
AN INVESTIGATION INTO THE PHYSICAL PROPERTIES OF METAKAOLIN-BASED GEOPOLYMER MODIFIED ASPHALT BINDER

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ABSTRACT

The development of sustainable, low-carbon construction materials has become a major priority in the construction industry. In response, this study evaluates the effectiveness of metakaolin-based geopolymer (MBG) as a modifier for improving the physical properties of asphalt binder. MBG was incorporated into a 60/70 penetration-grade asphalt binder at dosages ranging from 2% to 10% (by weight of binder, at 2% intervals). The effects on key physical properties including penetration, softening point, ductility, flash point, solubility, and storage stability were systematically investigated. Results showed that MBG addition produced a desirable hardening effect. Penetration decreased by an average of 17.94%, while the softening point increased by 6%, resulting in an improved penetration index. Ductility decreased by 39%, indicating reduced flexibility, whereas the flash point showed a marginal 1% increase. Solubility remained essentially unchanged, and storage stability was satisfactory across the tested range. These enhancements are attributed to strong interfacial interactions between the geopolymer particles and the asphalt matrix, leading to a stiffer and more stable binder structure. Overall, the findings demonstrate the potential of MBG as a sustainable, low-carbon modifier capable of enhancing asphalt binder performance. Based on the results, a 2% MBG dosage is recommended as the optimal level for effectively improving the physical properties without excessive compromise to ductility.

Keywords: Asphalt binder; Modification; Metakaolin; Geopolymer; Physical properties; Sustainability

1. INTRODUCTION

Several studies have demonstrated that polymer modification of asphaltic binders significantly enhances the performance of asphalt mixtures. For example, rubber-modified asphalt binders have been shown to improve toughness, low-temperature performance, and elastic properties, while ethylene-vinyl acetate (EVA) and high-density polyethylene (HDPE) reduce temperature susceptibility, and polyvinyl chloride (PVC) enhances heat resistance (Murana *et al.*, 2018; Murana *et al.*, 2020; Murana *et al.*, 2023; Abah *et al.*, 2024). In addition, polymer-modified asphalt contributes to notable environmental benefits by reducing pavement maintenance and repair frequency as well as lowering traffic noise (Hamid *et al.*, 2020). As a result, interest has increasingly shifted toward recycled polymers as modifiers. These materials offer clear environmental and economic advantages while often delivering superior performance compared to unmodified binders. Consequently, polymer modification of asphalt has become a highly promising approach for designing high-performance flexible pavements, with important implications for the engineering properties and long-term durability of road infrastructure.

Building on these developments, geopolymers have emerged as a promising alternative to conventional and recycled polymers for asphalt modification. Geopolymers are inorganic materials capable of delivering higher strength and superior performance in asphalt mixtures (Milad *et al.*, 2021). They are produced through geopolymerization, a reaction between alkaline activators and

aluminosilicate-rich precursors. A wide range of industrial by-products and wastes can serve as source materials, including metakaolin (Ibedu *et al.*, 2026), fly ash (Hamid *et al.*, 2020), bentonite (Jaimes *et al.*, 2020), rice husk ash (Thirukumaran *et al.*, 2023), mine waste, and blast furnace slag (Milad *et al.*, 2021; Amran *et al.*, 2021). Due to their low-CO₂ production process and utilization of waste materials that would otherwise require disposal, geopolymers are widely recognized as a greener, more environmentally friendly alternative to traditional binders and polymers. Their production significantly reduces greenhouse gas emissions compared to conventional materials, while also conserving natural resources and mitigating the environmental burdens of waste accumulation. This combination of technical performance, economic viability, and sustainability makes geopolymers highly attractive for advancing sustainable development in civil engineering. They hold strong potential for applications in asphalt concrete, pavement engineering, geotechnical engineering, and broader construction works.

However, a notable theoretical and experimental knowledge gap remains regarding the influence of metakaolin-based geopolymers on the physical properties of asphalt binders. The present study seeks to address this gap through a systematic evaluation of the physical performance of metakaolin-based geopolymer-modified asphalt binders. Such an assessment is essential to determine their suitability as a sustainable alternative to traditional bituminous binders. Bridging this gap will not only support the development of greener pavements but also provide deeper insights into the material's behavior under real-world service conditions.

2. METHODOLOGY

2.1 Materials

Metakaolin ($\text{SiO}_2 \cdot 2\text{Al}_2\text{O}_3$) was processed from Kankara kaolin. The metakaolin was used as precursor. The Alkali (Sodium Hydroxide and Sodium Silicate) were sourced from Cardinal Scientific Shop, Samaru Zaria. The NaOH was in pellet form (98.9% purity) and the Na_2SiO_3 was used for the alkali activation. The 60/70 asphalt binder was sourced at Gradient Emulsions and Polymers Limited, Located at kilometer 8, Abuja-Kaduna Expressway, Kakau Chikun LGA Kaduna State.

2.1.1 Geopolymer Synthesis

The geopolymer consists of metakaolin, and the alkaline activator. Due to the fact that there is no standard geopolymer mix design, the optimal molarity and mix ratio was adopted from Hamid, *et al.*, (2020) and further corroborated in the studies of Hauruna, *et al.*, (2018); Gambo, *et al.*, (2020). Sodium hydroxide (NaOH) pellet was diluted in water to produce 8 Molar (8M) NaOH solution and combined with the alkaline activator solution (Na_2SiO_3). The ratio of sodium hydroxide to sodium silicate ($\text{NaOH} / \text{Na}_2\text{SiO}_3$) was 2:1. A mixture of the sodium hydroxide solution and sodium silicate solution was prepared and stirred for 5 minutes to activate the aluminosilicate precursors in metakaolin. In this study, metakaolin with Class F which satisfies the chemical composition of $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ as stipulated by ASTM C168-19, (2019). The formed homogenous slurry geopolymer were poured into a size 75 cm x 75cm x 75 cm mold, the product was called metakaolin-based geopolymer. All samples were cured at room temperature for 24 hours and then further cured in the oven at 60°C for 24 hours. Generally, the process of geopolymerisation depends on the curing mode. The samples were cured at room temperature (25°C-26°C), to minimize moisture loss, allow for proper gelation. At higher temperature in the oven, the excess water is given out during this process, the reaction of geopolymerisation accelerates leading to faster setting and hardening. This improves the mechanical properties of geopolymer such as strength and durability. Plate I shows the mixing process to produce the geopolymer.



Plate I: Geopolymerisation process: (a) mixing at room temperatures (b) geopolymer paste (c) geopolymer cubes

2.1.2 Production of Metakaolin-Based Geopolymer Modified Asphalt Binder

To produce the metakaolin-based geopolymer modified asphalt binder, a two-stage mixing process was employed using high shear and low shear mixers. Initially, 3000g of pristine asphalt binder was heated to 180°C for 4 hours in an oven to achieve optimal fluidity. Subsequently, metakaolin-based geopolymer was added to the heated asphalt binder at varying concentrations (2, 4, 6, 8, and 10% by weight of bitumen). The mixture was then blended using a high shear mixer (Model Fluko SK560) at 4000 r/min for 30 minutes at 180°C. To ensure homogeneity, the blended asphalt was further mixed using a low shear mixer (Model Fluko FW30 electric stirrer) at 2000 r/min for 40 minutes at 180°C. The detailed mixing process is illustrated in Plate II.



Plate II: Production of metakaolin-based geopolymer modified asphalt binder (a) high shear mixing (b) low shear mixing

2.2 Methods

Test on Asphalt Binder

i. Fourier Transform Infra-Red (FTI-R) Spectroscopy

FTI-R spectroscopy was used to identify functional groups of metakaolin and metakaolin-based geopolymer. The wavelength were recorded from 4000 - 650 cm^{-1} . This was carried out in accordance with (ASTM D6348-12, 2020)

ii. Penetration

The penetration grade is a measure of the asphalt binder's consistency and stiffness, which correlates with its viscosity. This was conducted in accordance with ASTM standard (ASTM D5/D5M-20, 2020).

iii. Ductility

Ductility, a critical property of asphalt binders, refers to their ability to deform under tensile stress without cracking, reflecting the material's internal cohesion. The test was conducted following the guidelines outlined in the relevant ASTM standard (ASTM D113-17, 2017).

iv. Softening Point

Softening point of asphalt binders is used to grade asphalt binder and ensure performance in road construction. This was carried out in accordance with ASTM standard (ASTM D36/D36M-14, 2020).

v. Solubility

The solubility of asphalt binder was performed to determine the extent of asphalt binder solubility, having little or no organic impurities other than recovered asphalt binders in a specific solvent. For this study, Toluene was used as the solvent. This test was conducted in accordance with relevant ASTM standard (ASTM D2042-15, 2015).

vi. Flash point

The flash point of an asphalt binder is defined as the lowest temperature at which its vapors, when heated under specified conditions, will momentarily ignite (flash) upon application of a small test flame. The flash point testing was performed in accordance with the relevant ASTM standard (ASTM D92-18, 2018).

vii. Storage stability

The storage stability test measure or what extent a polymer stays mixed with asphalt binder under static storage conditions. This was conducted per ASTM Standard (ASTM D7173, 2020). Storage stability process is depicted in Plate 7.

3. RESULTS AND DISCUSSION

3.1 FTIR Spectra of Pristine Asphalt Binder

The FTIR spectrum of the pristine asphalt binder is shown in Figure 1. The major peaks characteristic of typical asphalt binders include: 2920.4 cm^{-1} and 2851.4 cm^{-1} , attributed to C–H stretching vibrations of aliphatic methylene (CH_2) groups. 1653.1 cm^{-1} , corresponding to C=C stretching of aromatic rings. 1459.3 cm^{-1} and 1375 cm^{-1} , assigned to C–H bending vibrations of aliphatic (CH_2 and CH_3) groups. In the fingerprint region, peaks at 864.7 cm^{-1} , 810.7 cm^{-1} , and 743.6 cm^{-1} indicate out-of-plane C–H bending of aromatic rings. These assignments align well with established literature on pristine asphalt binder characterization including studies by Weigel & Stephan, (2018); Yaro, *et al.*, (2021). Additionally, several less common peaks were observed, suggesting possible variations due to the specific source or crude oil origin of the asphalt binder: Weak bands at 2376.2 cm^{-1} and 2344.5 cm^{-1} , likely corresponding to S–H stretching (thiols). A peak at 2084.7 cm^{-1} , assignable to –N=C=S stretching of isothiocyanate groups (–NCS). Bands at 1597.2 cm^{-1} , 1541.3 cm^{-1} , and 1507.7 cm^{-1} , linked to –N=N– stretching in open-chain azo compounds connected to acyclic hydrocarbyl (alkyl or aryl) groups. A peak at 1032.5 cm^{-1} , attributed to P–O–C stretching of aliphatic phosphates. A band at 723.1 cm^{-1} , characteristic of out-of-plane aromatic C–H bending (Coates, 2000).

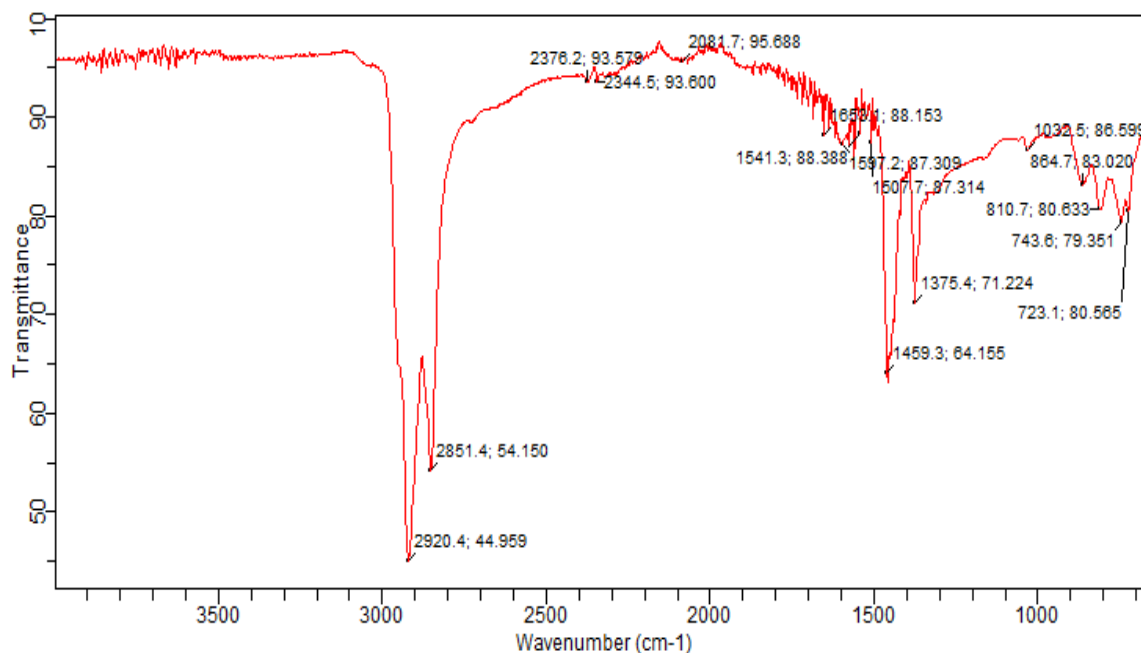


Figure 1: Infrared Spectra of Pristine Asphalt Binder

3.2 FTIR on Metakaolin-Based Geopolymer Asphalt Binder

Characteristic absorption bands at 2920.4 cm^{-1} and 2851.4 cm^{-1} , attributed to C–H stretching vibrations of methylene (CH_2 -) groups in saturated aliphatic chains (Silverstein, *et al.*, 2014), remained unchanged across all metakaolin-based geopolymer contents. Similarly, peaks at 1459.3 cm^{-1} , 1375.4 cm^{-1} , and 721.2 cm^{-1} , corresponding to C–H bending and rocking modes of aromatic rings, showed no significant shift, confirming the persistence of aromatic structures typically present in base asphalt binder (Lin-Vien, *et al.*, 1991; Coates, 2000). Weak bands observed at 2374.3 cm^{-1} and 2344.5 cm^{-1} are attributed to atmospheric CO_2 interference and are not indicative of sample composition (Smith, 2011). Several new or shifted absorption bands appeared with increasing geopolymer content, indicating chemical interaction between the metakaolin-based geopolymer and the asphalt binder. New peaks in the $2090\text{--}2165\text{ cm}^{-1}$ region (e.g., 2111.5 , 2145.1 , 2104.1 , 2161.5 , and 2092.1 cm^{-1}) suggest the formation of terminal and internal alkyne ($\text{C}\equiv\text{C}$) groups. The band originally at 1601.7 cm^{-1} in the neat asphalt binder shifted to higher (1658.1 and 1654.9 cm^{-1}) and lower (1541.3 cm^{-1}) wavenumbers, consistent with N–H bending in primary and secondary amines (Silverstein, *et al.*, 2014). A new band at 1155.5 cm^{-1} indicates C–N stretching of tertiary amines. Bands near $1994\text{--}1997\text{ cm}^{-1}$ may arise from transition metal carbonyls or cumulene ($\text{C}=\text{C}$) structures (Coates, 2000). Peaks in the $1000\text{--}1030\text{ cm}^{-1}$ region (1030.6 , 1006.4 , 1004.5 , and 1020.6 cm^{-1}) are assigned to C–H vibrations in cyclohexane ring structures or methyne groups. Additional aromatic C=C stretching bands emerged at 1597.2 , 1522.6 , and 1507.7 cm^{-1} (Silverstein, *et al.*, 2014). In the fingerprint region ($650\text{--}900\text{ cm}^{-1}$), multiple new peaks (864.7 , 862.9 , 810.7 , 808.8 , 807.0 , 745.5 , 743.6 , 659.7 , and 657.9 cm^{-1}) correspond to aromatic C–H out-of-plane bending vibrations (Lin-Vien, *et al.*, 1991). These observations demonstrate that incorporation of metakaolin-based geopolymer induces chemical modifications in the asphalt binder, resulting in the formation of new functional groups (particularly amines, alkynes, and additional aromatic structures) while core aliphatic and aromatic hydrocarbon features of the asphalt are largely retained. The FTIR spectra of metakaolin-based geopolymer asphalt binders (2–10% by weight of asphalt binder) in the range of $400\text{--}4000\text{ cm}^{-1}$ are presented in Figure 2(a–e),

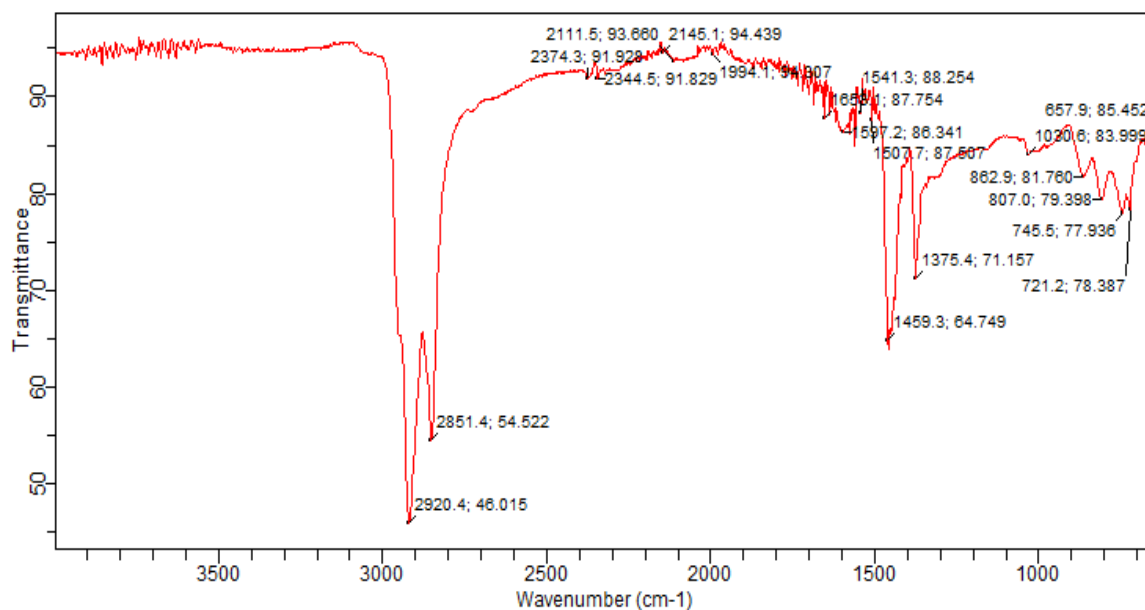


Figure 2a: Infrared Spectra of 2% Metakaolin-Based Geopolymer Asphalt Binder

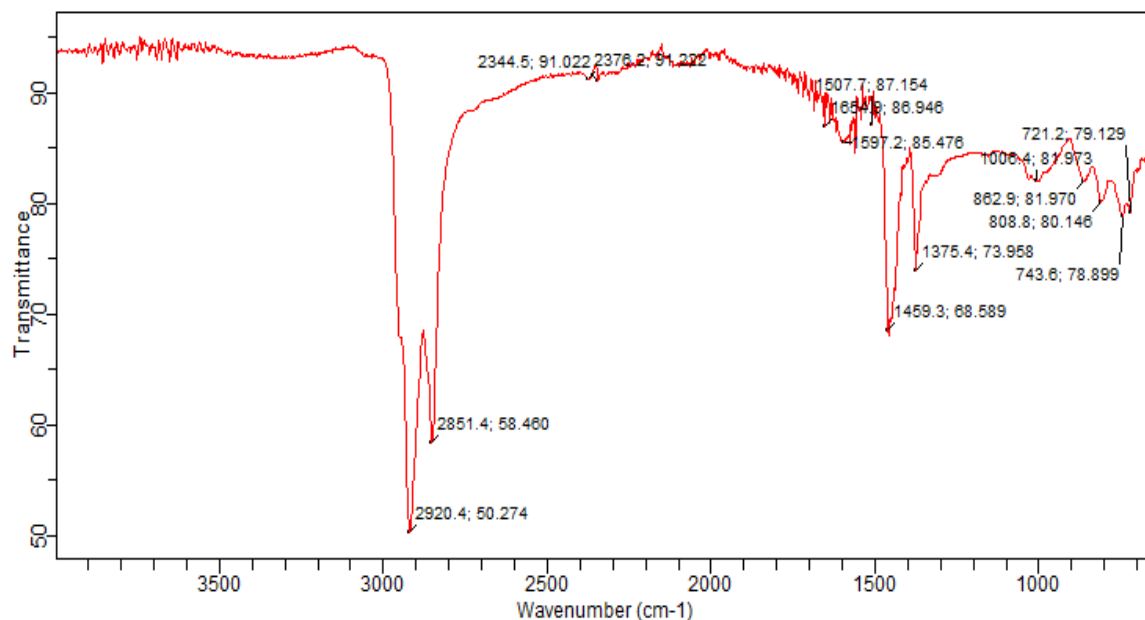


Figure 2b: Infrared Spectra of 4% Metakaolin-Based Geopolymer Asphalt Binder

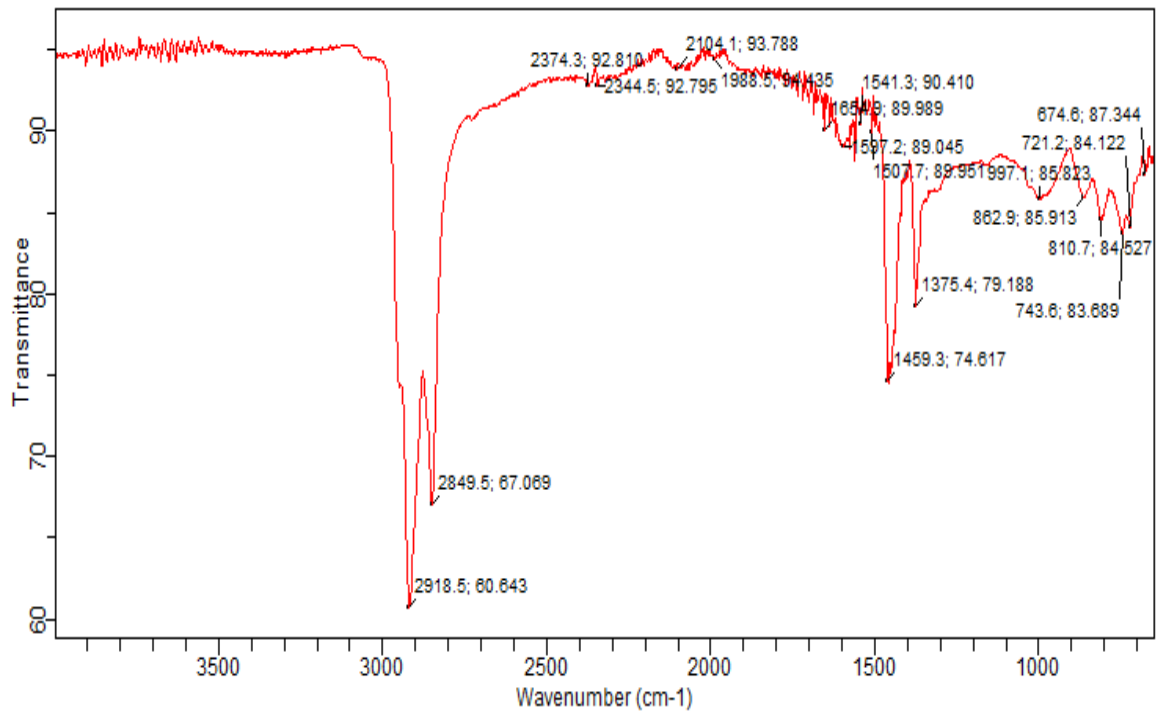


Figure 2c: Infrared Spectra of 6% Metakaolin-Based Geopolymer Asphalt Binder

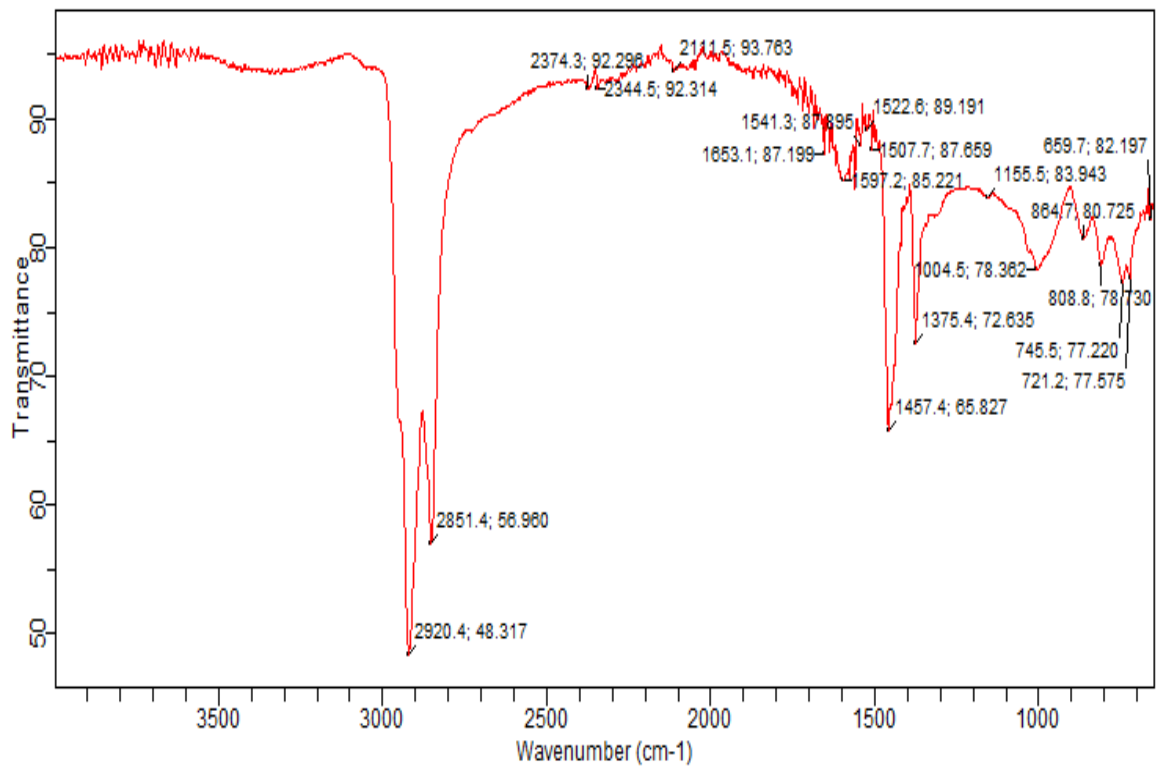


Figure 2d: Infrared Spectra of 8% Metakaolin-Based Geopolymer Asphalt Binder

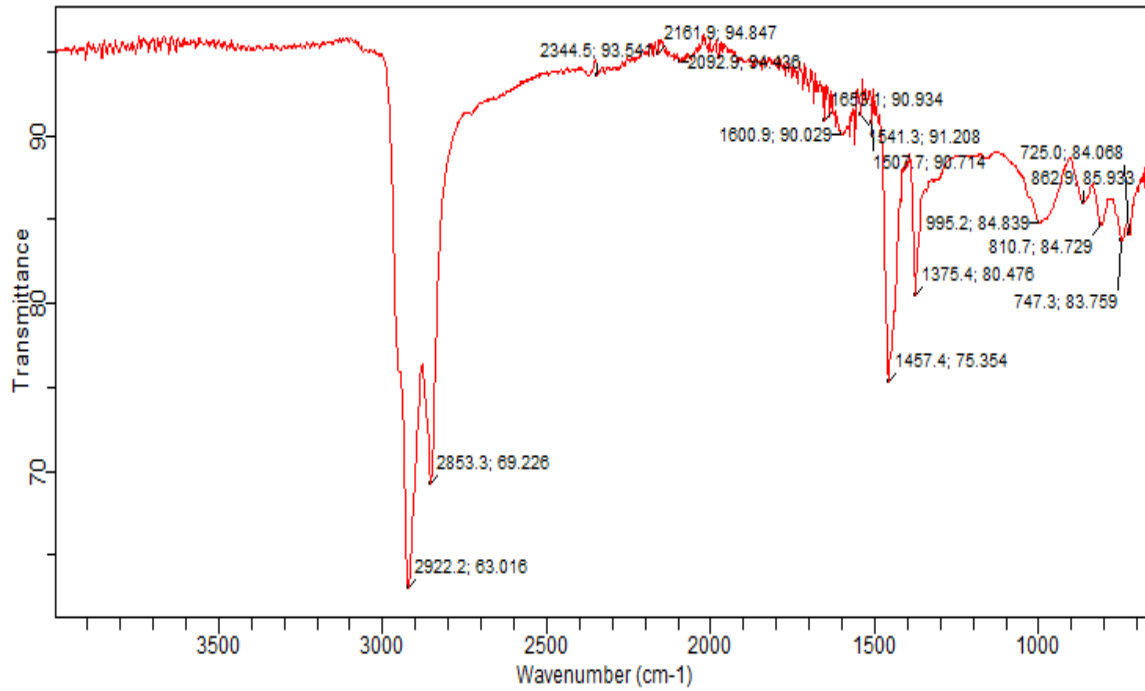


Figure 2e: Infrared Spectra of 10% Metakaolin-Based Geopolymer Asphalt Binder

3.3 Influence of Metakaolin-Based Geopolymer on the Properties of Asphalt Binder

3.3.1 Penetration

The results presented in Figure 4 demonstrate that increasing the content of metakaolin-based geopolymer (MBG) in the asphalt binder leads to a progressive decrease in penetration. This hardening effect can be primarily attributed to the strong interactions between the geopolymer particles and the asphaltene fraction of the binder, coupled with the absorption of lighter molecular components onto the geopolymer porous structure. All penetration values obtained for both pristine and MBG-modified asphalt binder fall within the penetration limits specified by (FMWH, 2016). These mechanisms reduce the mobility of the binder matrix, resulting in enhanced rigidity and stiffness, this is corroborated in the FTIR analysis by the appearance of extra aliphatic and aromatic bands in the infrared spectrum of modified asphalt binder. These findings align with previous studies on geopolymer-modified asphalt binders, including those by Yaro, *et al.*, (2023); Rosyidi, *et al.*, (2020); Tang, *et al.*, (2018). The outcome of this finding shows that MBG can effectively be used to modify conventional asphalt binder to lower penetration grades. The engineering implication of reduced penetration are enhanced asphalt binder's resistance to permanent deformation (rutting) and thermal susceptibility of asphalt mixtures produced with such asphalt binders, particularly in high-temperature climates, such as those prevalent in Nigeria. Reduced penetration also implies, reduced moisture infiltration, improved skid resistance and overall longer lifespan and durability of pavement.

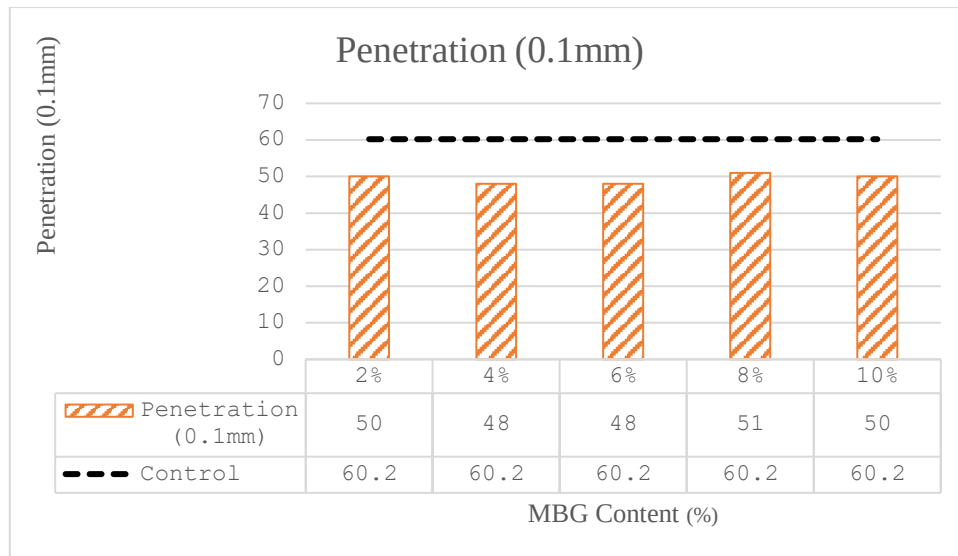


Figure 4: Influence of MBG on Penetration of Asphalt Binder

3.3.2 Softening Point

The results illustrated in Figure 5 indicate that incorporating MBG into the unmodified asphalt binder results in a significant increase in the softening point compared to the control. This trend shows the typical inverse relationship between softening point and penetration: lower penetration yields a higher softening point. All softening point values obtained are within the limits stipulated by FMWH (2016). The increased softening point can be attributed to the stiffening mechanisms induced by the MBG modifier. The geopolymer particles interact with the binder's components particularly by bonding with the aliphatic fractions leading to enhanced dispersion stability, reduced molecular mobility, and the formation of additional aromatic and aliphatic structures within the matrix. These observations are in agreement with prior research on geopolymer-modified asphalt binders, studies include those by Yaro, *et al.*, (2023); Rosyidi, *et al.*, (2020); Tang, *et al.*, (2018). From an engineering perspective, the higher softening point enhances the binder's resistance to permanent deformation (rutting) under elevated service temperatures.

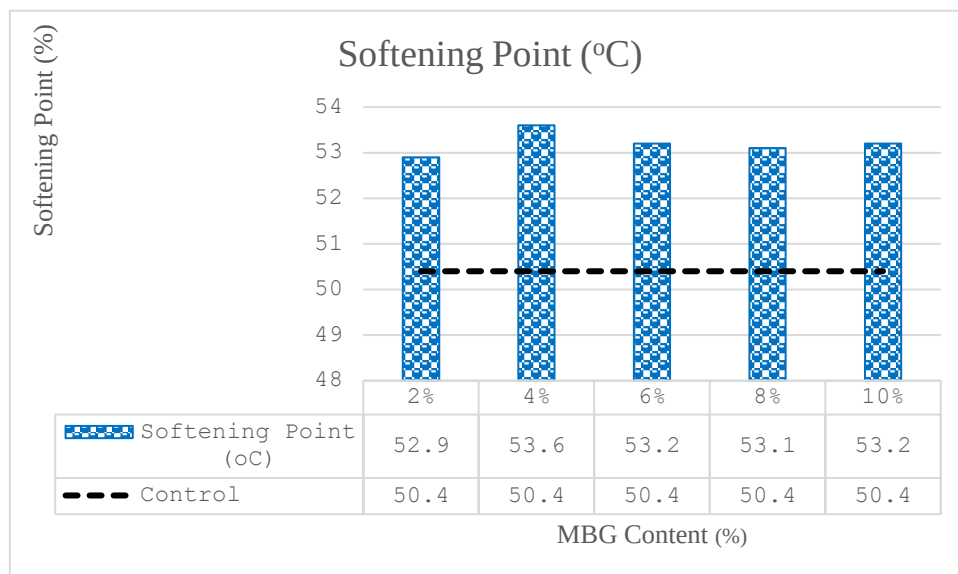


Figure 5: Influence of MBG on Softening Point of Asphalt Binder

3.3.3 Penetration Index

The penetration indices (PI) of the pristine (0%) and MBG-modified asphalt binders are presented in Table 1. The penetration index serves as a key indicator of the binder's temperature susceptibility. For conventional paving-grade asphalt binders, the acceptable PI range is typically -2 to $+2$. A PI of < -2 signifies high temperature susceptibility, while a PI of $> +2$ indicates low susceptibility. The calculated PI values decreased from the 0% binder to the MBG-modified binders. The observed reduction in PI for MBG-modified asphalt binder suggests improved overall temperature resistance. The MBG enhanced the binder's ability to resist both rutting (at high temperatures) and thermal cracking (at low temperatures). This improvement in temperature susceptibility can be attributed to the intermolecular interactions between dispersed MBG particles and the asphalt matrix, which stiffen the binder structure, reduce thermal volatility, and improve rheological property. These mechanisms are further supported by the FTIR results. From an engineering implication, the enhanced temperature performance of MBG modified asphalt binders is suitable for use in hot climate regions. This result is in align with the study by Yaro, *et al.*, (2021).

Table 1: Penetration indices of Pristine and MBG-modified asphalt binder

MBG Content	Penetration	Softening Point	Penetration index
0%	60.2	50.4	-0.666
2%	50	52.9	-0.574
4%	48	53.6	-0.439
6%	48	53.6	-0.532
8%	51	53.2	-0.413
10%	50	53.2	-0.436

3.3.4 Ductility

The results in Figure 6 indicate a decrease in ductility of MBG asphalt binder compared to the control. This reduction is linked to the increased stiffness observed in the penetration test, likely due to interactions between resins (carbonyl) and aromatic functional groups within the internal structure of the molecules. Only the control and the 2% MBG-modified asphalt binder satisfied the minimum 100 mm requirement specified by FMWH (2016). The higher dosages of MBG resulted in reduced ductility values that fell below the specified limit. However, despite the reduction in ductility with higher MBG dosages, the results suggest that the modified binders retain adequate adhesive and tensile properties. These characteristics are crucial for proper bonding between the asphalt binder and aggregates, indicating promising performance of the asphalt mixtures. This is consistent with findings from Yaro, *et al.*, (2023); Rosyidi, *et al.*, (2020); Tang, *et al.*, (2018).

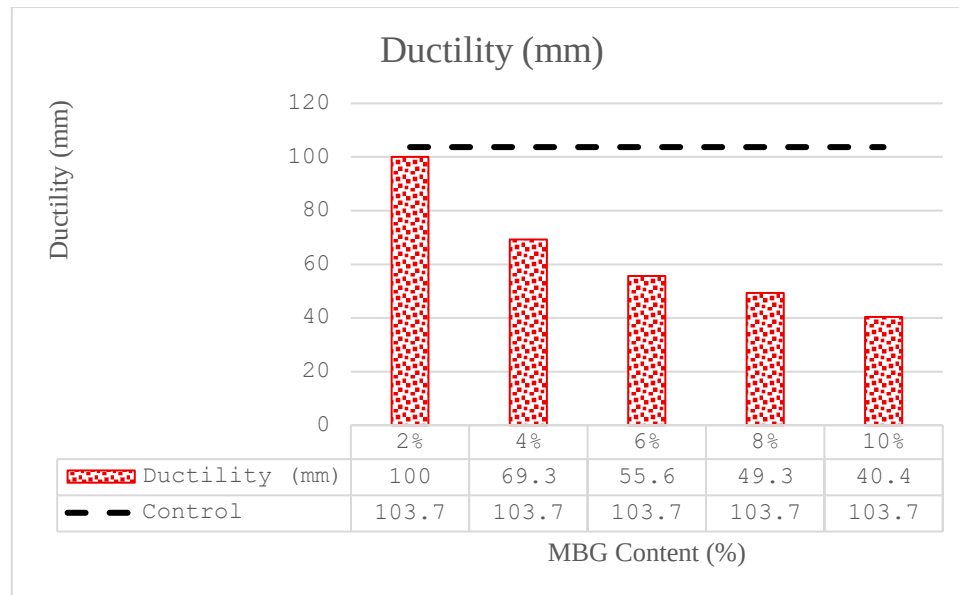


Figure 6: Influence of MBG on Ductility of Asphalt Binder

3.3.5 Flash point

The results from Figure 7 show that the flash point of the MBG-modified asphalt binder marginal increased by 1% with respect to the control binder. Nevertheless, the flash points of all MBG asphalt binders remain compliant the minimum requirement of 230 °C specified in AASHTO M 320 and higher than 250 °C by FMWH, (2016). The observed increase in flash point temperature for MBG-modified binders can be attributed to the dispersion of MBG particles within the asphalt matrix. These particles enhance the binder's thermal stability, thereby delaying the onset of flammability by reducing the volatility of lighter fractions and improving overall resistance to ignition. These findings suggest that MBG-modified asphalt binders exhibit reduced flammability compared to the control, offering a safety advantage. From an engineering perspective, a higher flash point enhances operational safety: the binder can be handled, stored, and transported at elevated temperatures with a significantly lower risk of fire hazards during production, mixing, or field application.

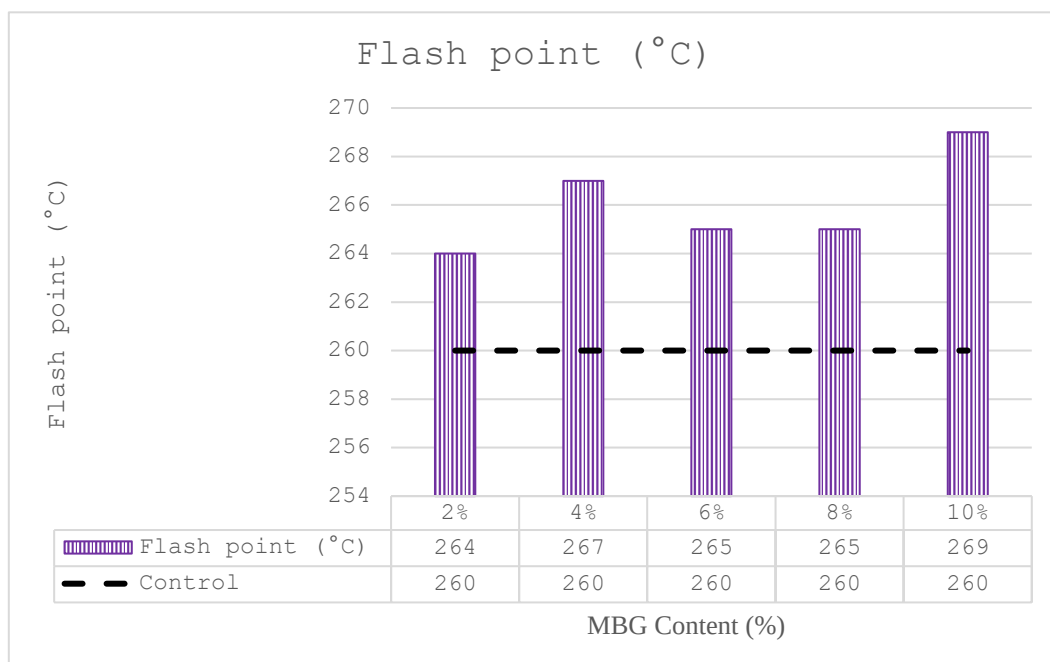


Figure 7: Influence of MBG on Flash Point of Asphalt Binder

3.3.6 Solubility

Solubility serves as an important indicator of asphalt binder purity. As shown in Figure 8, the solubility of the MBG-modified asphalt binders was lower than that of the control sample and decreased progressively with increasing MBG content. The control, 2 - 6% MBG-modified asphalt binder satisfied the minimum 99% requirement specified by FMWH (2016). The higher dosages of MBG fell below the specified limit. This reduction suggests the presence of minor insoluble organic impurities introduced by the modifier. Supporting evidence from FTIR analyses of metakaolin, as well as the FTIR spectra of the metakaolin-based geopolymer, confirmed the existence of such impurities. However, these impurities are present in minute quantities and are expected to have negligible effects on the overall performance of the asphalt binder. Therefore, their impact can be reasonably disregarded

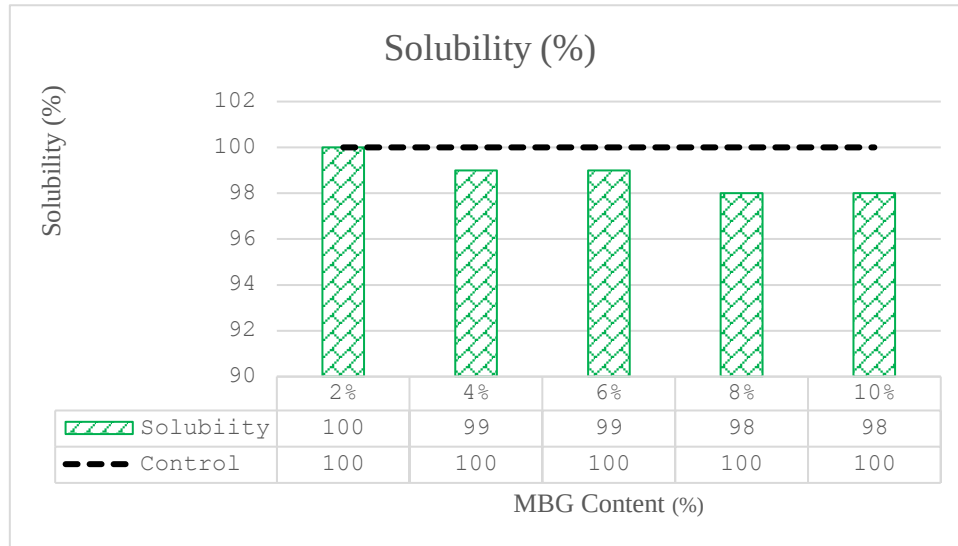


Figure 8: Influence of MBG on Solubility of Asphalt Binder

3.3.7 Storage Stability

The storage stability (also referred to as phase separation resistance or compatibility) of the asphalt binder was evaluated by measuring the difference in softening point between the top and bottom sections of a vertically stored sample tube after high-temperature conditioning. The results, as presented in Figure 9 showed that the storage stability for the pristine (0% MBG) asphalt binder, as well as the MBG modified asphalt binders at 2, 6, 8, and 10% dosages, remained below 2.5 °C (code acceptance criterion). This storage stability demonstrates excellent compatibility between the pristine asphalt binder and the MBG modifier, with negligible phase separation. Such performance is critical for preventing segregation during hot storage, transportation, and application, ensuring uniform binder properties in the field. From an engineering standpoint, this high temperature storage stability enhances operational reliability, reduces the risk of inconsistent binder behavior during mixing and paving, and supports practical handling in asphalt plants. Consequently, both the pristine and MBG-modified asphalt binders can be stored at elevated temperatures for extended periods, reheated, and reused without significant loss of homogeneity or performance. A similar trend of good storage stability with geopolymer based modifiers in asphalt was reported by Yaro *et al.*, (2021).

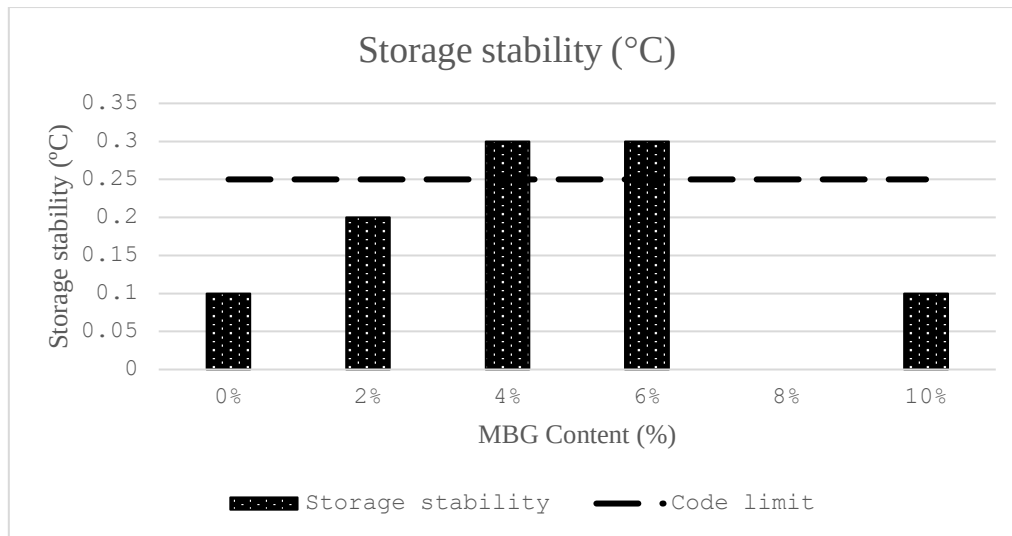


Figure 9: Storage stability of Asphalt Binders

4. CONCLUSION

This study presents the investigation on the physical properties of metakaolin-based geopolymer modified asphalt binder. The following conclusions were drawn based on the results of the research.

- i. FTIR spectroscopy of the pristine asphalt binder revealed the presence of characteristic functional groups, including C–H stretching (aliphatic methylene), C=C stretching (aromatic rings). Less prominent peaks corresponding to –N=N– (alkyl or aryl), S–H (isothiocyanate), and P–O–C (alkyl) were also identified. Upon incorporation of metakaolin-based geopolymer (MBG), the original functional groups in the asphalt binder remained largely unchanged, while new peaks emerged, notably C≡C (internal alkyne), additional C=C (associated with metal carbonyls or cumulene), and C–H (cyclohexane or methine structures). These spectral changes confirm the successful incorporation of MBG into the asphalt binder and indicate the formation of new chemical interactions. The resulting functional groups influenced the physical properties of the modified asphalt binder
- ii. The physical properties of the MBG-modified asphalt binders, when compared to the pristine binder, exhibited a typical pattern of effective stiffening modifications: decreased penetration, reduced ductility, and slightly lower solubility, accompanied by increases in softening point and flash point

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