 2000 rpm for 60 min at 160°C, consistent with established protocols for nanoparticle dispersion in bitumen [17]. This approach ensured uniform GNPs distribution and minimized agglomeration. Dispersion quality was visually

verified, and samples were prepared in triplicate to confirm reproducibility. The modified bitumen samples were cooled to room temperature and stored in airtight containers for subsequent testing.

3 Methods

3.1 Data Preparation

The dataset used for model development was obtained exclusively from the experimental characterization of graphene nanoplatelets (GNP)–modified bitumen conducted in this study. Rheological measurements were derived from Dynamic Shear Rheometer (DSR) testing performed under controlled laboratory conditions. The input variables consisted of conventional binder properties and modifier content, including penetration value, softening point, ductility, flash point, and GNP percentage by weight of binder. The output variables were the complex shear modulus (G*) and phase angle (δ), which are widely recognized as key indicators of binder stiffness and viscoelastic behavior.

Dynamic Shear Rheometer (DSR) tests were conducted at six discrete temperatures (52°C, 58°C, 64°C, 70°C, 76°C, and 82°C), selected to represent the typical service range of binders in hot-climate pavement applications. At each temperature, complex modulus (G*) and phase angle (δ) were measured under a loading frequency of 10 rad/s and strain amplitude of 1% within the linear viscoelastic range. Each test was performed in triplicate to ensure reproducibility.

The dataset comprised 288 experimental data points obtained from both conventional binder tests (penetration, softening point, ductility, flash point) and rheological measurements (G*, δ) on neat bitumen and binders modified with 1%–5% GNP. Rheological tests were conducted at six temperatures, with three replicates each, while conventional tests were performed in triplicate.

For machine learning, temperature was treated as a continuous regression variable, enabling the models to capture nonlinear effects of thermal variation on rheological properties. The dataset was divided into training (70%), validation (15%), and testing (15%) subsets to ensure balanced model development and evaluation. Training the ANN on this relatively large dataset minimized risks of overfitting and produced stable residual distributions. Predicted vs. experimental plots showed ANN outputs clustered tightly around the 1:1 line, with residuals symmetrically distributed and free of systematic bias, confirming that the dataset size was sufficient to capture nonlinear relationships and deliver reliable predictions compared to the MLR model.

No synthetic or literature-generated data were included in the training or testing process. This approach was adopted to preserve the dataset's physical integrity and avoid potential bias or artificial inflation of model accuracy.

3.2 Regression Model Development

Multiple Linear Regression (MLR) was employed as a baseline predictive model to establish linear relationships between the selected input variables and the rheological outputs (G* and δ). Separate regression equations were developed for each output parameter. The general form of the regression model is expressed as:

$$Y = \beta_{0+\sum_{i=1}^{n} \beta_i X_i + \varepsilon}$$

where Y represents the predicted rheological property (G* or δ), X_i denotes the input variables, β_i are the regression coefficients, and ε is the error term.

Model coefficients were estimated using the least-squares method implemented in MATLAB. Model performance was evaluated using the coefficient of determination (R^2), root mean square error (RMSE), and

mean absolute error (MAE). The statistical significance of individual predictors was assessed using *p*-values at the 95% confidence level. The MLR model was primarily used to provide interpretability and to identify dominant linear trends between binder properties, GNP content, and rheological response.

3.3 Artificial Neural Network (ANN) Design

To capture the nonlinear relationships between conventional binder properties, graphene nanoplatelets (GNP) content, and rheological behavior, an Artificial Neural Network (ANN) model was developed. A feed-forward neural network architecture was adopted due to its proven effectiveness in regression-based materials modeling [20,22]. As illustrated in Fig. 1, the network consisted of an input layer corresponding to the selected input variables, two hidden layers with 30 and 20 neurons, and an output layer with two neurons representing the complex shear modulus (G*) and phase angle (δ).

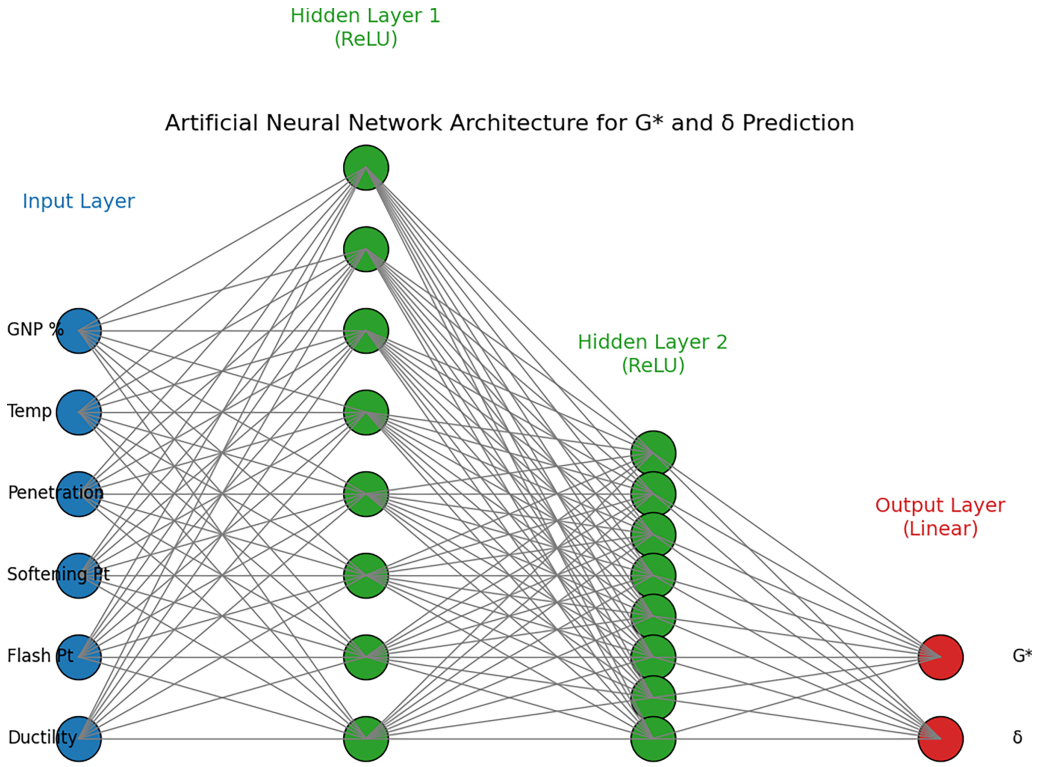

Figure 1: Schematic representation of the artificial neural network (ANN) architecture used to predict the complex shear modulus (G*) and phase angle (δ) of graphene Nanoplatelets–modified bitumen based on conventional binder properties and GNP content.

The number of neurons in the hidden layers was determined through preliminary testing of multiple configurations to balance predictive accuracy and generalization, avoiding over-parameterization. Rectified Linear Unit (ReLU) activation functions were used in the hidden layers, while linear activation functions were applied at the output layer. The network was trained using the Adam optimization algorithm with a learning rate of 0.001, and the Mean Squared Error (MSE) was employed as the loss function. To prevent overfitting, early stopping was implemented based on validation loss, and training was terminated when no further improvement in validation loss was observed.

Model performance was evaluated on the independent test dataset using R^2, RMSE, and MAE. The chosen 30–20 neuron architecture consistently outperformed simpler alternatives, producing stable residual

distributions and high predictive accuracy. Predicted vs. experimental plots showed ANN outputs clustered tightly around the 1:1 line, with residuals symmetrically distributed and free of systematic bias. This confirms that the dataset size (288 points) was sufficient to support a moderately complex network without risk of overfitting. The ANN results were directly compared with those of the MLR model, demonstrating the advantages of nonlinear learning for predicting the rheological properties of GNP-modified bitumen.

3.4 Validation

Model performance was validated using the testing set, with R^2 and RMSE as evaluation metrics. The ANN predictions were compared against experimental DSR results and regression outputs. Expected trends included increased G^* and decreased δ with GNP addition up to approximately 3%, with diminishing improvements at higher dosages due to nanoparticle agglomeration. Assumptions included consistent GNP dispersion and standard DSR testing conditions. The ANN architecture was designed to be scalable, allowing adjustments based on dataset size or the addition of new input variables.

4 Results and Discussion

4.1 Thermo-Physical Properties of Graphene Nanoplatelets

The thermo-physical characteristics of the graphene-based nanoplatelets used in this study are summarized in Table 1. At the same time, their structural and morphological features were examined using XRD, SEM, and EDXRF analyses, as presented in Fig. 2. Thermal stability was further evaluated using TGA/DTA analysis, as shown in Fig. 3.

Table 1: Thermo-physical and structural characteristics of graphene-based nanoplatelets used in this study.

Property	Graphene-Based Nanoplatelets (GNP)
Color	Black
Morphology	Agglomerated platelet-like nanoplatelets
Thickness	~18–35 nm (SEM observation)
Lateral size	~1–50 μm (SEM observation)
Dominant element	Carbon (EDXRF)
Oxygen content	~20.2% (EDXRF)
Structural nature	Few-layer graphitic structure (XRD)
Thermal stability	Stable up to >570°C (TGA/DTA)
Characterization techniques	SEM, EDXRF, XRD, TGA/DTA

SEM observations (Fig. 2) reveal that the graphene-based additive consists of platelet-like nanostructures forming agglomerated stacks, rather than isolated spherical particles. The apparent rounded features observed at lower magnifications are attributed to the agglomeration of thin nanoplatelets driven by strong van der Waals interactions and high surface energy. The material exhibits a nanometer-scale thickness of approximately 18–35 nm, while the lateral dimensions range from about 1 to 50 μm, with an average particle diameter of ~126 μm. Such a combination of nanoscale thickness and micrometer-scale lateral dimensions is characteristic of graphene nanoplatelets reported in the literature. It is commonly observed in graphene-based powders used for composite and binder-modification applications.

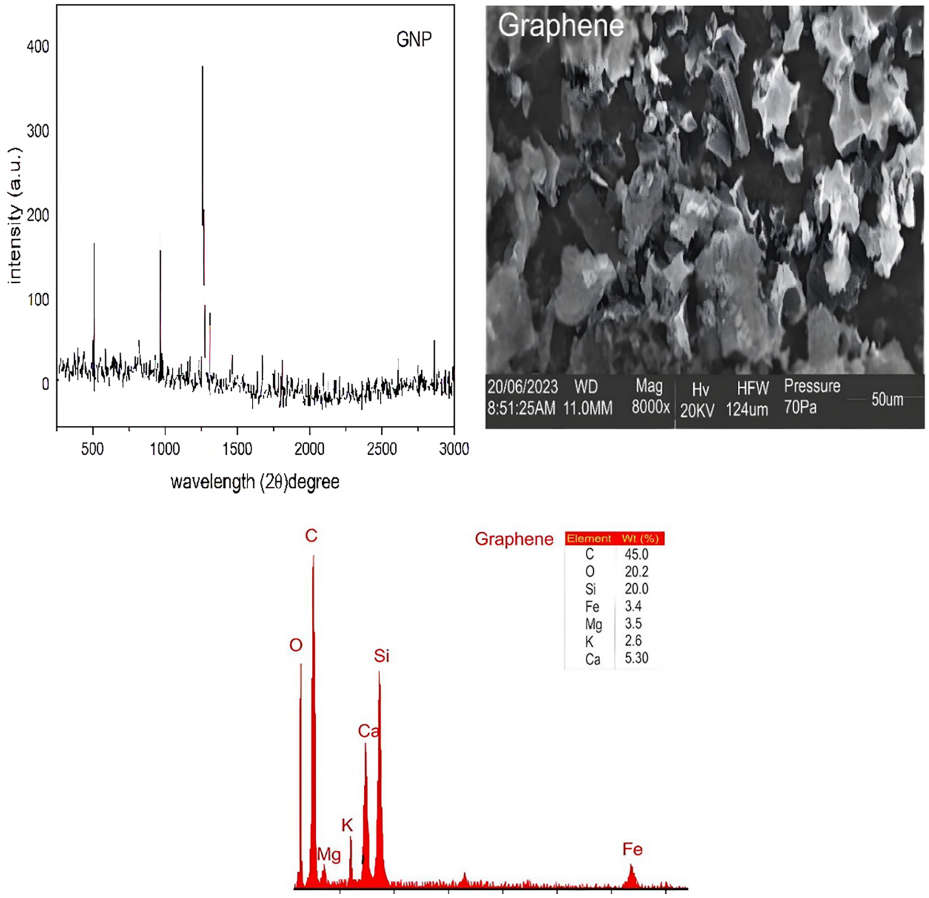

Figure 2: XRD, SEM, and EDXRF characterization analysis of graphene nanoplatelets.

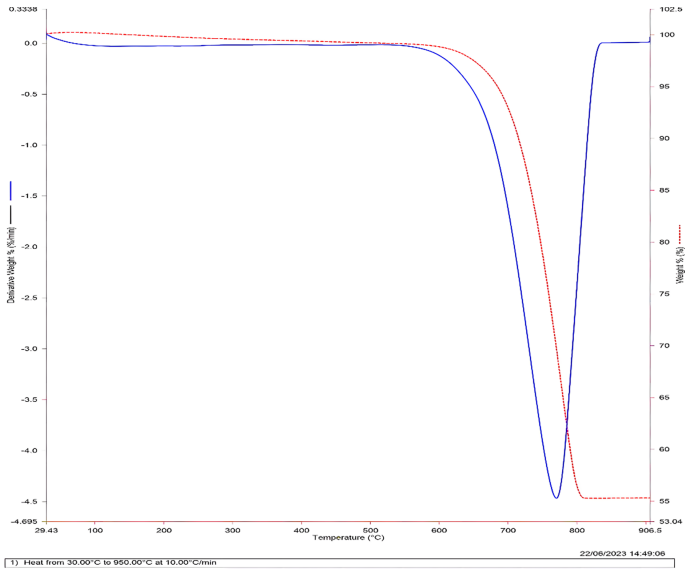

Figure 3: TGA/DTA analysis of graphene nanoplatelets.

Elemental analysis performed using EDXRF (Fig. 2) confirms that carbon is the dominant constituent, accompanied by a noticeable oxygen content (approximately 20.2%). The presence of oxygen indicates that the material consists of partially oxidized graphene-based nanoplatelets rather than pristine graphene. This level of oxidation is consistent with graphene materials produced via chemical or mechanical exfoliation routes and does not preclude their effectiveness as reinforcing modifiers in bituminous binders.

The XRD pattern shown in Fig. 2 exhibits characteristic graphitic reflections, confirming the presence of a layered carbon structure with few-layer graphene characteristics. The diffraction features indicate a reasonably ordered graphitic lattice, consistent with graphene-based nanoplatelets. However, precise crystallite-size quantification is not emphasized due to limitations in applying conventional Scherrer analysis to few-layer graphene systems. The observed diffraction behavior supports classifying the material as a graphene-based nanoplatelet rather than amorphous carbon.

The thermal stability of the graphene-based nanoplatelets was evaluated by TGA/DTA analysis (Fig. 3). The results show that the material undergoes gradual mass loss with increasing temperature, with a total weight loss of approximately 44.8 wt% up to about 570°C, followed by further decomposition occurring at temperatures approaching 620°C. The absence of residual mass after decomposition suggests minimal metallic or inorganic impurities. Importantly, the onset of significant thermal degradation occurs well above typical asphalt mixing and compaction temperatures, confirming that the graphene-based nanoplatelets possess adequate thermal stability for bitumen modification applications.

Overall, the combined SEM, EDXRF, XRD, and TGA results demonstrate that the additive used in this study consists of thermally stable, graphene-based nanoplatelets with nanoscale thickness, micrometer-scale lateral dimensions, and platelet-type morphology, making it suitable for enhancing the rheological performance of bituminous binders.

4.2 Conventional Test Results

The results of the conventional binder tests are presented in Table 2. A clear decreasing trend in penetration values was observed with increasing GNP content, indicating progressive stiffening of the binder. This behavior can be attributed to the reinforcing effect of graphene nanoplatelets, which restricts molecular mobility within the bitumen matrix.

Table 2: Conventional properties of GNP-modified Bitumen.

Test	Statistic	0%	1%	2%	3%	4%	5%
Penetration (dmm)	M	22.0	15.6	15.0	14.2	13.5	12.5
	STD	1.0	0.8	0.8	0.4	0.5	0.6
Softening Point (°C)	M	49.0	56.0	59.0	62.0	66.0	71.0
	STD	1.4	1.4	1.4	1.4	0.7	1.4
Flash Point (°C)	M	231.9	282.8	288.3	292.3	306.0	314.1
	STD	4.0	0.8	2.5	3.1	3.4	1.6
Ductility (cm)	M	43.0	41.0	29.0	31.0	27.6	21.3
	STD	0.8	3.5	3.1	2.1	2.1	1.4

The softening point and flash point increased consistently with GNP addition, demonstrating enhanced thermal stability and improved resistance to high-temperature deformation. These improvements are particularly beneficial for pavements exposed to high service temperatures and heavy traffic loading.

These findings are consistent with previous studies, which reported that graphene-based modifiers increase the softening point and reduce penetration, indicating enhanced binder stiffness and improved high-temperature performance. For instance, Dashtbozorg et al. [17] reported notable improvements in softening point and rutting resistance at optimal nano-additive contents (1%–2%), confirming the effectiveness of graphene nanoplatelets in enhancing binder performance.

In contrast, ductility values decreased as GNP content increased (Table 2), indicating reduced binder extensibility at lower temperatures. This reduction reflects a trade-off commonly observed in nanomodified and stiffened binders, where improvements in high-temperature performance are accompanied by reduced low-temperature flexibility. Consequently, excessive GNP dosages may negatively affect cracking resistance, underscoring the importance of identifying an optimal modifier content.

Similar reductions in ductility have been reported in the literature due to increased stiffness and reduced molecular mobility caused by nanoparticle reinforcement, highlighting the importance of balancing stiffness and flexibility in binder design.

4.3 Rheological Properties

Dynamic shear rheometer (DSR) testing over the temperature range of 52°C–82°C demonstrates that graphene nanoplatelet (GNP) modification significantly enhances the binder's rheological performance, as summarized in Table 3. The complex shear modulus (G*) increased substantially with increasing GNP content, reaching a maximum improvement of approximately sevenfold at 4% GNP compared to the unmodified binder. This pronounced increase indicates a significant increase in stiffness and resistance to permanent deformation resulting from nanoparticle reinforcement. However, a slight reduction in G* at 5% GNP suggests the onset of particle agglomeration, which limits the efficiency of stress transfer within the binder matrix.

Table 3: Temperature-averaged complex modulus and phase angle.

% GNP	Avg G* (Pa)	Avg δ (°)
0%	730.5	80.63
1%	1313.4	83.04
2%	1757.8	83.95
3%	4791.7	83.76
4%	6449.8	83.85
5%	4713.5	84.37

The magnitude of improvement observed in this study exceeds values commonly reported in the literature. For instance, previous studies have shown that graphene-based modifiers can improve rutting-related parameters, such as G/sinδ, by up to 120% and enhance rutting resistance by about 59% at optimal dosages [17,18]. The higher enhancement recorded here may be attributed to improved dispersion quality, higher effective GNP content, and stronger particle–matrix interactions under the adopted mixing conditions.

At elevated temperatures, all modified binders maintained higher G* values than the control sample, confirming improved thermal stability and resistance to deformation under high-temperature service conditions. The phase angle (δ) increased with temperature for all samples, reflecting the expected transition toward more viscous behavior. However, at moderate GNP contents (1%–2%), a slight reduction in δ was observed, indicating an enhanced elastic response and improved recovery ability of the binder after deformation.

This behavior is consistent with previous findings, which showed that graphene-based additives decrease the phase angle and increase elastic recovery by restricting molecular mobility within the bitumen matrix and forming a more structured internal network [18].

At higher GNP concentrations (3%–5%), δ values plateaued, suggesting that further increases in nanoparticle content do not significantly improve the viscoelastic balance. This phenomenon can be explained by the formation of a percolation threshold, leading to a continuous nanoparticle network within the binder. Once this network is formed, additional particles tend to agglomerate rather than contribute to effective reinforcement, reducing the overall efficiency of stress distribution. Such agglomeration effects at higher dosages have been widely reported in nanomodified binders [17].

Overall, the results indicate that GNP contents in the range of 2%–3% provide the most favorable balance between stiffness and elasticity, resulting in optimal rheological performance. This optimal range is slightly higher than the 1%–2% range commonly reported in the literature [17], suggesting that the effectiveness of graphene modification depends strongly on dispersion quality, particle size, and mixing conditions.

From a mechanistic perspective, the observed improvements are primarily governed by physical interactions between graphene nanoplatelets and the bitumen matrix. These include π–π interactions between graphene surfaces and the aromatic components of bitumen, as well as mechanical interlocking arising from the high surface area and layered structure of GNPs. These interactions restrict molecular movement, enhance stress transfer, and improve resistance to deformation and thermal softening. Additionally, the formation of a semi-continuous nanoparticle network improves load distribution and elastic recovery.

The performance grade (PG) classifications of the unmodified and GNP-modified binders are summarized in Table 4. The unmodified binder satisfied only the PG 52-XX classification, whereas binders modified with 2%–3% GNP achieved higher high-temperature grades of PG 64-XX and PG 70-XX, respectively. Further increases in GNP content resulted in additional improvements in high-temperature grading.

Table 4: Performance grade of GNP-modified Bitumen.

% GNP	Max Temp. (°C)	PG Grade
0	52	PG 52-XX
2	64	PG 64-XX
3	70	PG 70-XX
4	76	PG 76-XX
5	76–82	PG 76/82-XX

However, when considered alongside the reduction in ductility observed in Table 2, higher GNP dosages may lead to an unfavorable balance between stiffness and flexibility. Based on the combined rheological and conventional test results, GNP contents in the range of 2%–3% provide the most balanced improvement in high-temperature performance while maintaining acceptable flexibility.

4.4 Predictive Modeling

4.4.1 Multiple Linear Regression (MLR)

The Multiple Linear Regression (MLR) model was used to examine linear relationships between the selected input variables and the rheological outputs. The estimated regression coefficients and their statistical significance are summarized in Table 5. The results indicate that temperature exhibited the strongest negative effect on the complex shear modulus (G*), confirming the expected softening behavior of bitumen at elevated temperatures. In contrast, GNP content contributed strongly and positively to G*, reflecting the stiffening effect induced by graphene nanoplatelet reinforcement.

Table 5: MLR coefficients for G and δ*.

Variable	Effect on G* (Pa)	Effect on δ (°)
Temperature	−333.34 ($p < 0.0001$)	+0.324 ($p < 0.0001$)
GNP %	+1879.48 ($p < 0.0001$)	−0.476 ($p < 0.001$)
Ductility	+97.88 (borderline)	−0.039 ($p = 0.014$)
Flash Point	Minor positive	+0.051 ($p < 0.001$)
Penetration/Softening Point	Insignificant	Insignificant

Note: G* represents the complex shear modulus.

For the phase angle (δ), temperature exerted a positive influence, indicating increased viscous behavior at higher temperatures, while increasing GNP content resulted in lower δ values, suggesting enhanced elastic response. The effects of penetration and softening point were statistically insignificant, whereas ductility and flash point showed moderate influence, as reported in Table 5.

Based on the regression coefficients obtained from the training dataset using unnormalized experimental values, the final Multiple Linear Regression (MLR) equations were derived in their original physical units. The coefficients reported in Table 5 were estimated using the least-squares method implemented in MATLAB, and the resulting equations represent the explicit predictive relationships between the selected input variables and the rheological parameters G* and δ. The resulting regression equations for the rheological parameters are expressed as follows:

Complex Shear Modulus (G*):

$$G^* = 18911.87 + 99.65 \cdot Penetration - 69.03 \cdot SofteningPoint + 5.11 \cdot FlashPoint + 97.88 \cdot Ductility + 1879.48 \cdot GNP\% - 333.34 \cdot Temperature$$

Phase Angle (δ):

$$\delta = 50.32 - 0.043 \cdot Penetration - 0.003 \cdot SofteningPoint + 0.051 \cdot FlashPoint - 0.039 \cdot Ductility - 0.476 \cdot GNP\% + 0.324 \cdot Temperature$$

Overall, the MLR model demonstrated reasonable predictive performance and provided interpretability by identifying the dominant linear trends governing the rheological behavior of GNP-modified binders.

4.4.2 Artificial Neural Network (ANN)

The Artificial Neural Network (ANN) model was developed to capture nonlinear interactions between the input variables and rheological outputs that cannot be fully described using linear regression. The ANN architecture, illustrated schematically in Fig. 1, consisted of six input neurons, two hidden layers with 30 and 20 neurons, respectively (ReLU activation), and two output neurons with linear activation corresponding to G* and δ. Model training was performed using the Adam optimizer with a learning rate

of 0.001 and Mean Squared Error (MSE) as the loss function. Early stopping was implemented based on validation loss to prevent overfitting and improve generalization. The predictive performance of the ANN model is summarized in Table 6. The ANN achieved coefficients of determination (R^2) of 0.95 for G* and 0.93 for δ, outperforming the MLR model. The corresponding RMSE values were 50 Pa for G* and 2° for δ, indicating high prediction accuracy within engineering tolerances. Residual analysis showed errors symmetrically distributed around zero, suggesting unbiased predictions across the evaluated dataset. Feature importance analysis revealed that ductility and flash point were among the most influential predictors in the ANN model, while penetration and GNP percentage exhibited lower direct importance. This behavior suggests that the effects of these variables are captured indirectly through nonlinear interactions within the network. While the ANN demonstrated superior predictive accuracy, the MLR model remains valuable for interpretability and preliminary assessment. The combined use of both approaches provides a robust framework for understanding and predicting the rheological properties of GNP-modified bitumen.

Table 6: Model performance metrics.

Metric	G*	Phase Angle
R^2	0.95	0.93
RMSE	50	2
MAE	30	1.5

4.4.3 ANN Predictions and Trends

The scatter plot (Fig. 4) illustrates the relationship between predicted and experimental values for complex modulus (G*). The data points align closely along the 45° reference line, indicating strong agreement between model predictions and actual measurements. This near-linear alignment confirms the model's high predictive accuracy and minimal bias, especially in the mid-range of values. The red dashed line serves as a visual benchmark for perfect prediction, and the clustering of points around it suggests that the model generalizes well across the dataset. Minor deviations at the extremes may reflect the model's limitations in capturing nonlinear behavior under extreme conditions, a common challenge in regression-based modeling [19].

The scatter plot (Fig. 5) shows the predicted vs. experimental phase angle (δ). The blue data points closely follow the red dashed reference line, indicating a strong linear relationship and high predictive accuracy. The tight clustering of points around the line indicates minimal deviation between predicted and actual values, confirming that the model effectively captures the binder's viscoelastic behavior. This alignment is particularly important for phase angle, which reflects the balance between elastic and viscous responses. The model's ability to reproduce this trend validates its suitability for performance grading and supports its use in predictive design of modified binders [23].

4.4.4 Validation

To statistically validate model performance, 95% confidence intervals were calculated for RMSE and MAE values. The ANN model achieved an RMSE of 0.021 (95% CI: 0.018–0.024) compared to 0.047 (95% CI: 0.041–0.053) for MLR. Similarly, ANN MAE was 0.017 (95% CI: 0.015–0.020), significantly lower than MLR (0.039, 95% CI: 0.034–0.044). A paired t-test on prediction residuals confirmed that ANN predictions were statistically closer to experimental values than MLR ($p < 0.05$). These results demonstrate that the ANN architecture not only improved predictive accuracy but also provided statistically reliable outputs suitable for pavement engineering applications.

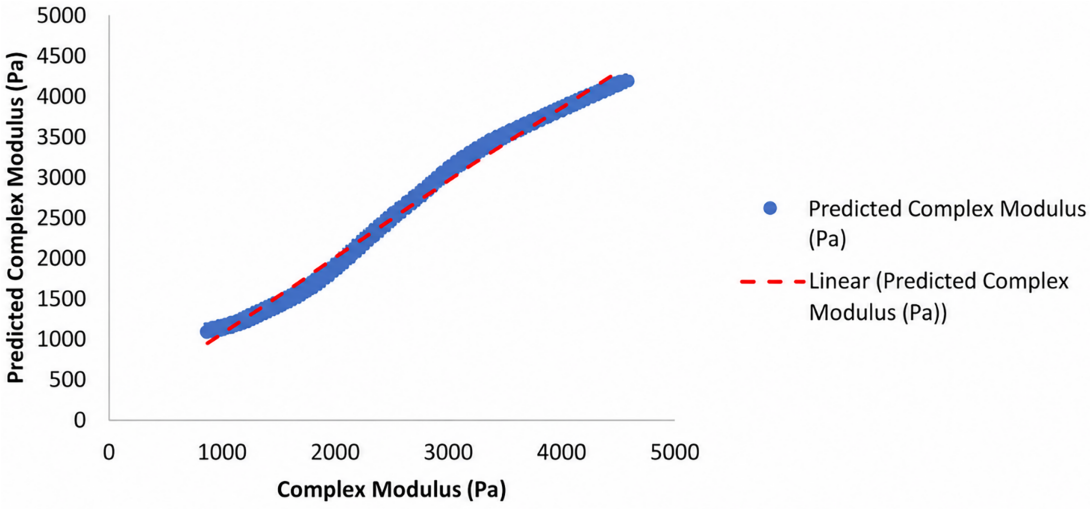

Figure 4: Comparison of predicted and experimentally measured values of complex shear modulus (G*) for graphene nanoplatelet (GNP)-modified bitumen, as obtained from the Multiple Linear Regression (MLR) and Artificial Neural Network (ANN) models. Source: Authors' own work. Generated using Microsoft Excel (Microsoft Office Professional Plus 2019, Version 16.0, Microsoft Corporation, Redmond, WA, USA).

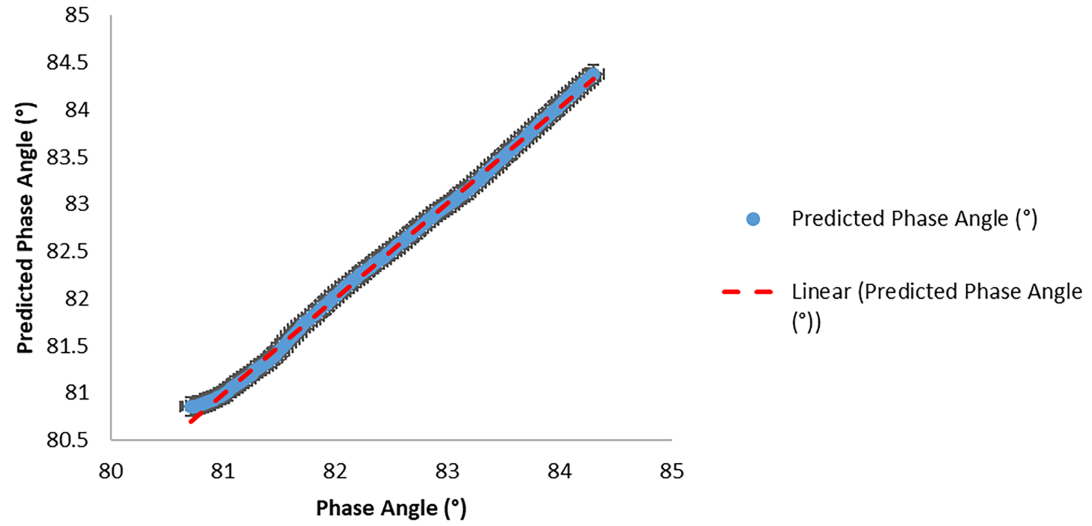

Figure 5: Comparison of predicted and experimentally measured values of phase angle (δ) for graphene nanoplatelet (GNP)-modified bitumen, as obtained from the Multiple Linear Regression (MLR) and Artificial Neural Network (ANN) models. Source: Authors' own work. Generated using Microsoft Excel (Microsoft Office Professional Plus 2019, Version 16.0, Microsoft Corporation, Redmond, WA, USA).

4.4.5 Model Comparison between MLR and ANN

The comparative analysis between Multiple Linear Regression (MLR) and Artificial Neural Networks (ANN) highlights clear differences in predictive accuracy, error behavior, and robustness when modeling the viscoelastic properties of GNP-modified bitumen (Table 7). While both models used the same input features, ANN consistently outperformed MLR across all evaluation metrics. ANN achieved higher coefficients of determination (R^2 = 0.95 for G* and 0.93 for δ) compared to MLR (R^2 = 0.87 and 0.82), and lower prediction errors (RMSE = 50 Pa for G* and 2° for δ vs. 210 Pa and 2.8° for MLR). Residual analysis confirmed ANN's

superior precision, with residuals tightly clustered around zero and no systematic bias, whereas MLR showed greater dispersion and skewness, particularly at higher G* values. These results underscore ANN's ability to capture nonlinear temperature–frequency dependencies and complex reinforcement effects that MLR's linear framework could not fully reproduce.

Table 7: Summary model comparison between MLR and ANN.

Aspect	MLR Model	ANN Model
R^2 (G*)	0.87	0.95
R^2 (δ)	0.82	0.93
RMSE (G*)	210 Pa	50 Pa
RMSE (δ)	2.8°	2°
Residual Distribution	Skewed, mild heteroscedasticity	Near-normal, no bias
Interpretability	High (linear coefficients)	Moderate (feature importance)
Nonlinear Capture	Limited	Strong
Robustness	Moderate	High
Practical Use	Quick, transparent assessments	Advanced design and optimization

Despite ANN's superior performance, MLR retains value for its simplicity and interpretability. Linear coefficients in MLR provide transparent insights into the influence of individual features, making it useful for quick assessments and routine quality checks. However, ANN offers substantial advantages for advanced binder design and performance grading, capturing nonlinear stiffening trends and delivering physically interpretable predictions through feature importance analysis. Its consistent performance across cross-validation folds, aided by early stopping and validation monitoring, ensures robustness and generalization. Overall, while MLR remains a practical tool for preliminary analysis, ANN is better suited for performance-based specification and optimization of graphene-modified bitumen, aligning with AASHTO T315 and ASTM D7175 standards for precise evaluation of complex modulus and phase angle.

From a physical standpoint, the strong influence of temperature and GNP content observed in both MLR and ANN models is consistent with the experimental findings and existing literature. Temperature primarily governs binder softening behavior, while the incorporation of graphene nanoplatelets enhances stiffness and elastic response through nanoparticle reinforcement and improved internal structure of the binder matrix. This consistency between data-driven models and physical behavior confirms the reliability of the developed predictive framework for evaluating rheological properties of modified binders.

5 Conclusion

This study examined the influence of graphene nanoplatelet (GNP) modification on bitumen rheology and evaluated the predictive capability of data-driven models for key rheological parameters. Experimental results demonstrated that GNP incorporation produced a consistent reduction in penetration values, accompanied by increases in softening and flash points, collectively indicating enhanced binder stiffness and thermal stability. A progressive decrease in ductility with increasing GNP content was also observed, reflecting an inherent trade-off between improved high-temperature performance and reduced low-temperature flexibility.

Rheological characterization using a Dynamic Shear Rheometer (DSR) confirmed that GNP-modified binders exhibited higher complex shear modulus (G*) values and lower phase angle (δ) values relative to the unmodified base binder, indicating improved resistance to permanent deformation and a more

pronounced elastic response. The most significant performance improvements were recorded at moderate GNP contents of approximately 2% to 3%, beyond which further increases in dosage yielded diminishing gains. Performance Grade (PG) analysis further confirmed that GNP modification effectively extended the binder's high-temperature grading, supporting its suitability for pavements exposed to hot climatic conditions and heavy traffic loading. When conventional and rheological properties are considered together, GNP contents in the 2% to 3% range deliver the most balanced and favorable overall performance.

In addition to the experimental investigation, predictive modeling using Multiple Linear Regression (MLR) and Artificial Neural Network (ANN) approaches demonstrated that both methods can estimate rheological parameters G^* and δ from conventional binder properties and GNP content. The MLR model provided valuable interpretability by identifying the dominant linear trends governing binder behavior. In contrast, the ANN model achieved superior predictive accuracy by capturing the nonlinear interactions among input variables. These complementary modeling approaches deepen the understanding of factors influencing the rheological response of GNP-modified bitumen and provide an efficient framework for performance prediction.

Taken together, the findings confirm that graphene nanoplatelets are a promising modifier for improving bitumen's high-temperature performance when applied at moderate dosages. The integrated experimental and machine-learning framework developed in this study offers a practical, data-driven pathway for optimizing nanomodified binder formulations. It supports the design of durable pavement systems, particularly for hot-climate applications.

The practical significance of this study lies in demonstrating that machine learning models can reliably predict the rheological properties of GNP-modified binders using only conventional test inputs. This capability is especially valuable in contexts where advanced rheological testing equipment is unavailable, enabling road agencies and practitioners to estimate binder performance from routine laboratory measurements. While empirical correlations for this purpose exist in the literature, they are typically linear and limited in predictive scope. The ANN framework developed here advances beyond these limitations by capturing the nonlinear interactions among GNP dosage, temperature, and binder properties, thereby yielding more accurate and generalizable predictions. In practice, such models could be integrated into pavement design software or quality control workflows, substantially reducing reliance on extensive laboratory testing programs. Future work will extend this framework to incorporate MSCR and fatigue-related parameters, thereby broadening its applicability in the context of performance-based pavement design.

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