

## Research Article

# Effect of Multiple-Walled Carbon Nanotubes (MWCNTs) on Asphalt Binder Rheological Properties and Performance

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Growing traffic loads, soaring summer temperatures, and moisture damage will render conventional asphalt binder insufficient to maintain the performance standards of asphalt concrete pavement. Thus, it is necessary to modify the virgin asphalt using various polymers or nanomaterials. The primary goal of this research was to examine the rheological effects of combining multiple-walled carbon nanotubes (MWCNTs) and styrene butadiene styrene (SBS) in an asphalt binder. In this study, MWCNTs and SBS were mixed with virgin asphalt at concentrations of 1%, 3%, and 5% by weight. The performance grade (PG) and asphalt binder qualities were determined through Superpave system testing. The addition of 1% MWCNTs had no effect on the (PG) of virgin asphalt, whereas the addition of 3% and 5% MWCNTs resulted in increases of 2° and 4°, respectively. When 1% SBS is added to asphalt, the PG rises by an average of 1°; when 3% and 5% SBS are used, the PG rises by an average of 2° and 3°, respectively. The results also showed that the rutting parameter ( $G^*/\sin$ ) increased by 10%, 73%, and 208% when asphalt was changed with 1%, 3%, and 5% of SBS, and by 18% and 130% when MWCNTs were applied.

## 1. Introduction

Asphalt mixtures were used for highway construction for a long time, and asphalt cement is considered one of the major components [1]. The asphalt properties have a great influence on the performance of pavements [2]. It is anticipated that the highway must resist rutting at high temperatures and cracks at low temperatures [3]. Therefore, modification of asphalt cement is required to improve asphalt cement rheological properties, enhance asphalt temperature susceptibility, and improve the asphalt binder to rutting, fatigue, and stripping [4, 5].

Alhamali et al. [6] studied the influence of blending asphalt cement with 5% SBS and nanosilica at a percentage of 2%, 4%, and 6%. They found that nanosilica increase ductility, softening point, and viscosity, while it decreases penetration. Shu et al. [3], investigated the effect of mixing multiple-walled carbon nanotubes (MWCNTs) at a percentage of 0.5%, 1%, 1.5%, 2%, and 3% with 3.5% SBS. They concluded that MWCNTs increases rutting parameter

( $G^*/\sin \delta$ ) as the percentage of MWCNTs content increased. Faramarzi et al. [7], studied the effect of modifying asphalt binder with 0.1%, 0.5% and 1% of carbon nanotube (CNT). They showed that adding CNT work to increase softening point and decrease both ductility and penetration. The results obtained by Zheng et al. [8], depicted that adding CNTs to asphalt modified with SBS can increase rutting parameter and this parameter is increased as the percentage of CNTs increased.

Yan et al. [9], observed that modifying asphalt binder with waste tire rubber (WTR) and reclaimed low-density polyethylene (RPE) can increase the complex shear modulus and decrease the magnitude of phase angle comparing with virgin asphalt cement. Zafari et al. [10], evaluated the effect of adding nanosilica as a modifier to the base asphalt binder. They concluded that nanosilica may be used as an antiaging modifiers. Galooyak et al. [11], studied the effect of nanosilica on asphalt binder rheological properties. The results demonstrated that the modified asphalt has higher rutting resistance than the virgin asphalt. They also concluded that

TABLE 1: DSR test results of virgin asphalt cement, PG (64-16).

Parameter	Temperature measured	Test results	Criteria
RV, Pa s	@135°C	0.540	3 Pa s, max
Aging		Unaged binder	
G*/sin δ, kPa	@70°C	1.91	1 kPa, min
	@76°C	0.75	
Aging		Rolling thin film oven (RTFO)	
G*/sin δ, kPa	@64°C	4.39	2.2 kPa, min
	@70°C	1.72	
Aging		Pressure aging vessel (PAV)	
G*. sin δ, kPa	@28°C	4,305	5,000 kPa, max

TABLE 2: Asphalt cement physical properties.

Test	Unit	AC (40–50)	
		Test results	SCRB, 2003 criteria
Penetration (100 gm, 25°C, 5 s)	(0.1 mm)	44	40–50
Rotational viscosity at 135°C	Pa s	0.540	.....
Ductility (25°C, 5 cm/min)	cm	115	>100
Flash point (cleave land open cup)	°C	298	Min. 232
Softening point (4 ± 1) °C/min	°C	52	.....
Specific gravity	.....	1.03	1.01–1.05

the rut depth is decreased as the percentage of nanosilica increased.

Galooyak et al. [11] and Shan et al. [12] mentioned that the elastic portion and nonlinearity are increased when modifying asphalt with SBS as compared with unmodified asphalt, and this increase increases as the percentage of SBS increases. The results also show that SBS makes the asphalt binder fluidity to decrease [11, 12]. The results achieved by Zhang et al. [13], found that modifying asphalt with SBS can enhance asphalt binder performance at elevated temperatures and reduce the sensitivity of asphalt to high temperatures. Moreover; it is found that SBS increased softening point and viscosity and, as a result, improved rutting resistance of asphalt pavement. Shafabakhsh and Tanakizadeh [14] tested the influence of using SBS on asphalt binder stiffness. The results demonstrated that the stiffness of modified asphalt was approximately three times that of unmodified asphalt under a long time of loading at 40°C. ul Haq et al. [15], examined the effect of using CNTs as modifiers for asphalt binder. They concluded that CNTs enhanced the stiffness of bitumen and, consequently, increased asphalt resistance against permanent deformation. Anwar et al. [16], found that the addition of MWCNTs decreased penetration by 14.4% and increased softening point and ductility by 10.2% and 40%, respectively. Moreover; results illustrated that there was a considerable enhancement in phase angle and complex shear modulus. Słowik [17], observed that modifying asphalt cement with an SBS copolymer made a significant increase in complex shear modulus. Eisa et al. [18], studied the effect of CNTs on the mechanical properties of asphalt binder and mixtures. They concluded that CNTs decreased penetration and

increased both of softening point and viscosity. They also stated that rutting parameter increased while rut depth reduced by 45% when adding upon 0.5% CNTs.

The major goal of the current study is to examine the influence of styrene–butadiene–styrene (SBS) and MWCNTs on the rheological properties of asphalt binder.

## 2. Materials and Methods

**2.1. Asphalt Cement.** The AL Daurah refinery in Baghdad supplied the asphalt cement, which had a penetration grade of (40–50). Table 1 shows the results of dynamic shear rheometer (DSR) tests performed in accordance with AASHTO T-315 to determine asphalt binder rheological qualities at high and moderate temperatures, whereas Table 2 presented the physical properties of asphalt cement.

**2.2. Modifiers.** In this research, two kinds of modifiers were used to enhance asphalt cement rheological properties, SBS and MWCNTs.

**2.2.1. Styrene Butadiene Styrene (SBS).** SBS is one of the most extensively used elastomer polymers in asphalt binder modification. Kraton Company in France supplied the recycled SBS that was imported. The SBS employed in this research is depicted together with its physical and mechanical properties in Table 3.

**2.2.2. Multiple-Walled Carbon Nanotubes (MWCNTs).** CNTs define a family of nanomaterials made of carbon. The properties and the appearance of the MWCNTs are presented in Table 4 and Figure 1(a). Structurally MWCNTs compose of multilayers of graphite overlapped and rolled in on

TABLE 3: Physical and mechanical properties of SBS.

Bulk density (Kg/m <sup>3</sup> )	Elongation (%)	Specific gravity	Tensile strength (MPa)	Point of melting (°C)	Color
940	88	0.94	32 min	180	White

TABLE 4: Physical properties of (MWCNTs).

Purity	Inner diameter	Outer diameter	Length	Surface area	Bulk density	Appearance
90%	5–10 nm	10–30 nm	10–30 $\mu$ m	>200 m <sup>2</sup> /g	0.06 g/cm <sup>3</sup>	Black powder

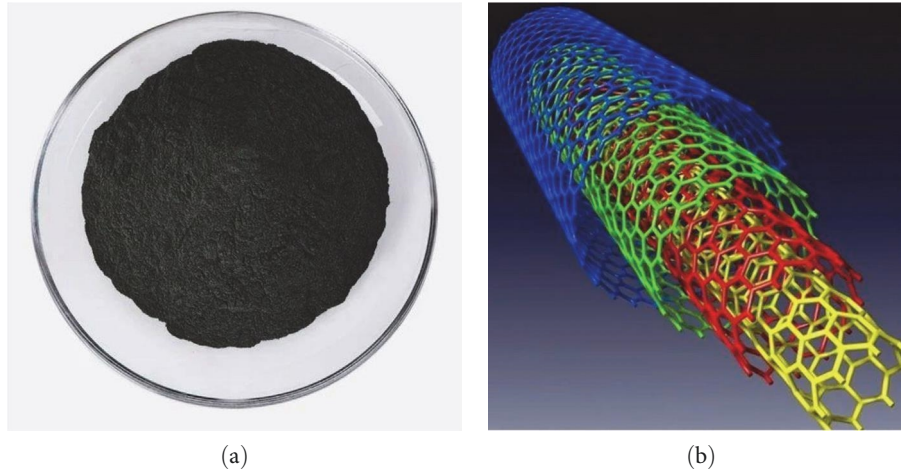


FIGURE 1: (a) Appearance of MWCNTs and (b) multilayers of graphite overlapped and rolled in on themselves to form a cylindrical shape.

themselves to form a cylindrical shape, as depicted in Figure 1(b). The used MWCTs was brought from Cheap Tubes Company in America.

### 3. Samples Preparation

In order to mix virgin asphalt with SBS, at the beginning, the virgin asphalt is heated until it becomes sufficiently fluid to pour; then 1%, 3%, and 5% of SBS by weight of asphalt are gradually added and blended by a shear mixer device at 2,220 rev/min. Furthermore, the revolution was maintained for approximately 3.5–4.0 hr while the temperature was kept at 180°C to ensure good homogeneity and compatibility [19]. In the same manner, 1%, 3%, and 5% of MWCTs by weight of asphalt were gradually added and blended by a shear mixer device at 1,550 rev/min for 40 min at 160°C [20]. It is worthwhile to mention that 28 specimens were tested to determine the effect of modifiers on asphalt binder rheological properties by using DSR and RV tests taking into account short term aging by RTFO and long term aging by conducting pressure aging vessel (PAV). All of the experimental studies were carried out at Al-Nahrain University, civil engineering department in Baghdad. Figure 2 shows the experimental flowchart.

## 4. Results and Discussion

4.1. *Effect of SBS on Asphalt Binder Rheological Properties.* It is obvious from Table 5 and Figure 3 that ( $G^*/\sin \delta$ ) for

modifying asphalt is greater than that of virgin asphalt. It is observed that ( $G^*/\sin \delta$ ) is increased by an average of 10%, 73%, and 208% when adding 1%, 3%, and 5% of SBS, respectively. Moreover, the PG of virgin asphalt is enhanced by one degree PG (70-16), two degrees PG (76-16), and three degrees PG (82-16) for asphalt blended with 1%, 3%, and 5% of SBS, respectively, as illustrated in Figure 4.

For rotational viscosity at 135°C, it is obvious from Figure 5 below that the viscosity increased by 48%, 176%, and 287% when the asphalt was mixed with 1%, 3%, and 5% of SBS, respectively. The fatigue parameter ( $G^* \cdot \sin \delta$ ) went up by 12% when asphalt was mixed with 1% SBS. However, the fatigue parameter ( $G^* \cdot \sin \delta$ ) for asphalt mixed with 3% and 5% SBS got better and met Superpave specifications at 31°C. The increase in the rotational viscosity and performance grade (PG) belongs to the characteristics of SBS. SBS has a tendency to swell through aromatic constituents from bitumen up to nine times more than its original volume so as to build a strong net between bitumen and polymer. So, in this case SBS behaves as a cross-linker and as a result, this cross-linkage increases cohesion of bitumen and also increases complex shear modulus, which leads to improve asphalt resistance to rutting [21, 22].

4.2. *Effect of MWCNTs on Asphalt Binder Rheological Properties.* Table 6 and Figure 3 show that modifying virgin asphalt with 1%, 3%, and 5% MWCNTs increases ( $G^*/\sin \delta$ ) by 18%, 130%, and 264%, respectively, when compared to

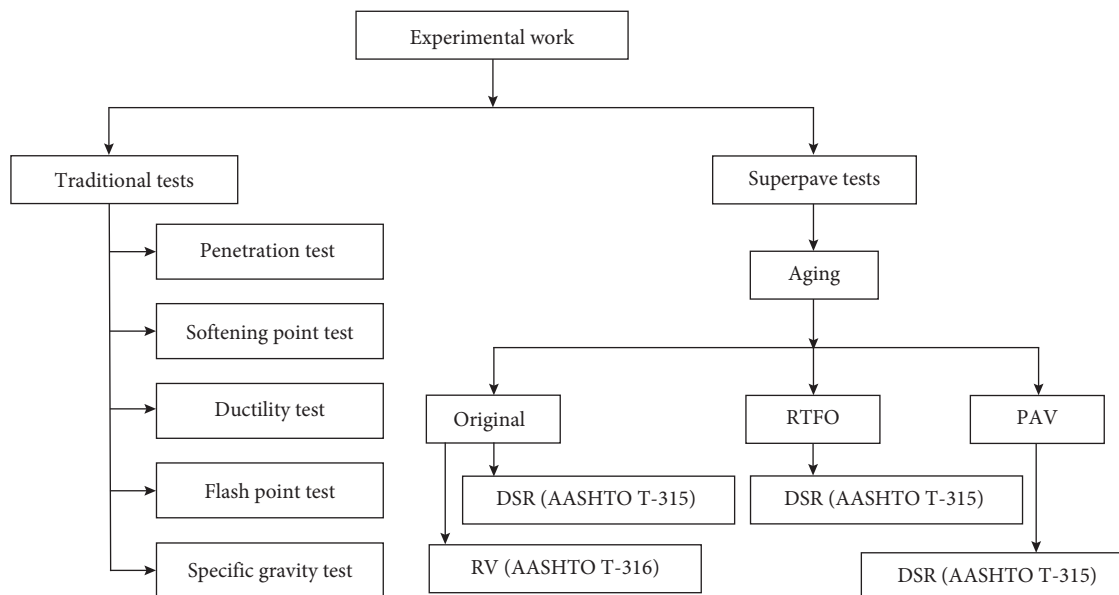


FIGURE 2: The experimental work.

TABLE 5: Test results of modified asphalt binder with SBS.

Parameter	Criteria	Standard specifications	Modified asphalt binder					
			1% SBS		3% SBS		5% SBS	
			Test results	Measured temp.	Test results	Measured temp.	Test results	Measured temp.
RV, Pa s	3 Pa s, max	(AASHTO) (T-316)	0.8	@135°C	1.49	@ 135°C	2.09	@135°C
Aging			Unaged binder					
$G^*/\sin \delta$ , kPa	1 kPa, min	(AASHTO) (T-315)	1.87	@76°C	1.77	@82°C	2.89	@82°C
			0.923	@82°C	0.88	@88°C	0.78	@88°C
Aging			Rolling thin film oven (RTFO)					
$G^*/\sin \delta$ , kPa	2.2 kPa, min	(AASHTO) (T-315)	2.98	@70°C	2.43	@76°C	3.05	@82°C
			1.25	@76°C	1.62	@82°C	2.11	@88°C
Aging			Pressure aging vessel (PAV)					
$G^* \cdot \sin \delta$ , kPa	5,000 kPa, max	(AASHTO) (T-315)	4,810	@28°C	4,210	@ 31°C	4,528	@31°C

virgin asphalt. Furthermore; PG of virgin asphalt PG (64-16) does not change with the addition of 1% MWCNTs, while it is enhanced by two degrees PG (76-16) and four degrees PG (88-16) when modifying asphalt with 3% and 5% of MWCNTs, respectively, as demonstrated in Figure 4.

For rotational viscosity, the results Figure 5 showed that viscosity was increased by 43%, 107%, and 309% when add 1%, 3%, and 5% of MWCNTs were added, respectively. Furthermore, ( $G^* \cdot \sin \delta$ ) increased by 5% for asphalt modified with 1% MWCNTs, while ( $G^* \cdot \sin \delta$ ) was enhanced and passed specifications of Superpave 31°C for asphalt modified with 3% and 5%, respectively. The increase in PG and rutting parameters can be attributed to the high-surface energy, and the existence of interaction forces among MWCNTs that made the asphalt binder stiffer [1].

## 5. Effect of SBS and MWCNTs on Pavement Performance

**5.1. Pavement Structure and Design Criteria.** The correlation between pavement deterioration and asphalt layer thickness was calculated using Mechanistic-Empirical Pavement Design Guide (MEPDG) (AASHTOWare version 2.3). Expressway No. 1-Iraq (part R4/B) was used as a case study to find the pavement performance. For MEPDG calculation, the mentioned section typically has a four-layer pavement structure consisting of 12, 15, and 20 cm, and A-7-6 for asphalt concrete, base, subbase, and subgrade (semi-infinite) layer, respectively. The level-2 design was chosen for the computation, and the traffic volume was set to 10 million ESALs. All other fields had their default values chosen for them.

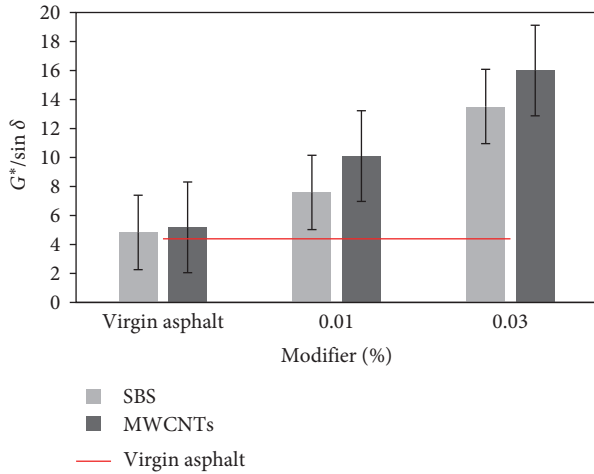


FIGURE 3: Effect of SBS and MWCNTs on rutting parameter.

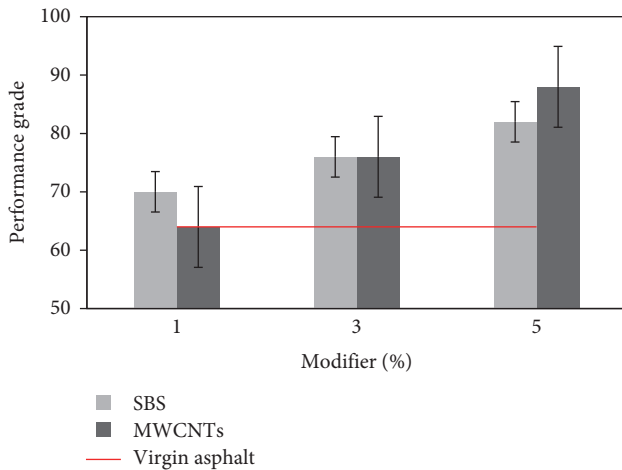


FIGURE 4: Effect of SBS and MWCNTs on performance grade.

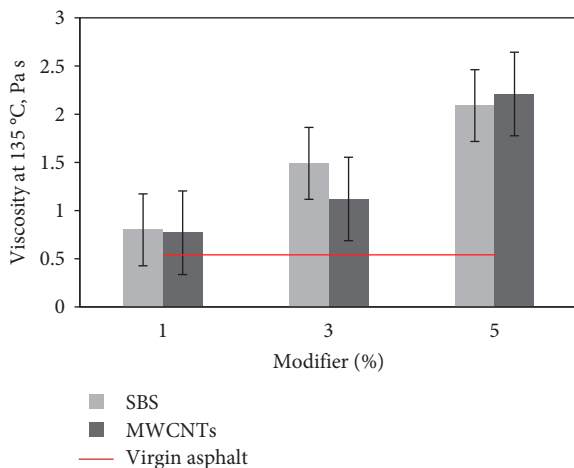


FIGURE 5: Effect of SBS and MWCNTs on viscosity.

Over a 30-year time period, the effect of modifier variation on pavement distresses was investigated to establish a baseline. Some of the most vital performance indicators included asphalt layer rutting, total pavement rutting,

top-down cracking, heat cracking, alligator (bottom-up) fatigue cracking, and top-end smoothness (international roughness index (IRI)). The current version of pavement ME's unreliable performance model means that reflected cracking is being ignored for the time being. To evaluate the effect of modifier modification on pavement thickness, we can consult Table 7, which provides the performance criterion based on the MEPDG defaults for roughness, design information, and distress limits.

Kadhim [24] previously investigated the elastic modulus of the conventional pavement section's binders and foundation courses via the laboratory indirect tensile stiffness test. Table 8 displays the results of taking the same measurements for each pavement section's wearing course and underlying layers (subbase and subgrade) using the same properties [24]. Linear elastic materials were assumed and uniform loading was applied in this investigation (single load).

Table 9 further shows that new pavements were designed using median traffic scenarios. The axle load spectra and default vehicle class distribution and were used in the study of truck traffic categorization on expressways (TTC1: bus > 2%, multitrailer 2%, predominantly single-trailer trucks) (Level 3 input). In addition, the AASHTO 1993 pavement design guide's load equivalent factor was employed to convert traffic statistics across the two-decade design period into ESALs for comparative purposes.

**5.2. Effect of Modifier on Rutting.** Thinner asphalt layers can be maintained with the use of modifiers. The AASHTOWare, ME program was used to determine the overall permanent deformation of typical pavement constructions when varying percentages of SBS and MWCNTs modifiers were applied. Figure 6 displays the MEPDG results for MWCNTs, while Figure 7 displays the results for SBS. According to Table 6 [26] of the MEPDG design guidelines, the maximum allowable permanent deformation is 1.9 cm.

Figure 6 shows that the addition of 1% MWCNTs produces the same permanent deformation as when using a virgin binder. According to the findings, both the virgin and modified versions (1% of modifier) of the pavement will have an age of 15 years. While 3% and 5% of MWCNTs modifiers, respectively, could result in an age of 17.5 and over 25 years, respectively.

Figure 7 shows that the service life of binders modified with SBS is significantly less than that of binders treated with MWCNTs, which can last for up to 13, 14, and 25 years at 1%, 3%, and 5% SBS, respectively. The results show the influence of these variables on the PG.

## 6. Effect of Modifier on Bottom-Up Cracking

Figures 8 and 9 display the results of an MEPDG calculation for bottom-up cracking based on modulus values, input of asphalt layer thickness, and all other default values stated in Tables 6 and 7. For bottom-up cracking, MEPDG currently uses a 25% threshold [26].

According to the findings of the tests, the lifetime of the construction made with virgin binder is 17 years, and the lifetime of the pavement was increased by 5 years due to the

TABLE 6: Test results of modified asphalt binder with MWCNTs.

Parameter	Criteria	Standard specifications	Modified asphalt binder					
			1% MWCNTs		3% MWCNTs		5% MWCNTs	
			Test results	Measured temp.	Test results	Measured temp.	Test results	Measured temp.
RV, Pa s	3 Pa s, max	(AASHTO) (T-316)	0.77	@135°C	1.12	@ 135°C	2.21	@135°C
Aging				Un-aged binder				
$G^*/\sin \delta$ , kPa	1 kPa, min	(AASHTO) (T-315)	1.89	@70°C	1.69	@76°C	3.43	@82°C
			0.95	@76°C	0.887	@82°C	1.01	@88°C
Aging				Rolling thin film oven (RTFO)				
$G^*/\sin \delta$ , kPa	2.2 kPa, min	(AASHTO) (T-315)	5.18	@64°C	3.651	@76°C	6.12	@82°C
			2.09	@70°C	1.13	@82°C	2.96	@88°C
Aging				Pressure aging vessel (PAV)				
$G^*/\sin \delta$ , kPa	5,000 kPa, max	(AASHTO) (T-315)	4,532	@28°C	4713	@31°C	4710	@31°C

TABLE 7: M-E pavement design criteria [23].

Design criterion	Design information		
R (%)	90	Base layer construction started	May 2023
IRI° (m/km)	1.0	Surface construction	July 2023
Terminal IRI (m/km)	2.71	Traffic opening	Sep. 2023
Top-down fatigue cracking (m/km)	379	Climate station	(Taxes, station 133,191)*
AC bottom-up fatigue cracking (%)	25		
Thermal cracking (m/km)	189	Assumed ESAL	10 million
Rutting (AC only) (cm)	0.63	AADT (veh./day)	3,000
Rutting (total) (cm)	1.9		

Due of its proximity to Iraq, the researchers chose Taxes as their climatic station.

TABLE 8: HMA properties used for thickness reduction estimation.

Layer	Thickness (cm)	Poisson's ratio	Elastic modulus (MPa)	References
Wearing course	4	0.35	1194	[25]
Binder and base (stabilizer) courses	8/15	0.35	2609	[24]
Subbase course (granular material)	20	0.4	104	[25]
Subgrade	–	0.5	40	[25]

TABLE 9: Data related to traffic volume.

	Medium traffic level
ESAL	10 million
AADTT (veh./day)	3,000
Design speed (V) (km/h)	80
Truck in design direction (D) (%)	50
Truck in design lane (%)	95
Growth rate (G%)	3

addition of 1% of the MWCNTs modifier. The pavement's durability also improves when more modifier is added to the mix.

Neither MWCNT- nor SBS-modified pavements experience significantly shorter service lives due to bottom-up

cracking. Pavements built using stiff binders, including MWCNTs and SBS, exhibit less bottom-up cracking than those built with virgin binders, according to the software results.

## 7. Effect of Modifier on International Roughness Index (IRI)

To account for the cumulative effects of distresses such as cracking, rutting, faulting, and punchouts on the loss of pavement smoothness during the whole design period, IRI is computed incrementally in MEPDG [27–29].

The predicted values of IRI for MWCNTs and SBS are shown in Figures 10 and 11, respectively. The pavement service life for MWCNTs modification reaches the threshold

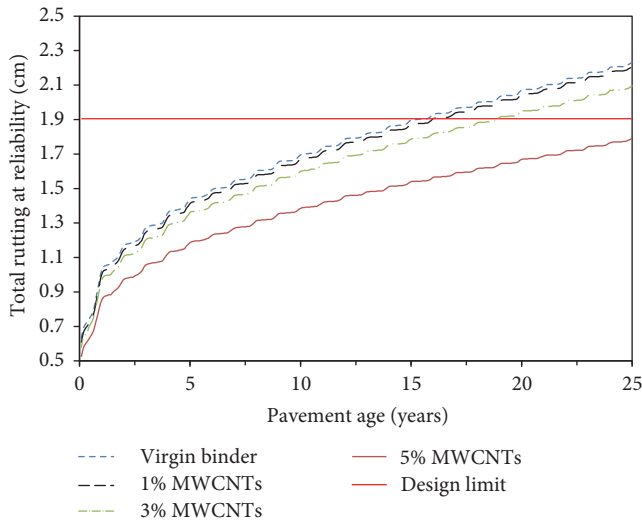


FIGURE 6: Effect of MWCNTs percentage on total permanent deformation.

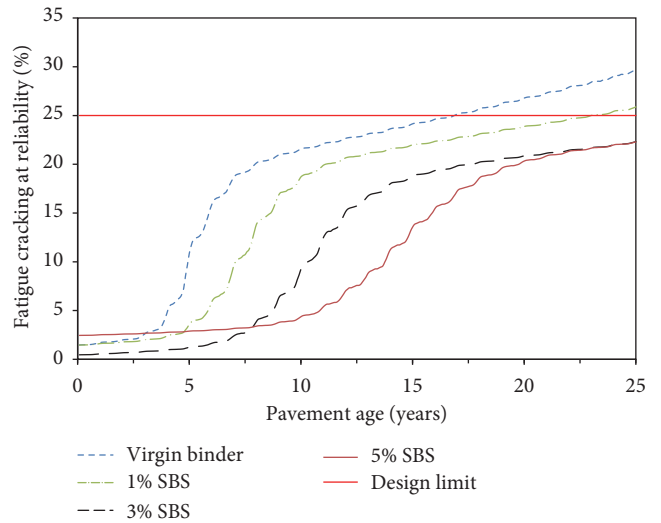


FIGURE 9: Effect of SBS percentage on bottom-up cracking.

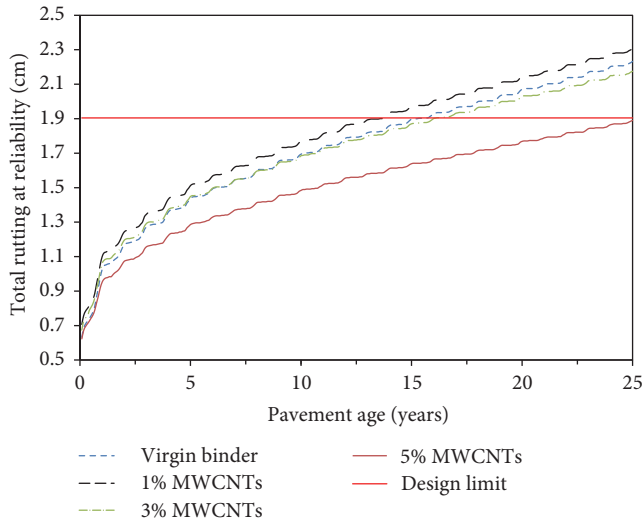


FIGURE 7: Effect of SBS percentage on total permanent deformation.

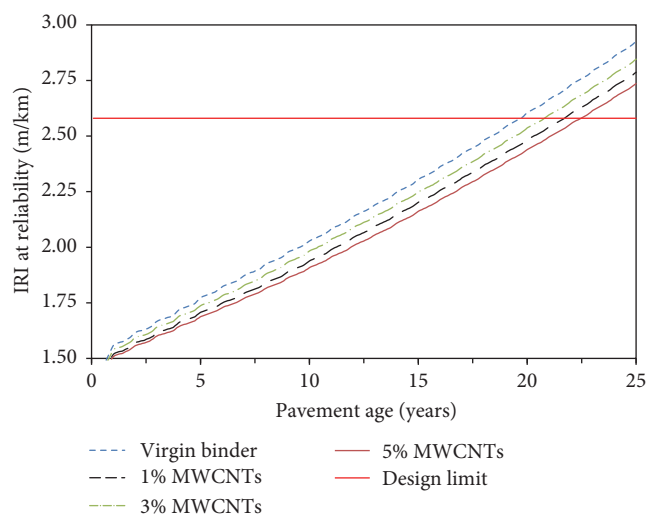


FIGURE 10: Effect of MWCNTs percentage on IRI.

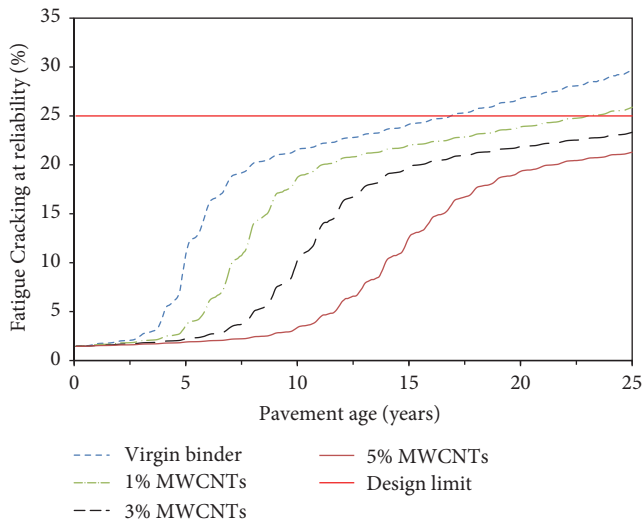


FIGURE 8: Effect of MWCNTs percentage on bottom-up cracking.

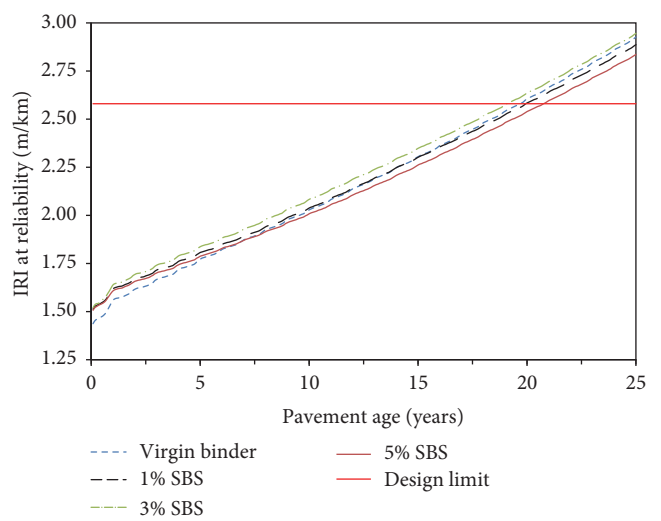


FIGURE 11: Effect of SBS percentage on IRI.

after 19 years for virgin under and after 23 years for the service life. This indicates that raising the modifier percentages will improve the performance of the pavement. In general, results show that the higher modifier percentages predicted lower IRI and higher pavement service life.

## 8. Conclusions

This study's objective was to assess the impact of adding SBS elastomer polymer and MWCNT nanomaterials to virgin asphalt on various asphalt binder rheological parameters. According to the study's findings, adding SBS to virgin asphalt increased its PG by up to 3° and increased its viscosity and rutting parameter by up to 208% and 287%, respectively. On the other hand, the addition of MWCNTs resulted in an increase in PG of up to four grades, a 264% rise in rutting parameter, and a 309% increase in viscosity.

The study comes to the conclusion that nanotechnology offers an alternate strategy for enhancing the effectiveness and longevity of materials used in road construction. The findings imply that MWCNTs are more successful than SBS in enhancing the rheological performance of asphalt binder, and it is advised to add 3% MWCNTs to produce improved asphalt binder features. These discoveries have important ramifications for the creation of more resilient and environmentally friendly road construction materials that can endure the demands of heavy traffic and unfavorable weather. To make the most of the utilization of MWCNTs in the modification of asphalt binder, more research in this area is required.

From the results obtained, it is clear that using of additives as modifiers play a very important role in the enhancement of PG for virgin asphalt after short- and long-term aging and as a result increase the resistance of asphalt to rutting deterioration.

## Abbreviations

AADT:	Annual average daily traffic
AASHTO:	American association of state highway and transportation officials
AC:	Asphalt concrete
CNT:	Carbon nanotube
DSR:	Dynamic shear rheometer
ESAL:	Equivalent single axle load
$G^* \cdot \sin \delta$ :	Fatigue parameter
$G^*/\sin \delta$ :	Rutting parameter
IRI:	International roughness index
MWCNTs:	Multiple-walled carbon nanotubes
PAV:	Pressure aging vessel
PG:	Performance grade
RPE:	Reclaimed low-density polyethylene
RTFO:	Rolling thin film oven
RV:	Rotational viscometer
SBS:	Styrene-butadiene-styrene
SCRB:	State corporation of roads and bridges
WTR:	Waste tire rubber.

## Data Availability

The datasets that support results are obtained from experimental work which was performed in the lab, so data are already available and can be found directly in the manuscript.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Acknowledgments

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