Uludağ University Journal of The Faculty of Engineering, Vol. 29, No. 2, 2024 REVIEW

DOI:10.17482/uumfd.1337107

THE USE OF GRAPHENE IN THE MODIFICATION OF BITUMEN: A LITERATURE REVIEW

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Received: 03.08.2023; revised: 30.01.2024; accepted: 28.05.2024

Abstract: In recent years, nanomaterials in bitumen modification have become widespread due to their superior properties. Graphene and its derivatives are prominent examples of this. Therefore, this review study was conducted to evaluate the effect of graphene on bitumen in detail. Accordingly, by examining the literature studies, general information about graphene and its derivatives was given, and the preparation conditions of graphene-modified bitumen were evaluated. Then, the effect of graphene on the physical and rheological properties of bitumen was investigated. In addition, the effect of graphene modification on the performance of asphalt mixtures and the use of graphene in composite modification was investigated. As a result, it was determined that graphene improves the high-temperature performance of bitumen, but its effect on the low-temperature and fatigue performance of bitumen is mostly negligible. In addition, it has been determined that graphene increases the rutting resistance of bituminous mixtures and positively affects the cracking resistance of bituminous mixtures in general.

Keywords: Bitumen, Bitumen Modification, Nanomaterials, Graphene, Rutting

Grafenin Bitüm Modifikasyonunda Kullanımı: Bir Literatür Araştırması

Öz: Son yıllarda bitüm modifikasyonunda üstün özelliklerinden dolayı nano malzemelerin kullanımı yaygınlaşmıştır. Grafen ve türevleri bunun belirgin örneklerindendir. Bu nedenle, grafenin bitüm üzerindeki etkisini detaylı bir şekilde değerlendirmek için bu derleme çalışması yapılmıştır. Bu doğrultuda, literatürdeki çalışmalar incelenerek ilk olarak grafen ve türevleri hakkında genel bilgiler verilmiş ve grafen modifiyeli bitümlerin hazırlanma koşulları değerlendirilmiştir. Ardından, grafenin bitümün fiziksel ve reolojik özellikleri üzerindeki etkisi incelenmiştir.Buna ilaveten, grafen modifikasyonun asfalt karışımların performansı üzerindeki etkisi ve kompozit modifikasyonda grafen kullanımı araştırılmıştır. Sonuç olarak, grafenin bitümün yüksek sıcaklık performansını geliştirdiği ancak bitümün düşük sıcaklık ve yorulma performansı üzerindeki etkisinin çoğunlukla ihmal edilebilir düzeyde olduğu belirlenmiştir. Ayrıca, grafenin bitümlü karışımların tekerlek izi direncini geliştirdiği ve bitümlü karışımların çatlama direnci üzerinde genel olarak olumlu bir etki gösterdiği belirlenmiştir.

Anahtar Kelimeler: Bitüm, Bitüm Modifikasyonu, Nanomalzemeler, Grafen, Tekerlek İzi

1. INTRODUCTION

Bituminous pavements have been used in various areas for many years. Although it showed adequate service performance in the first years of its use, deformations in bitumen pavements increased with rising living standards over time. In other words, the developing conditions, the increase in the human population, and the increasing commercial transportation caused an increase in

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the traffic volume. Due to these reasons and the effect of environmental conditions such as climate, deformations have increased in bitumen pavements. Therefore, there is a need to improve the properties of the pavements by modifying bitumen or bituminous mixtures. Researchers have conducted studies in this context (Erkuş et al., 2020; Feng et al., 2016; Navarro et al., 2009; Sengoz and Isikyakar, 2008; Yeşilçiçek et al., 2022) and continue to do so. Although it is possible to increase the performance of the pavement with bitumen or bituminous mixture modification, bitumen is mostly modified because the effect of bitumen on the pavement is pronounced.

With bitumen modification, the resistance of the pavement to rutting, fatigue, or low-temperature cracking can be increased. Many additives such as polymers (Navarro et al., 2009; Padhan and Gupta, 2018; Ziari et al., 2016), waste materials (Kumandaş et al., 2022; Mohd Hasan et al., 2016; Zhang and Hu, 2016), various waxes (Feitosa et al., 2016; Polacco et al., 2012; Zhang et al., 2021c), and synthesis additives (Bozdemir et al., 2023; Zhang et al., 2021a) are used in bitumen modification.

Recently, nanomaterials have attracted the attention of the asphalt industry due to their unique properties, such as small sizes ranging from 1 nm-100 nm and high specific surface area (Polaczyk et al., 2023). Various nanomaterials such as carbon nanotubes (Gong et al., 2018), nano clay (You et al., 2011), nano-silicon dioxide (Fang et al., 2016), nano fly ash (Naskar et al., 2013), carbon nanofiber (Khattak et al., 2012), nano titanium dioxide (Enieb et al., 2023), and graphene (Moreno-Navarro et al., 2018) have been used in bitumen modification. Researchers have shown great interest in graphene and its derivatives because of their excellent properties among nanomaterials (Akbari et al., 2023). Due to its high mechanical strength and specific surface properties, using graphene and graphene-based materials in bitumen modification is becoming increasingly common. Based on this, this review was made to comprehensively evaluate the effect of graphene and its derivatives on bitumen properties.

In this study, it was aimed to investigate the effects of graphene modification on the physical and rheological properties of bitumen by examining the studies in which bitumen was modified with graphene-based materials. In this context, firstly, basic information about graphene and its types is given. Then, the preparation conditions of graphene-modified bitumen, the effect of graphene modification on bitumen properties, and the effect of graphene-modified bitumen on the performance of asphalt mixtures were investigated. In addition, studies on the preparation of composite-modified bitumen using graphene and other additives are also mentioned.

2. METHOD

In this study, Web of Science and Google Scholar databases were used. A literature search was carried out by searching the terms “graphene modified asphalt”, “graphene modified bitumen”, “graphene”, “asphalt”, and “bitumen” as combinations in specified databases. While the article pool of this study was created with studies involving bitumen modification from the publications found as a result of this search, studies including the use of graphene in other fields were eliminated.

When the publications in the article pool are examined, it is seen that there are two types of studies in terms of control bitumen sample. The first of these includes studies in which base bitumen is modified with graphene. The second is the studies in which composite-modified bitumen is prepared by adding graphene to the bitumen modified with another additive. In this review, studies in which base bitumen was modified were reviewed mainly to evaluate the effect of graphene on the physical and rheological properties of bitumen independently of any other additive. Studies on the use of graphene in composite modification are examined in a separate section.

In the sub-headings of this study, explanations about graphene are given in Section 3 to provide basic information to the reader about the properties, types, and production of graphene. Then, since the preparation conditions of the modified bitumen are of great importance in laboratory and field conditions, the preparation conditions/mixing parameters of the graphene-modified bitumen were examined in Section 4. In Sections 5 and 6, the effects of graphene modification on the physical and rheological properties of bitumen are investigated, respectively. In Section 7, studies examining the performance of asphalt mixtures prepared with graphene-modified bitumen are evaluated. In Section 8, studies in which composite-modified bitumen has been produced using other additives besides graphene are reviewed. In the last part of the study, in light of the information obtained, general comments and evaluations on the use of graphene in bitumen modification were made by the authors.

3. CARBON MATERIALS AND GRAPHENE

Carbon is an element with atomic number 6, represented by the symbol C, and has nonmetallic properties. Carbon, the building block of life and living resources is also important in nanotechnology. The carbon atoms form three different bonds: sp, sp^2, and sp^3 hybridization. The type of hybridization made affects the geometry of the structure to be formed and the two- or three-dimensional material structure (Yeter, 2018). The two most well-known allotropes of carbon are diamond and graphite. While carbon atoms form a tetrahedral arrangement by sp^3 hybridization in diamond, they form hexagonal and layered structures by sp^2 hybridization in graphite.

The primary carbon materials are diamond, graphite, fullerenes, carbon fibers, amorphous carbon, glassy carbon, carbon nanotubes (CNTs), black carbon, coal tar, carbon black, and graphene and its derivatives (Yeter, 2018).

3.1. Graphene

Graphene was added to the literature in 2004 when Andre Geim and Konstantin Novoselov obtained only one-atom-thick carbon layer from graphite (Gerstner, 2010). Andre Geim and Konstantin Novoselov were awarded the 2010 Nobel Prize in Physics for being the first to isolate and demonstrate the properties of this remarkable material, graphene (Gerstner, 2010). Graphene, one of the current research topics of condensed matter physics, has unique properties in many ways. It is the thinnest crystalline material imaginable yet the hardest, strongest, and best conductive material known (Sahin et al., 2015). Due to these properties, graphene, which is essential in nanotechnology applications, has applications in different industries, from energy to medicine. Since graphene is a strong and light material, it has attracted the attention of researchers in the construction industry. It has been used in bitumen modifications and as an additive in cement (Gholampour et al., 2017; Yang et al., 2019).

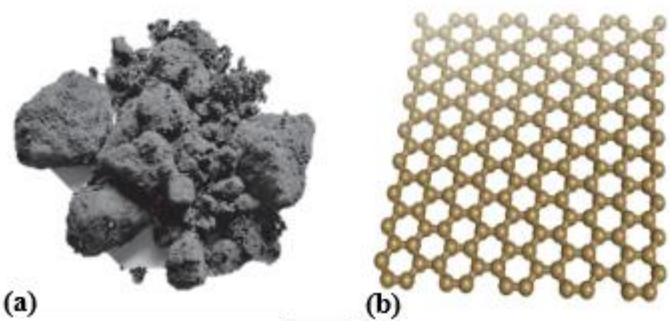

Figure 1:
a. *macro* ***b.*** *structural images of graphene <(Liu et al., 2021; Radic et al., 2013)>*

3.2. Graphene Oxide (GO)

Graphene oxide is a single-layer material obtained by oxidation of graphite layers (Bedeloğlu and Mahmut, 2016). It is produced by oxidizing graphite with oxidants such as sulfuric acid, sodium nitrate, and potassium permanganate (Uygunoğlu and Şimşek, 2019). GO's usage areas are increasing due to its dielectric properties, ease of changing electronic properties, transparency, and mechanical properties (Yazıcı et al., 2016). In addition, its easy dispersion in solvents facilitates its use in bitumen modification and homogeneous distribution in bitumen (Yazıcı et al., 2016).

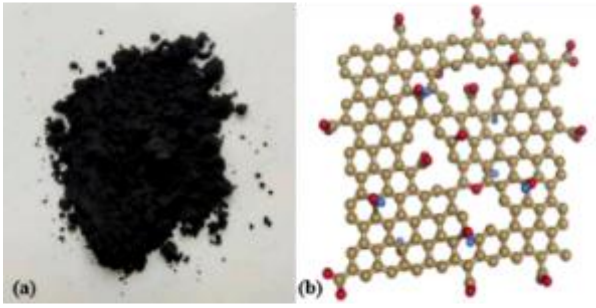

Figure 2:
***a.** macro **b.** structural images of GO <(Wu et al., 2022c; Radic et al., 2023)>*

4. CONDITIONS OF PREPARATION OF MODIFIED BITUMEN

The preparation conditions of the modified bitumen are essential in terms of producing a stable bitumen by homogeneously dispersing the additives in the bitumen. The mixing parameters chosen during the preparation of modified bitumen significantly affect the performance of the bitumen. Mixture preparation parameters can be listed as the amount of additive to be added to the bitumen, type of mixer, mixing time of the additive and bitumen, mixing temperature, and speed.

When the mixing parameters given in Table 1 are examined, it is seen that there are differences between the studies. The studies show that while producing modified bitumen, temperatures between 120-180 ˚C and mixing time between 10-180 minutes are used. However, although some of the authors of the studies are are the same, there is a concentration on the temperature of 160 °C and a mixing time of 45 min. On the other hand, when the speed parameter is examined, speeds ranging from 500 rpm to 15000 rpm are seen, but the density is above 4000 rpm. From this, it is thought that the optimum mixing parameters for graphene modified bitumen production can be considered as 45 min, 160 ˚C and 4000 rpm for time, temperature and speed, respectively. In addition, a gradual mixing procedure was applied in some studies. The most prominent example of this is the work by Yang et al. (2020). In this study, mixing was carried out for 20, 20, 20, 20, and 40 minutes at 145-150 °C and at speeds of 300, 500, 1000, 2000, and 2500 rpm, respectively. Similarly, Wang et al. (2022) mixed the graphene-bitumen mixture manually for 5 minutes at 155 °C and then mixed it with a high-speed mixer at 180 °C at 15000 rpm for 60 minutes. It can be said that the primary aim of these and other studies, in which the gradual mixing procedure is adopted, is to obtain a homogeneous bitumen by dispersing the graphene in the bitumen without clumping.

Table 1. Preparation conditions of graphene-modified bitumen

Graphene type	Mixer type	Mixing parameters			Ref.
		Time (min)	Temperature (˚C)	Speed (rpm)	
Graphene	-	10	150		(Moreno-Navarro et al., 2018)
Graphene	high-speed shear	30	170	5000	(Yang et al., 2019)
Graphene	-	60	140	1720	(Ahmad Nazki et al., 2020)
Graphene	high-speed shear	180	135	1000/3000	(Li et al., 2021a)
Graphene	-	60	135	2000	(Zhou et al., 2017)
Graphene	manual/mechanical	-/60	145	500	(Li et al., 2021b)
Graphene	high-speed mec.	60	150	3000	(Eisa et al., 2021)

Table 1. ***Continued***

Graphene type	Mixer type	Mixing parameters			Ref.
		Time (min)	Temperature (˚C)	Speed (rpm)	
Graphene	-	20/20/20/40	145-150	300/500/1000/2000/2500	(Yang et al., 2020)
Graphene	manual/high-speed shear	5/60	155/180	15000	(Wang et al., 2022)
Graphene	manual/high-speed shear	-/30	155/180	12000	(Polaczyk et al., 2023)
GO*	high-speed shear/ultrasonic disperser	45/30	120/120	5000	(Wang et al., 2019)
GO	high-speed shear	45	160	4000	(Adnan et al., 2020, 2022a; Adnan et al., 2021a, 2021b, 2023; Adnan et al., 2022b)
GO	high-speed shear	45	160±5	4000	(Akbari et al., 2023)
GO	high-speed shear	30	150	4000	(Zeng et al., 2020)
RGO**	mechanical/high-speed shear	20/40	130-145	2000/5000	(Chen et al., 2021)
GO	stir/high-speed shear	30/60	150	500rad / 5000 rpm	(Wu et al., 2022a)
GO	high-speed shear	8/5	160	3000/5000 rad/min	(Jiang et al., 2022)
GO	high-speed shear	-	135	-	(Zeng et al., 2017)
GO	high-speed shear	30	150,170	4000	(Li et al., 2018b)
GO	high-speed shear/ultrasonic disperser	45/30	120,150	5000	(Wang et al., 2020)
GO	high-speed shear	30	155,170	4000	(Wu et al., 2017)
Graphite	high-speed shear	-	145	2000	(Yao et al., 2016)

* GO: Graphene oxide
** RGO: Reduced graphene oxide

5. EFFECT ON PHYSICAL PROPERTIES

The physical properties of bitumen include penetration, softening point, and ductility. Penetration and softening point values give information about the hardness-softness of the bitumen, while the ductility value represents the extensibility of the bitumen. These properties, determined by conventional bitumen tests, give an idea about the performance of bitumen rather than precise information. For example, bitumen with a low penetration value and high softening point is expected to have superior high-temperature performance. Bitumen with high ductility is expected to have high resistance to thermal cracking. To examine the effect of graphene on the physical properties of bitumen, the studies that included conventional tests in the article pool of this study are listed in Table 2. This table indicates the effect of graphene by considering the base bitumen grade, graphene type, and additive ratio used in the studies. By examining each study, the effect of graphene on the penetration, softening point, and ductility of bitumen is indicated by the up and down arrows for increase and decrease, respectively. If any of these physical properties have not been examined, the "-" sign indicates this. Finally, the findings from conventional tests are briefly presented. When Table 2 is examined, it is seen that graphene causes a decrease and an increase in the penetration and softening point value of bitumen, respectively, in all the studies given. These results show that graphene increases the consistency of bitumen, indicating that graphene modified bitumen can have high deformation resistance at high temperatures. On the other hand, the addition of graphene negatively affected the ductility of the bitumen. Therefore, graphene modification can reduce the resistance of bitumen to thermal cracking.

Table 2. Effect of graphene modification on the physical properties of base bitumen

No	Base Bit.	Graphene		Effect of graphene			Results	Ref.
		Type	Ratio (%)	Pen.*	S.P.*	Duc. *		
1	N/A*	Graphene	0.5, 1, 1.5, 2	-	↑	-	It has been stated that there is no significant difference in the softening point of modified bitumen compared to base bitumen. It has been noted that this may be because the softening point test may not be sensitive enough to evaluate the effect of graphene on bitumen.	(Ahmad Nazki et al., 2020)
2	B70/ 100	Graphene	0.5, 1, 1.5	↓	↑	↓	It was determined that while the softening point of bitumen increased with the addition of graphene, the penetration and ductility values decreased.	(Li et al., 2021a)
3	B70/ 100	Graphene	1, 2, 3, 4, 5	↓	↑	↓	It has been stated that graphene increases the PI and equivalent softening point of bitumen, improving its heat sensitivity and high-temperature stability. On the other hand, the addition of graphene significantly reduced the ductility of the bitumen.	(Li et al., 2021b)
4	B50/ 70	GO	0.5, 1, 1.5	↓	↑	↓	In this study, the physical properties of bitumen were investigated before and after aging by various methods. It has been determined that the percentage of permanent penetration of modified bitumen after aging is high, and the softening point increase is low compared to base bitumen. It has been stated that this is indicative of better-aging resistance. Similarly, it was noted that the percentage of permanent ductility of modified bitumen is higher than that of base bitumen and that GO can reduce the ductility weakening during aging.	(Wang et al., 2019)
5	B50/ 70	GO	0.2, 0.5, 0.8	↓	↑	↓	It has been stated that using graphene increases the softening point of bitumen while decreasing the penetration and ductility value.	(Akbari et al., 2023)
6	B50/ 70	GO	0.1, 0.3, 0.5, 0.7, 0.9	↓	↑	↓	It was observed that as the graphene content increased, the rate of increase in softening point and decrease in penetration and ductility decreased. After aging with RTFOT, modified bitumens showed a higher percentage of permanent penetration than control bitumen. In addition, it was determined that the optimum GO content in terms of physical properties was between %0.5 and %0.7.	(Zeng et al., 2020)
7	B70/ 100	RGO	0.1, 0.3, 0.5	↓	↑	↓	It has been determined that modified bitumen has higher PI and equivalent softening point compared to base bitumen. Therefore, it has been stated that modified bitumen can be used in hot regions to resist rutting.	(Chen et al., 2021)
8	B70/ 100	GO	0.5, 1, 1.5, 2	↓	↑	↓	While the penetration and ductility values decreased with the addition of GO, the softening point value increased. However, it was seen that the increase or decrease with increasing GO content was not linear. As the GO content increased, the penetration and ductility values first decreased, then increased, and the reverse was true for the softening point.	(Wu et al., 2022a)

Table 2. ***Continued***

No	Base Bit.	Graphene		Effect of graphene			Results	Ref.
		Type	Ratio (%)	Pen.*	S.P.*	Duc.*		
9	B70/ 100	GO	5, 10, 15	↓	↑	↓	It has been stated that GO may have a positive effect on the high-temperature performance of bitumen as it causes an increase in the softening point of the bitumen. Still, because it reduces ductility, the increase in the GO content negatively affects the low-temperature properties of the bitumen.	(Jiang et al., 2022)
10	B50/ 70 B70/ 100	GO	1, 3	↓	↑	-	From the penetration and softening point values obtained before and after TFOT and PAV aging, it was stated that GO could improve anti-aging properties, but this effect was not significant.	(Zeng et al., 2017)
11	B50/ 70	GO	0.5, 1, 1.5, 2	↓	↑	↓	Bitumen containing 2.5% GO gave a lower softening point than that containing 2%. It has been stated that this is an indication that excess GO causes agglomeration in the asphalt, reducing homogeneity.	(Adnan et al., 2022a)
12	B50/ 70	GO	0.006, 0.012, 0.024, 0.048	↓	↑	↓	It was determined that GO caused little increase in the softening point of bitumen, and its addition in low ratios did not affect the ductility of the bitumen much.	(Yu et al., 2019)
13	B70/ 100	GO	1, 3	↓	↑	↓	It has been noted that binders containing GO tend to harden.	(Li et al., 2018b)
14	B70/ 100	GO	1, 3	-	-	-	In this study, the relevant test results are not shared directly, but permanent penetration and permanent ductility rates and softening point increases are given after UV aging. It has been stated that while GO increases the permanent penetration and ductility rates, it decreases the softening point increase.	(Wu et al., 2017)
15	B50/ 70	Graphene	0.5, 1, 1.5	↓	↑	-	It has been determined that graphene decreases the penetration of bitumen and increases the softening point. From this, it was stated that graphene can increase the deformation resistance of asphalt.	(Eisa et al., 2021)

*Pen.: Penetration; S.P.: Softening point; Duc.: Ductility; N/A: Not available

6. EFFECT OF GRAPHENE ON RHEOLOGICAL PROPERTIES

6.1. Effect of graphene on workability

In general, the viscosity of bitumen increases in parallel with the increase in consistency. Values from the rotational viscosity (RV) tests are often used as a measure of the viscosity of bitumen. The measured viscosity values give an idea of the workability of the bitumen. For the constructionof the pavement, the bitumen must have sufficient workability. Therefore, the Superpave specification (ASTM D6373-15, 2015) states that the viscosity values of bitumen measured at 135 °C should not exceed 3000 Pa. To evaluate the effect of graphene on the workability of bitumen, studies involving the RV test were reviewed in the article pool of this study. In common in these studies, it was stated that graphene increases the viscosity of bitumen (Eisa et al., 2021; Jiang et al., 2022; Li et al., 2021a; Wu et al., 2022a; Yang et al., 2020; Yao et al., 2016; Zeng et al., 2020). These findings align with the hardening effect of graphene on the consistency of bitumen mentioned in the previous section. Increasing the viscosity of the bitumen negatively affects the workability by increasing the mixing and compaction temperatures of the bitumen. Therefore, adding graphene can reduce the workability of bitumen, increasing the energy and carbon emissions required for pavement construction.

6.2. Effect of graphene on rutting, fatigue, and low-temperature performance

One of the most common tests used to determine the resistance of bitumen to rutting at high temperatures is the dynamic shear rheometer (DSR) test. As a result of DSR tests carried out at high temperatures, the complex shear modulus (G*), phase angle (δ), and the rutting parameter, also known as the G*/sinδ parameter, are determined from these values (Oruç and Yılmaz, 2016). An increase in the rutting parameter indicates an increase in the rutting resistance. The studies examining the rutting resistance and other rheological properties of graphene-modified bitumen are given in Table 3. Since it is impossible to provide the results obtained in these studies numerically, the parameters examined in each study are marked in the table. When the rutting parameters obtained in the references given in the table were concerned, it was observed that there was a significant increase in the G*/sinδ value with the graphene modification. This result shows that the rutting resistance of graphene-modified bitumen will be high.

Another method used to examine the rutting performance of bitumen is the multiple stressed creep and recovery (MSCR) test. With this test, which is more up-to-date than the DSR test, the response of modified bitumen under stress is evaluated over two parameters: non-recoverable creep compatibility (Jnr) and the percent recovery (%R). When the studies, including the MSCR test given in Table 3, are examined, it is seen that the Jnr value of the bitumen decreases. In contrast, the %R value generally increases with adding graphene. These results show that with the addition of graphene, the elasticity of the bitumen increases, and the rutting resistance improves.

Another rheological property of bitumen is its resistance to fatigue at intermediate service temperatures. The G*sinδ parameter obtained from the DSR test is often taken as the measure of this resistance. It is desirable that this parameter, also known as the fatigue parameter, is small. As the parameter increases, resistance to fatigue cracks decreases. When the effect of graphene on fatigue resistance is examined, there is no stability in the literature. Adnan et al. (2021b) and Wu et al. (2017) reported that graphene improved fatigue resistance. However, Li et al. (2018b) determined that the G*sinδ value increased slightly (0.55%) by adding 1% graphene to base bitumen, decreasing by 3.52% by adding 3%. Another study (Wang et al., 2022) stated that while the graphene added to base bitumen was below 0.65%, the fatigue parameter was minimal. Still, a significant increase was observed at 5% and 7% rates. From these results, it can be said that the effect of graphene on the fatigue resistance of bitumen does not show a clear trend. For the most part, the impact of graphene on the G*sinδ parameter was negligible. However, due to the high specific surface area of graphene, like other nanomaterials, it can be expected that it can prevent the free movement of asphalt; its surface pores can absorb the light components of asphalt, and for this reason, bitumen can be expected to behave more brittle (Wang et al., 2022).

Table 3. Effect of graphene modification on the rheological properties of base bitumen

No	Base bitumen	Graphene		Rheological effect								PG	Ref.
		Type	Ratio (%)	G*	δ	G*/sinδ	G*sinδ	S(t)	m-value	MSCR	Other		
1	B50/70	Graphene	0.1, 0.5, 1	✓	✓	✓				✓			(Moreno-Navarro et al., 2018)
2	B70/100	Graphene	0.2, 0.4, 0.6, 0.8, 1, 1.5	✓	✓					✓			(Yang et al., 2019)
3	N/A	Graphene	0.5, 1.0, 1.5, 2.0	✓	✓					✓			(Ahmad Nazki et al., 2020)
4	B70/100	Graphene	0.5, 1, 1.5	✓	✓								(Li et al., 2021a)
5	B50/70	Graphene	2, 4, 6, 8, 10	✓	✓								(Yang et al., 2020)

Table 3. ***Continued***

No	Base bitumen	Graphene		Rheological effect								PG	Ref.
		Type	Ratio (%)	G*	δ	G*/sinδ	G*sinδ	S(t)	m-value	MSCR	Other		
6	B70/100	Graphene	0, 1, 2, 3, 4, 5	✓	✓	✓						PG64-XX PG64-XX PG64-XX PG64-XX PG70-XX PG70-XX	(Li et al., 2021b)
7	B50/70	Graphene	2, 3, 4	✓		✓				✓		PG70-XX PG76-XX PG76-XX PG82-XX	(Liu et al., 2023)
8	PG64-22	Graphene	0.3, 0.65, 1, 1.5, 2.5, 5, 7	✓	✓	✓	✓	✓	✓				(Wang et al., 2022)
9	PG64-22	Graphene	0.3, 0.65, 1, 2.5, 5	✓	✓	✓	✓	✓	✓				(Polaczyk et al., 2023)
10	B50/70	GO	0.5, 1.0, 1.5	✓	✓		✓						(Wang et al., 2019)
11	B50/70	GO	0.5, 1.0, 1.5, 2, 2.5	✓	✓	✓				✓			(Adnan et al., 2021a)
12	B70/100	GO	0.5, 1.0, 1.5, 2	✓	✓	✓		✓	✓	✓			(Wu et al., 2022a)
13	B70/100	GO	5, 10, 15	✓	✓	✓							(Jiang et al., 2022)
14	B50/70 B70/100	GO	1, 3	✓	✓								(Zeng et al., 2017)
15	B50/70	GO	0.5, 1.0, 1.5, 2, 2.5				✓	✓	✓		LAS test		(Adnan et al., 2021b)
16	B50/70	GO	0.006, 0.012, 0.024, 0.048					✓	✓				(Yu et al., 2019)
17	PG64-22	GO	0.02, 0.05, 0.1, 0.2			✓		✓	✓	✓			(Liu et al., 2018a)
18	B70/100	GO	1, 3	✓	✓		✓	✓	✓				(Li et al., 2018b)
19	B50/70	GO	0.5, 1.0, 1.5	✓	✓					✓			(Wang et al., 2020)
20	B70/100	GO	1, 3	✓	✓		✓						(Wu et al., 2017)
21	B50/70	Graphite	2, 4, 6, 8, 10	✓	✓	✓					Storage and loss modules		(Oladunjoye et al., 2021)
22	B50/70	Graphene	0.5, 1.0, 1.5			✓							(Eisa et al., 2021)
23	PG52-34	Graphite	3, 6	✓	✓			✓	✓				(Le et al., 2020)

Another rheological property of bitumen is its resistance to cracking at low temperatures. The bending beam rheometer (BBR) test is often used to determine the resistance of bitumen to low-temperature cracking. As a result of this test applied to the bitumen sample aged by the pressure aging vessel (PAV) method, two parameters are obtained: creep stiffness (S) and m-value. The decrease in the S value of the bitumen and the increase in the m value indicates improved low-temperature performance. Studies examining the S and m parameters of graphene modified bitumen are given in Table 3. When the findings obtained in these studies were examined, it was determined that there was no definite trend regarding the effect of graphene on low-temperature performance. Some studies (Adnan et al., 2021b; Li et al., 2018b; Wu et al., 2022a; Yu et al., 2019) showed a positive trend on low temperature, while others (Polaczyk et al., 2023; Wang et al., 2022) showed the opposite. However, generally, the positive or negative effect of graphene is negligible. In only one study (Polaczyk et al., 2023), low-purity graphene significantly affected low-temperature performance negatively, even at low doping ratios. From these results, it can be said that the effect of graphene on the low-temperature performance of bitumen depends on its homogeneous distribution and the type, purity, and size of graphene used.

7. EFFECT OF GRAPHENE MODIFICATION ON THE PERFORMANCE OF ASPHALT MIXTURES

The performance of bituminous pavements against deformations in traffic and environmental conditions can be predicted by applying various tests to bituminous mixtures. Marshall stability and flow test, rutting test, and semi-circular bending (SCB) test are commonly used in this context. Although the number of studies on mixtures prepared with graphene-modified bitumen is limited in the literature, there are studies including the specified tests (Adnan et al., 2020, 2022a; Adnan et al., 2022b; Akbari et al., 2023; Eisa et al., 2021; Le et al., 2020; Tabasi et al., 2023). The results from the studies in which Marshall stability and flow test were applied to asphalt mixtures prepared with graphene-modified bitumens are consistent. In these studies (Adnan et al., 2022a; Eisa et al., 2021), it was determined that the stability values of the mixtures containing graphene were higher, and the flow values were lower compared to the control mixtures prepared with base bitumen. These results indicate that pavements constructed with graphene-modified bitumen will have better-rutting resistance than pavements prepared with base bitumen. The results of the studies examining the effect of graphene modification on the rutting resistance of bituminous mixtures overlap with each other. Under cyclic loading, the graphene mixture samples showed less rutting depth than those without (Akbari et al., 2023; Eisa et al., 2021). These results show that the deformation resistance of asphalt mixtures is improved by graphene modification.

In studies in which the effect of graphene on the cracking resistance of bituminous mixtures was investigated with the SCB test, mostly similar results were obtained. Akbari et al. (2023) and Adnan et al. (2022b) determined that adding GO improves the crack toughness of bituminous mixtures at low temperatures. In another similar study (Le et al., 2020), it was stated that the mixture prepared with bitumen containing 3% graphene showed higher cracking energy, but the mixture prepared with bitumen containing 6% graphene showed lower cracking energy compared to the control mixture. From these results, it can be said that the effect of graphene on the low-temperature performance of bituminous mixtures is generally favorable. Still, more studies are needed on this subject. On the other hand, in studies where SCB tests were carried out at medium temperatures, it was determined that adding GO to bitumen increased the cracking energy of bituminous mixtures (Adnan et al., 2020; Tabasi et al., 2023). These results show that graphene modification increases the cracking resistance of bituminous mixtures at medium temperatures.

8. USE OF GRAPHENE IN COMPOSITE MODIFICATION

Composite modifications are modifications in which bitumen is modified with more than one additive. The primary aim of these modifications is to improve the behavior of bitumen in many ways by utilizing the properties of different additives. As the use of graphene in bitumen modifications has become widespread, the use of graphene in composite modification has also increased. The studies in which graphene was used in composite modifications, and the additives used with graphene in these studies are given in Table 4. It can be seen from this table that the effect of graphene on bitumen is often examined together with SBS. This result is a natural consequence of SBS being one of the most common additives in bitumen modification. In these studies, it was determined that graphene further improved the elastic properties and rutting resistance of SBS-modified bitumen (Li et al., 2023; Liu et al., 2018a; Lu et al., 2022; Polaczyk et al., 2023; Wang et al., 2021; Wang et al., 2022; Wu et al., 2022b). In Table 4, it is seen that another commonly used additive with graphene is rubber powder. These studies show that graphene improves the high-temperature performance of rubber-modified bitumen (Chen et al., 2020; Gao et al., 2023; Li et al., 2022e; Lin et al., 2019). From these results, it is seen that graphene has positive effects, especially on rutting resistance in studies where it is used in composite modifications. For this reason, it is thought that the use of additives that have a negative effect on rutting resistance while improving some properties of bitumen together with graphene will be better in terms of the high-temperature performance of bitumen.

Table 4. Additives used with graphene in bitumen modification

Composite additive	Ref.
Styrene Butadiene Styrene (SBS)	(Duan et al., 2019; Huang et al., 2022; Jiangmiao et al., 2020; Le et al., 2020; Li et al., 2023; Li et al., 2018b; Li et al., 2022d; Liu et al., 2018a; Lu et al., 2022; Polaczyk et al., 2023; Wang et al., 2020; Wang et al., 2021; Wang et al., 2022; Wu et al., 2022b; Wu et al., 2017; Zhu et al., 2020)
Polystyrene+SBS	(Han et al., 2018a; Li et al., 2018a; Wei et al., 2020)
Octadecyle Amine+SBS	(Han et al., 2018b; Li et al., 2018a; Wei et al., 2020)
TiO_2+Polystyrene+SBS	(Yang et al., 2021a)
Polyvinylpyrrolidone (PVP)+SBS	(Liu et al., 2021b)
PMMA+SBS	(Li et al., 2020)
Polyurethane+SBS	(Li et al., 2022a; Li et al., 2022b; Li et al., 2022c)
Polyurethane	(Wang et al., 2023; Wu et al., 2022c; Yu et al., 2021)
Rubber Powder	(Chen et al., 2020; Gao et al., 2023; Guo et al., 2022; Li et al., 2022e; Liao et al., 2022; Lin et al., 2019; Xie et al., 2023)
Epoxy Resin	(Si et al., 2023; Zhang et al., 2023; Zhang et al., 2022a; Zhang et al., 2021b; Zhao et al., 2022a, 2022b)
Polyethylene (PE)	(Hu et al., 2022; Zhou et al., 2021)
Carbon Nanotube (CNT)	(Yang et al., 2021b; Yu et al., 2022)
Crumb Rubber	(Singh et al., 2020)
Carbon Black+Crumb Rubber	(Liu et al., 2021a)
Tourmaline	(Guo et al., 2021; Guo et al., 2019)
Ethylene Bis (Stearamide) (EBS)	(Zhang et al., 2019)
Waste EVA	(Zhang et al., 2022b)
Polypropylene	(Liu et al., 2022)
Sasobit, WCO	(Liu et al., 2018b; Liu et al., 2019; Zhu et al., 2019)
Bio-Oil	(Qian et al., 2022)

9. CONCLUSIONS

This review investigated the use of graphene and its derivatives in bitumen modification and their effects on the physical and rheological properties of bitumen. In this context, the studies in the literature were examined in detail, and an evaluation was made by comparing the findings obtained from these studies. The results obtained from this study are listed below.

- Graphene modification caused a decrease in the penetration value of the bitumen and an increase in the softening point. On the other hand, the addition of graphene had a negative effect on the ductility of the bitumen.
- When the effect of graphene on the rutting resistance of bitumen is evaluated, the parameters obtained from both the DSR and MSCR tests indicate that the graphene modification has an enhancing effect.
- When the effect of graphene on the fatigue performance of bitumen was evaluated, there was no noticeable trend.
- When the effect of graphene on the low-temperature performance of bitumen was evaluated, it was determined that a positive or negative effect observed depending on the preparation conditions of the modified bitumen, the type of graphene used, the additive ratio, size, and purity.
- When the effect of graphene on the performance of bituminous mixtures was examined, the graphene mixture showed higher stability, less flow value, and less rutting depth than the control mixtures. On the other hand, graphene has generally improved the crack resistance of bituminous mixtures at low and medium temperatures, but more research is needed.

In light of the information obtained from this study, it has been determined that graphene and its derivatives significantly improves the rutting resistance of bitumen and bituminous mixtures. For this reason, it is thought that using graphene-modified bitumen in pavements to be built in hot climates exposed to high temperatures would be beneficial. In further studies, it is believed that researchers will contribute to the literature by examining the effects of graphene and its derivatives on the cracking resistance of bitumen at low and medium temperatures, including graphene type, size, additive ratio, and different mixing procedures. In addition, since there are limited studies examining the effect of graphene on the properties of bituminous mixtures, it is thought that investigating the impact of graphene and its derivatives on the performance of bituminous mixtures with various experimental methods will fill the gap in the literature.

CONFLICT OF INTEREST

The author(s) confirm that there are no known conflicts of interest or common interests with any organization or individual.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

Neslihan ŞAHAN Conceptualization, Investigation, Writing – original draft, Visualization; Aytuğ KUMANDAŞ Visualization, Writing – original draft; Şeref ORUÇ Conceptualization, Supervision.

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