

Review

A Review on Improving Asphalt Pavement Service Life Using Gilsonite-Modified Bitumen

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Abstract: Hot mix asphalt has various benefits such as good workability and durability. It is one of the most general materials used as asphalt mixtures in road pavements. Asphalt mixtures and binders can be improved by modifying them with various additives. Gilsonite is a natural asphalt hydrocarbon which may be used as an additive to hot mix asphalt. It is used as an asphalt binder modifier (wet process) and an asphalt mixture modifier (dry process) to improve the properties of the mix. It provides the option of improved rheological properties, stability, strength rutting resistance and moisture sensitivity. This paper examines the current research relating to the use of gilsonite to improve the asphalt properties (binder and mixture). The rheological properties of the modified asphalt binders and mechanical properties of the modified asphalt mixtures will be reviewed. The influence of adding gilsonite individually or combined with other additives will be discussed. Furthermore, assessment of the environmental and economic perspectives of the studied asphalt along with some suggestions to improve the asphalt binders and mixtures will be explored.

Keywords: polymer modified asphalt; asphalt gilsonite; natural asphalt; powder asphalt



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1. Introduction

The life cycle of asphalt mixtures (surfacing layer) can be determined by measuring the fatigue and stress characteristics including permanent deformation resulting from traffic loading [1]. Therefore, improving the quality of the asphalt mixture such as rutting resistance and moisture damage will lengthen the pavement service life, thus, accordingly enhancing the sustainability [2]. Hot Mix Asphalt (HMA) is an essential material in the paving industry, and offers several advantages including versatility, good durability, and cost-effectiveness [3]. HMA is a combination of two primary ingredients: aggregates and asphalt binder [4]. HMA deteriorations including aging, low temperature and fatigue cracking, rutting, and moisture-induced damages are mostly due to the mechanical and rheological properties of the binder and asphalt mixtures [5]. The global consumption of asphalt binder has rapidly increased since the 1900s, being used mostly as the binder for asphalt mixtures for road construction and maintenance [6]. Poor asphalt not only reduces the pavement service life but also causes further issues, for example increased repair, premature failure, increased maintenance costs [7], hazardous conditions for road users and finally, reduced safety.

Road authorities, the asphalt industry, and the related researchers have paid increased attention to modify the asphalt binder with various materials to decrease the life-cycle cost

and develop the service performance of roads. Asphalt modification is one of the most popular approaches [8]; modified asphalt has been developed to produce an alternative to traditional HMA. In the process, additives were added to the binder to improve asphalt binder characteristics, such as the adhesion to aggregate, the properties of final asphalt mixture, and workability [9]. Additives are used to increase the pavement service life and delay deterioration. These additives can be either added to asphalt binders (wet process) or added immediately into the mixture in plants (dry process) [10,11]. Although numerous additives increase the performance of asphalt binder and mixture, the proper performance of an additive must not be the primary principle for selecting it; there are also other aspects including environmental and economic concerns that should be considered when choosing an additive [12]. One of the important additive materials is gilsonite. Gilsonite is a resinous hydrocarbon belonging to the hydro carbonates in the classification of asphalt binder modifiers [12,13], and has been assessed and utilized in numerous industrial aspects [12]. Gilsonite, the scientific name of which is “uintaite”, was discovered in the 1860s, and later referred to as “gilsonite” after Samuel H. Gilson [14]. Gilsonite is also referred to as asphaltum, natural asphalt, mineral asphalt binder, asphalt binder powder mineral tar or drilling mud [14].

This paper focuses on using gilsonite in asphalt pavement, and aims to give a comprehensive overview of the research on numerous studies. This paper also provides a comprehensive comparison of the environmental and financial aspects, in addition to the potential future development of asphalt modified with gilsonite.

Characterisation of the Gilsonite

Physical properties of gilsonite: a black and breakable mineral, which is simply crushed into powder [12], and used in numerous industrial contexts [15]. Table 1 illustrates the physical properties of gilsonite. At the start of the process, the natural asphalt binder must be separated from the underground stone reservoir and exploited through ground layer failures. It may undergo steady solidification and oxidization, ending up as a solid and hard-mineral asphalt binder if it stayed underground or near the ground surface [14,15]. Gilsonite is found abundantly mainly in Iran and America, and can simply be mixed with asphalt binder as a result of their comparable structures [16]. Hence, gilsonite was involved in numerous studies to modify asphalt binders, and it was stated that asphalt binder performance such as stiffness, elasticity and resistance to moisture damage increased as a result [12,17].

Table 1. Physical properties of gilsonite [18].

Property	Value
Penetration (mm) at 25 °C	0
Softening Point, °C	225 ± 5
Specific Gravity at 25 °C	1.11
Colour in Mass	Black
Colour in Powder	Brown

Chemical compositions of gilsonite: It is well understood that the chemical composition of gilsonite as a raw material has an important effect on the properties of the resultant asphalt mixture [12]. Therefore, the chemical composition of gilsonite should be examined to reveal the performance of these materials when in combination with asphalt binder or being chemically triggered by other additives. However, based on Fourier transform infrared (FTIR) spectroscopy test results, no chemical reactions between the neat asphalt binder and gilsonite powder were evidenced [18]. This was due to the obvious similarities in the spectra of these materials; it was expected that adding gilsonite powder to neat

asphalt binder would not change the peak locations in its spectrum [18]. The chemical composition of gilsonite as collected from the elemental analysis are presented in Table 2.

Table 2. Chemical properties of gilsonite [14].

Gilsonite Chemical Components											
Nitrogen, wt.%	Carbon, wt.%	Hydrogen, wt.%	Sulfur, wt.%	Oxygen, wt.%	H/C, atomic ratio	O/C, atomic ratio	C/N, atomic ratio	S/C, atomic ratio	Heating value (Btu/Lb)	Organic matter, wt.%	Inorganic matter, wt.%
3.25	84.36	10.05	0.27	1.36	1.44	0.01	39.05	0.01	17	99.32	0.68

According to reviewed papers, FTIR and thin layer chromatography-flame ionization detection (TLC–FID, in an Iatroscan instrument) demonstrated the gilsonite properties. FTIR spectroscopy is an important tool for the characterization of asphalt binder and its products, as it provides a rapid comprehensive assessment of the structure and composition of the materials [19]. It is also a suitable technique for obtaining qualitative mineralogy [14,20]. Based on FTIR analysis (as shown in Figure 1), there are many similarities between gilsonite powder and neat asphalt binder [18]. The hydrogen-bonded O–H and the greater pyrrolic N–H show the presence of long aliphatic chains in gilsonite, and the pronounced aliphatic characters are linked with their suitability for liquefaction [14]. However, there is a difference between gilsonite and asphalt binder, which is the high intensity of the peak at 1030 cm^{-1} due to the sulfoxide group. The peaks in the range of 700 cm^{-1} to 900 cm^{-1} wavenumbers are correlated with the aromatic compounds that are also evident for both neat asphalt binder and gilsonite powder [17].

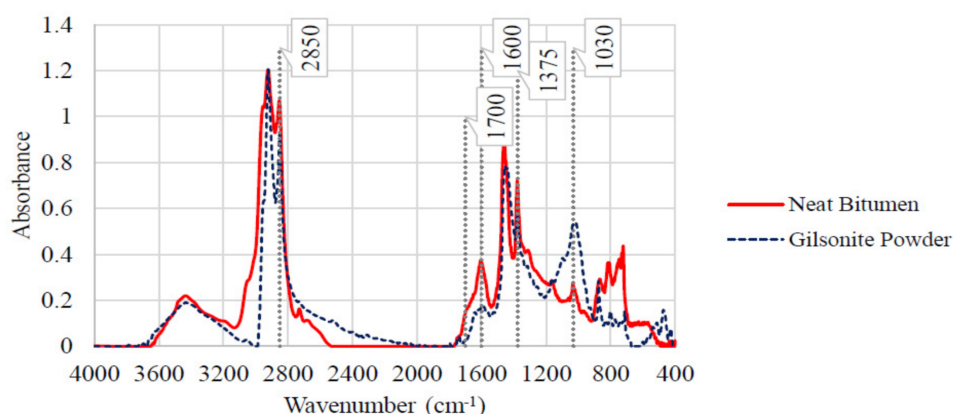


Figure 1. FTIR spectroscopy for gilsonite analyses [18].

TLC–FID is used as an inexpensive and rapid method to calculate saturate, aromatic, resin and asphaltenes (SARA) fractions in asphalt binder extracts. The quantitative SARA of gilsonite has been analyzed by [14] and showed high asphaltenes quantity around 79.9% and low saturated and resins with 0% aromatic. The most interesting finding is the marked significant asphaltenes with no aromatic content noticed within the gilsonite, which is not usual for neat asphalt binder. The comparative SARA fractions between gilsonite [14] and asphalt binder [21] have been illustrated in Figure 2. It is shown in Figure 2 that the asphaltene and aromatic content of asphalt binder and gilsonite demonstrates their respective dissimilar physical and chemical behavior. Furthermore, according to [16], gilsonite is a mixture with a complicated broad-ranging continuum of fragments that have variable molecular weight. In gilsonite, not every molecule reflects the structure of gilsonite, but they may exhibit some similar structural characteristics.

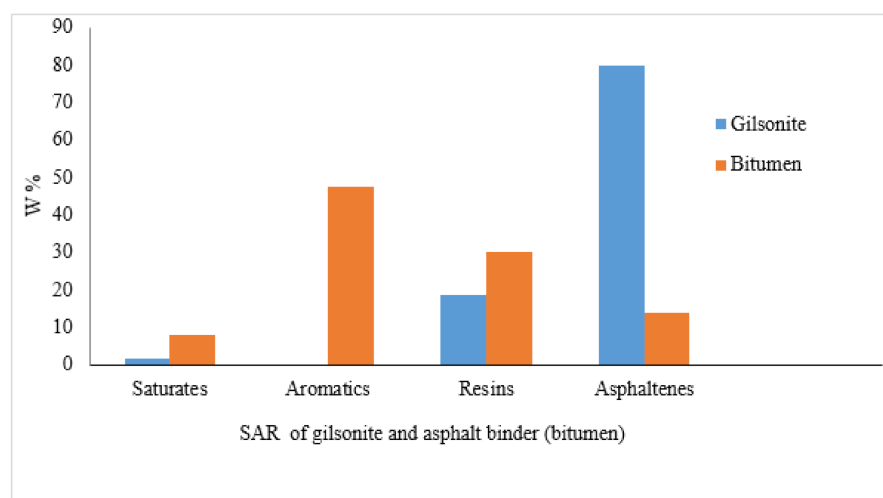


Figure 2. Difference in SARA rates between gilsonite [14] and asphalt binder (bitumen) [21].

2. Review of Research

Gilsonite is often used to harden softer petroleum products. It is usually mixed with petroleum products and refinery-produced asphalt binder to achieve a certain desired characteristic [14]. Gilsonite can be inserted into asphalt mixture by two processes: a first addition to the asphalt binder (wet process) or a second addition to aggregates (dry process) during the premixing cycle while making mixture at the batch plant [12,15]. In the typical wet process, gilsonite is used as the asphalt binder modifier, blended with asphalt binder in the blending tank for 45–60 min, at a temperature of 170–205 °C [22]. However, in the dry process, gilsonite is usually used as an aggregate substitution in the mixture rather than a specific binder modifier, blended with aggregate in the drum at the batch plant [15,22]. This paper focused on asphalt–gilsonite modification both in the wet and/or dry process in order to assess the comparative effectiveness in improving the engineering properties of asphalt in both industrial and paving applications. In addition, asphalt modified with gilsonite alone or combined with other additive materials will be discussed.

2.1. Asphalt Modified with Gilsonite as a Sole Modifier

The purpose of asphalt modification is to increase the stability and strength of mixtures, improve the cohesive strength within the mixtures, improve oxidation and resist aging and consequently reduce life cycle costs of the pavement [9]. Many researchers have paid attention to modifying the asphalt with gilsonite as a sole modifier.

Liu et al. [23] indicated that the addition of gilsonite tends to increase the rutting resistance of asphalt modified binder; however, it increases the tendency for fatigue cracking and low temperature cracking. Their study concluded that adding a small amount of gilsonite (around 3%) improved the rutting resistance of asphalt modified binder with no reduction of cracking resistance at low temperature.

Ameri et al. [12] determined that there is a direct correlation between the fraction of gilsonite and the viscosity; as the proportion of gilsonite in the mix increases, the viscosity of the modified asphalt binder increases. In their investigation, it was noted that when gilsonite increased from 4% to 12%, the modified asphalt binder performance at high temperature enhanced by extending the temperature at which the asphalt binder can sustain its elasticity. In addition, the results highlighted the positive impact of gilsonite in improving the performance of asphalt binder at high temperatures (around 80 °C) and increasing the shear strength of the modified asphalt binder. However, the BBR test showed that the gilsonite negatively affected the modified asphalt binder performance at low service temperatures (−6 & −24 °C), causing a decrease in the elasticity and an increase in the stiffness.

Babagoli et al. [24] investigated the benefits of modifying asphalt binder with gilsonite on asphalt mixtures. The results of rheological properties tests indicated a negative impact on the degree of penetration and ductility when increasing the gilsonite percentage; however, in the case of SP and viscosity of the modified binder, the impact was opposite. It is revealed that adding gilsonite leads to stiffening of the binder. The penetration index value demonstrated that gilsonite resulted in the binder being more favourable for hot climates and less susceptible to temperature. The results showed that the mixture with 10% gilsonite was the most resilient modulus, reflecting the impact of gilsonite on the mixture stiffening. In addition, mixtures without gilsonite showed a lower tensile strength value compared to mixtures with gilsonite. The results also indicated that gilsonite-modified binder had enhanced resistance to moisture damage and permanent deformation, as well as improved rutting performance. The study concluded that using gilsonite-modified asphalt binder was efficient and more effective in preparing mastic cover aggregates resulting in increased shear strength for asphalt mixture.

Djakfar et al. [25] evaluated the performance of porous asphalt mixtures using recycled concrete and investigated the impact of gilsonite addition to the mixture. The results showed that using gilsonite at ranges of 8–10% (the optimum content was 9%) increased the Marshall characteristics of the mixture, especially its stability without lessening the permeability capability of the porous asphalt mixtures. However, the study concluded that the porous asphalt mix content of recycled concrete material and gilsonite was suitable for roads with medium traffic load.

Akbari Nasrekani et al. [17] assessed the influence of using gilsonite to asphalt binder and asphalt mixtures at high temperatures. The results indicated that the SP of modified asphalt binder increased when the rate of gilsonite increased. For instance, adding 10% gilsonite showed a noticeable improvement in resistance to the flow of gilsonite-modified binder by increasing the SP by 7.8 °C. In addition, the results of DSR indicated that the complex modulus is improved significantly by adding gilsonite to the asphalt binder. It is determined that the rutting performance of the gilsonite-modified binder is improved in high temperatures. Nevertheless, increased modulus may lead to reduced performance at low and intermediate temperatures for asphalt modified binder. The DCT results showed that increasing gilsonite content increased resistance to rutting by the reduced secondary stage gradient of a creep curve. This enhancement is observed as a result of the high content of asphaltenes in gilsonite, which increased the rutting resistance of gilsonite-modified mixture.

Quintana et al. [26] carried out experimental work to investigate the impact of using gilsonite on HMA by wet and dry processes. The study has indicated that when the mixture was modified with gilsonite at ratios of 5% and 10%, a significant improvement in stiffness and mechanical strength was obtained. The results also indicated that the gilsonite generated an increase in the viscosity and SP, and a significant decrease in the penetration. The results also showed an increase in the rutting resistance and stiffness for the gilsonite-modified binder and mixture based on the DSR test and the resilient modulus test, respectively. In addition, the study indicated that the large increases in stiffness and mechanical strength were achieved when the asphalt mixture was modified by the wet process with 10% gilsonite. This increase in stiffness displays the role of gilsonite addition in enhancing the rutting resistance of the asphalt mixture at high service temperatures in hot weathers. In contrast, during the investigation, based on the BBR test, the gilsonite asphalt mixture experienced embrittlement at low temperatures (−12 °C and −18 °C), consequently reducing its resistance against low temperature fatigue cracking. Concerning the TSR value, compared with the unmodified asphalt mixtures, the gilsonite-modified mixtures had similar magnitudes, suggesting that the gilsonite had no effect on the resistance against moisture damage.

Jahanian et al. [27] reported that the addition of gilsonite to asphalt binder in HMA significantly increased the resilient modulus parameter and Marshall stability in gilsonite asphalt mixtures. Nevertheless, the resilient modulus test showed a large decrease in the

flexibility property of gilsonite asphalt mixtures, which caused pavement to be brittle when the resilient modulus increased greatly. The results from the DCT indicated that adding gilsonite increased the flow number (load cycle number) and this resulted in an increased rutting resistance. Moreover, the flow number of asphalt mixture with a content of 4.6% gilsonite was about double the flow number of HMA without gilsonite. Also, the TSR ratio results indicate that adding gilsonite to asphalt mixtures reduced the moisture sensitivity.

Tang et al. [28] demonstrated that the addition of more gilsonite benefits the elastic property due to the asphaltene content in gilsonite, which increases the polarity of asphalt, causing an increase in elastic binder. The results indicated that adding gilsonite improved the rutting resistance according to DCT and WTT. The study concluded that the gilsonite modifier serves generally as a filler that induces stiffness of the asphalt binder.

Nasrekani et al. [18] applied gilsonite as an anti-stripping additive to paving asphalt mixtures containing siliceous aggregate. The results displayed that gilsonite addition significantly improved the moisture sensitivity of mixtures with siliceous aggregates compared to those with lime aggregate mixtures. Therefore, it can be stated that gilsonite plays a role as an anti-stripping agent for siliceous aggregates. In addition, the results indicated that gilsonite asphalt modified mixtures had higher ITS values and TSR values that result in lower moisture sensitivity. During the investigation, the results indicated that for both aggregate types, a further increase in gilsonite content (more than 5%) increased ITS values but did not affect TSR values. In other words, the gilsonite dosage effect on moisture sensitivity was not significantly different between 5% and 10%. Furthermore, based on FTIR analyses, blending gilsonite with asphalt binder increased other aliphatic functional groups and asphaltenes, and resulted in an overall reduction of the sulfoxide ratio in the asphalt binder. Hence, a reduction in the sulfoxide groups and carboxylic acids of asphalt binder resulted in increased moisture resistance of gilsonite asphalt mixtures. Therefore, it is expected that mixing gilsonite with asphalt binder results in improved resistance against moisture damage.

Ameri et al. [29] investigated fatigue cracking and rutting resistance in asphalt binder modified with gilsonite. The MSCR test results, at high temperatures and high stress levels, showed that gilsonite increased elastic recovery and the rutting resistance of the base binder. For instance, modifying asphalt with 12% gilsonite gave the highest rutting resistance. The results of the LAS test indicated that gilsonite-modified asphalt binders had a better fatigue resistance at low strain levels than the base binders. However, increasing the shear strain levels improved the fatigue performance for the base asphalt binder. Ultimately, the results of the study showed that gilsonite-modified binders have better fatigue and rutting performance than the base binder.

Rondón-Quintana et al. [30] studied resistance under cyclic load and monotonic load, and moisture damage resistance in HMA when part of the coarse fraction of the natural aggregate is substituted with blast furnace slags (BFS) and used a binder modified with 10% gilsonite. The results indicated that adding gilsonite (10%) demonstrated a significant increase in asphalt stiffness (reduced penetration and increased SP and viscosity at high service temperatures). In addition, the results have shown an improvement in the moisture damage resistance when using gilsonite as an asphalt modifier. The HMA with gilsonite underwent an improvement in fatigue life; however, these improvements were not statistically significant concerning the traditional HMA. The HMA that substituted natural aggregate with BFS and used binder modified with gilsonite had increased rutting resistance in high temperature climates, and therefore its use is advised for the forming of thin asphalt layers. However, it may undergo premature cracking (brittle behavior) in low temperature climates and when used as thin asphalt layers.

Mirzaiyan et al. [15] evaluated the temperature susceptibility and performance of gilsonite and SBS-modified asphalt binders. The results of rheological and physical tests indicated that adding gilsonite to the binder can decrease the penetration grade while increasing the SP and PI. Based on RV test results, adding gilsonite to asphalt binder increased the viscosity; as the gilsonite content increased, the modified asphalt binder

viscosity also increased. The BBR test results showed that the stiffness increased when the gilsonite content increased, and this impacted on the low temperature performance grade of the modified asphalt binder. The results of the DSR test indicated that adding gilsonite to asphalt binder increased the rutting resistance of the modified asphalt binder compared to the neat asphalt binder. The results of FTIR test could be a sensible explanation for the behavior of modified binders regarding temperature susceptibility. Therefore, as a result of the presence of sulfoxide groups in gilsonite, modified binders containing higher gilsonite content have greater resistance against temperature susceptibility.

Kusuma et al. [31] evaluated the use of gilsonite additive mixed with asphalt binder. The results indicated that the penetration value of the modified asphalt inclines to decline along with a rise in gilsonite level. However, SP, flash point and density values of the gilsonite-modified asphalt binder tend to increase along with the addition of gilsonite. Furthermore, the ductility value of the modified asphalt is stable along with the addition of gilsonite; however, there was a decrease in ductility value when adding 8.5% gilsonite. Based on the result of the tests, adding 6.5%, 7.5%, and 8.5% of gilsonite to asphalt showed a better characteristic than base asphalt binder. Therefore, addition of gilsonite can improve the quality of asphalt mixture.

Zhou et al. [32] evaluated the adhesion and healing properties of asphalt binders modified with gilsonite. Regarding initial adhesion performance, the results indicated that gilsonite can enhance the asphalt binder adhesion performance; nevertheless, overdosage of gilsonite (more than 20%) had a negative effect on the binder adhesion performance, and had considerably increased the binder stiffness and reduced its flowability due to gilsonite addition. Based on the four-point bending fatigue test results, modifying binder with gilsonite indicated much higher stiffness and effective fatigue property than base asphalt in the initial fatigue test. The micro-crack healing in the gilsonite asphalt mixture is challenging after the fatigue failure point, pointing out a lower fatigue life in the second fatigue test following healing. With respect to the self-healing performance and adhesion, it is recommended to modify asphalt with 12% or 20% of gilsonite.

Sianturi and Sulaiman [33] studied the effect of gilsonite on the asphalt porous mixture by using concrete waste as the coarse aggregate. Analysis of data obtained from the testing program indicated that concrete waste as the coarse aggregate can improve the characteristics of porous asphalt mixtures, specifically stability. The stability value carried on increasing as the percentage of replacing concrete waste (such as coarse aggregate) increased. Moreover, the stability value likewise carried on increasing as the percentage of gilsonite increased. The results showed that 8.5% of gilsonite had greater stability than others. At the same time, using gilsonite reduced the voids in the mixture and increased the flow (melt) value; therefore, porous asphalt mixture without gilsonite is more rigid and susceptible to cracking. Furthermore, the optimum performance and value characteristic of the porous asphalt mixture had been obtained when using a gilsonite content of 8.5%, compared to the normal porous asphalt mixtures. Eventually, porous asphalt mixture using 8.5% gilsonite and 100% concrete waste experienced increase performance, by increasing the friction force between the vehicle wheels and road surface, increasing the resilient modulus, and decreasing the depth of the track of the vehicle wheels.

The Zuluaga-Astudillo et al. [34] study determined that the gilsonite increased the stiffness of asphalt mixture, while negatively affecting the mechanical properties when the normal aggregate is replaced with recycled aggregate. Their study concluded that gilsonite-modified asphalt binder presents the possibility to replace the normal aggregate with recycled aggregate. Moreover, this replacement improved the rutting and fatigue resistances. Replacing the aggregate by volume can be advised for asphalt layers of more than four inches in tropical environment, whereas asphalt layers of less than two inches may experience fatigue cracking in cold environments.

A summary of key experimental research work on asphalt modified with gilsonite as sole modifier has been tabulated in Table 3.

Table 3. Research work on asphalt modified with gilsonite as a sole modifier.

References	Rate of Additive	Mode of Mix	Tests Conducted	Outcomes (Effect of Gilsonite on Modified Asphalt)
[23]	3%, 6%, 9%, and 12%	wet	Superpave specifications, asphalt binder test.	Improved the rutting resistance and reduced the cracking resistance.
[12]	4%, 8%, and 12%	wet	Rotational viscosity (RV), dynamicshear rheometer (DSR), and bending beam rheometer (BBR).	Increased the viscosity, the stiffness at low temperatures and the elasticity at high temperatures.
[24]	5%, 10%, and 15%	wet	Rheological tests such as penetration, softening point (SP), ductility and viscosity. Asphalt mixture tests such as Marshall stability, indirect tensile strength (ITS), moisture susceptibility, resilient modulus test, and rutting resistance.	Improved the adhesion to aggregate, resistance to moisture damage, rutting resistance and resilient modulus.
[25]	7%, 8%, 9%, and 10%	wet	Marshall stability and flow.	Improved the stability of the porous asphalt mix.
[17]	5% and 10%	wet	SP test, DSR test, and dynamic creep test (DCT).	Increased the SP, stiffness and elasticity. Improved rutting resistance significantly.
[26]	5%, 10%, and 15%	wet and dry processes	Penetration test, SP, Marshall tests, ITS, BBR, DSR, and resilient modulus test.	Increased the SP, Marshall stability, strength, stiffness and rutting resistance. Reduced the penetration value.
[27]	In the range of 0–10% with 2% increments	wet	Marshall stability and flow, ITS, DCT, resilient modulus test, and moisture sensitivity.	Improved Marshall stability, resistance tensile stress, rutting resistance, resilient modulus and moisture damage resistance.
[28]	4, 12, 16, 20, and 24%	wet	DSR, multiple stress creep and recovery (MSCR) test, and wheel tracking test (WTT).	Increased elastic binder at high temperatures. Improved the rutting resistance.
[18]	5% and 10%	wet	ITS test, FTIR analyses.	Improved strength and moisture sensitivity.
[29]	4%, 8%, and 12%	wet	MSCR test, linear amplitude sweep (LAS) test.	Increased the rutting resistance and fatigue resistance.
[30]	10%	wet	Marshall tests, resilient modulus, ITS, permanent deformation, and fatigue resistance tests.	Increased asphalt stiffness, moisture damage resistance, fatigue resistance (slightly) and rutting resistance.
[15]	4%, 8%, and 12%	wet	Penetration test, SP, RV, DSR, and BBR.	Decreased the penetration and increased SP, viscosity, rutting resistance, stiffness and PI.
[31]	4.5; 5.5; 6.5; 7.5; and 8.5%	wet	Penetration, SP, flashpoint, ductility, specific gravity and construction temperatures (mixing and compaction).	Reduced penetration and ductility. Increased the SP, density, flashpoint and construction temperatures.
[32]	4%, 8%, 12%, 20%, and 24%	wet	Binder bond strength, FTIR, and four-point bending fatigue test.	Improved the adhesion and healing properties of asphalt modified binder.

Table 3. Cont.

References	Rate of Additive	Mode of Mix	Tests Conducted	Outcomes (Effect of Gilsonite on Modified Asphalt)
[33]	4.5; 5.5; 6.5; 7.5 and 8.5%	dry	Marshall tests, WTT, skid resistance and resilient modulus.	Increased stability, rutting resistance, skid resistance and resilient modulus.
[34]	10%	wet	Resilient modulus, fatigue resistance, permanent deformation and ITS.	Increased stiffness and rutting resistance of asphalt binder, increased fatigue resistance and reduced stiffness of asphalt mixture with recycled aggregate.

2.2. Asphalt Modified with Gilsonite in Combination with Other Additives

The modified asphalt produced should fulfil a list of criteria such as sufficient mechanical properties, storage stability, high-temperature viscosity appropriate to the road-building approaches and types of equipment, and acceptable cost, which remains of essential importance [35]. There are many additives used as modified and reinforcing material included in the asphalt mixes [36]; asphalt can be modified using two or more additives, making it challenging to understand the characteristics of modified binders [28]. Many studies have investigated asphalt modified with gilsonite and other materials.

Kök et al. [37] evaluated gilsonite as an asphalt binder modifier with styrene–butadiene–styrene (SBS) content, based on several rheological tests. The results showed that when the two modifiers are blended in the same binder, around 3–4% more gilsonite is required to substitute 1% of SBS. The results also showed that the viscosity of modified binders (when SBS was replaced with gilsonite) is continuously less than the SBS-modified binder. This denoted that adding gilsonite to SBS in the same binder can decrease the binder's viscosity. This reduction in the viscosity of the binder helps reduce the compaction energy and improves the workability of the asphalt mixture during manufacturing. The results of the DSR test indicted that adding gilsonite to the asphalt binder modified with SBS improved rutting resistance. The study suggested using gilsonite as a substitute modifier to minimize the cost of asphalt mixture production and improve the rutting resistance.

Yilmaz and Erdoğan Yamaç [38] investigated the rheological properties of bituminous binders and the mechanical properties of HMA when a combination of gilsonite and SBS were added to asphalt binder. The tests conducted in the study determined that using additives improved the rheological properties of binders resulting in asphalt-modified mixtures that have a positive impact on stability, moisture-induced damage resistance, stiffness, and fatigue life. The results of the DSR test indicated that use of gilsonite resulted in lower viscosity than SBS, and therefore gilsonite offers some advantages such as energy consumption. Analysis of data obtained from the Marshall stability and flow test revealed that using additive prompted an increase in Marshall stability values. Moreover, indirect tensile fatigue test results showed that using additive increased fatigue life, and the load repetition count required creating a (1 mm) deformation in the mixes. Tests results confirmed that 18% gilsonite or 3% SBS + 10% gilsonite were the most influential additives relative to Marshall stability and ITS values. On the other hand, 18% gilsonite, 2% SBS + 13% gilsonite, and 3% SBS + 10% gilsonite were the most influential additives against moisture-induced damage. Overall, considering all of the study's results, it can be stated that combining gilsonite and SBS provides greater advantages over utilizing gilsonite or SBS alone.

Ren et al. [39] investigated the influences of SBR on the properties of gilsonite-modified asphalt. The results of rheological tests indicated that the penetration and ductility values of gilsonite-modified asphalt decreased, while SP increased with increased gilsonite content. This finding means that gilsonite could significantly increase the high temperature properties of asphalt. Conversely, gilsonite reduced the low temperature performance

(such as cracking resistance) of asphalt due to its high stiffness characteristic, while adding SBR provided low temperature cracking resistance of gilsonite-modified asphalt effectively. Also, the results indicated that using SBR and gilsonite enhanced a high temperature performance, viscosity, and rutting resistance of asphalt binder. Furthermore, modifying asphalt with gilsonite resulted in a reduced fatigue resistance performance and compatibility, whereas adding SBR to gilsonite-modified asphalt resulted in increased fatigue resistance performance, storage stability, and compatibility. The results of FTIR and FM tests showed that, during the modification process, SBR and gilsonite interact with asphalt physically more than chemically. Generally, in consideration of storage stability and high-low temperature performance of SBR/gilsonite-modified asphalt, asphalt with 7.5% SBR and 30% gilsonite has been recommended. The study showed that gilsonite contributes to high temperature performance efficaciously, while also having an adverse impact on the low temperature performance of asphalt; the addition of SBR could resolve this problem and increase high and low temperature properties of gilsonite-modified asphalt.

Shi et al. [40] studied the rheological properties of gilsonite and nano-silica as compound additives to asphalt binder. The results of RV indicated that increased gilsonite and/or nano-silica improved the viscosity of modified binder. According to results of the DSR test, the gilsonite and nano-silica as solo modifiers or in combination together increased the rutting resistance of modified binder at high temperatures. However, there is a negative effect of gilsonite and/or nano-silica modified binder on performance at low temperatures, due to increased stiffness according to BBR test results. Therefore, the modified asphalt (with gilsonite and/or nano-silica) is more applicable for high temperature weathers, while being inappropriate for cold weather conditions.

Yalçın et al. [41] carried out experimental work to investigate the effect of using gilsonite on the storage stability of SBS-modified asphalt binder. The results of penetration tests indicated that the most stable modified binders based on storage stability were 18% gilsonite and 2% SBS + 13% gilsonite. Based on the SP test results, the SP rates of the modified asphalt binder samples obtained from the bottom of the tubes were lower compared to those of the samples obtained from the top of the tubes. In addition, the results of the viscosity test showed that 5% SBS-modified binders had the largest viscosity rates whereas 18% gilsonite-modified binders had the least viscosity rates due to their density. The results of the BBR test indicated that SBS modification had negative effects on storage stability. Nevertheless, SBS-modified binders exhibited elastic behavior at low temperatures, and vice versa with gilsonite. For that reason, combining SBS and gilsonite in the modification results in improved storage stability compared to the use of only SBS modification; also, more flexible binders were achieved compared to binders modified only with gilsonite. The results of the DSR test indicated that a major difference between the complex shear modulus of samples collected from the bottom and the top of the tubes was observed in 5% SBS, while 18% gilsonite-modified binder samples showed a minor difference between the DSR test results. Thus, using gilsonite produced stable results if compared to SBS modification. Generally, considering all conducted tests, the more stable binders (with higher storage stability) were achieved using gilsonite and SBS together, compared to using SBS solely in asphalt binder modification.

Sobhi et al. [42] evaluated the durability and mechanical performance of Warm Mix Asphalt (WMA) with an asphalt binder containing 3% Sasobit and gilsonite (composite binder). The results of rheological tests indicated that the composite binder showed improved performance compared to Sasobit-modified asphalt binder or gilsonite-modified asphalt binder. For instance, the addition of gilsonite into the asphalt binder with a content of 3% Sasobit reduced the penetration grade and increased the softening point and viscosity; also, gilsonite had better temperature susceptibility compared to the Sasobit-modified asphalt binder and the neat asphalt binder. However, further addition of gilsonite (more than 9%) decreased the resistance to moisture damage, due to the fracture mechanism and brittle behavior. Moreover, the composed binder enhanced resistance to moisture damage more than the neat binder due to gilsonite increasing the adhesion between the

asphalt binder and aggregates. Based on the Marshall tests, adding 5% and 9% gilsonite to the binder modified with 3% Sasobit increased the Marshall stability values; while increasing the gilsonite rate more than 9% made the mixtures more brittle. Furthermore, the higher the gilsonite content in the composite modifier, the higher the resilient modulus values. The addition of 9% and 13% gilsonite (along with adding 3% Sasobit) mitigated cracking resistance at low temperatures. Ultimately, the study suggested using a composite modifier made from Sasobit and gilsonite at low and intermediate temperatures, but not in colder regions. The results of the Marshall stability test, the resilient modulus test, and DCT showed that the modified mixtures provided significantly better performance in hot regions compared to conventional HMA. A summary of key experimental research work on gilsonite with other additives (all these additives were in the wet process) has been tabulated in Table 4.

Table 4. Research work on bitumen modified with gilsonite and other additives.

References	Other Additives	Rate of Gilsonite Additive	Tests Conducted	Outcomes (Effect of Gilsonite on Modified Asphalt)
[37]	styrene-butadiene-styrene (SBS)	In the range of 0–12% with 1% increment	RV test and DSR test.	Reduced the viscosity of binder modified with SBS. Improved rutting resistance.
[38]	SBS	12%, 14%, 16%, 18%, and 20%	RV, Marshall stability test, ITS Test, indirect tensile and fatigue test.	Improved the rheological properties, stability, moisture damage resistance, stiffness, and fatigue life.
[39]	styrene butadiene rubber (SBR)	5, 10, 15, 20, 30, and 40%.	Penetration, SP, DSR, ductility test, BBR, RV, FTIR and fluorescence microscopy (FM).	Improved the viscosity, rutting resistance, fatigue resistance, storage stability and compatibility.
[40]	nano-silica	2%, 4%, and 6%	RV test, DSR test, and BBR test.	Increased viscosity, rutting resistance and stiffness.
[41]	SBS	6%, 10%, and 13%	Penetration, SP, RV, BBR, and DSR.	Improved the storage stability.
[42]	Sasobit 3%	5, 9, and 13%	Dynamic creep test, ITS, resilient modulus, Marshall stability and semi-circular bending.	Gilsonite increased the moisture damage resistance; Sasobit decreased the high construction temperatures related with the use of gilsonite.

3. Critical Evaluation of Gilsonite as Asphalt Modifier

A wide range of gilsonite has been added to the asphalt binder and asphalt mixtures by wet and dry processes, respectively. Different processes and range of sources may contribute to the different performance properties of the modified asphalt mixtures. For example, Quintana et al. used high purity gilsonite and had a low carbon and sulfur content [26], compared with other studies where the gilsonite had higher carbon and sulfur contents [18].

Compared to the control asphalt mixtures, modified asphalt mixtures in the wet process provide better performance properties [22]. However, the dry process is cost-effective and it is much easier for a manufacturer to produce the asphalt mix as it does not need mixing tanks [22]. A modified wet process was used in most of the research, and the reviewed studies indicated that gilsonite asphalt pavement using the wet process exhibited good performance [17,24,30].

The methodologies have been developed and subjected to numerous research investigations. Several tests were conducted to assess the performance of the resulting products following different standards. Physical properties tests such as penetration, softening point (SP), ductility and viscosity were the common tests considered by most of the re-

viewed studies to evaluate the rheological properties of modified binders. According to the reviewed studies [17,24,30,31], adding gilsonite mitigated the penetration value and increased the softening point, ductility and viscosity values; based on these test results, modification of the asphalt binder with gilsonite can increase the quality of the asphalt mixture. However, increased viscosity of modified binders led to escalated mixture and compaction temperatures [31]. Therefore, gilsonite modification can lead to binder oxidation and decline of the design life because of reduced flexibility and cracking fatigue [43]. Therefore, adding another additive, such as waxes, to the gilsonite-modified binder was necessary to decrease the viscosity [44].

Other tests used in assessing the performance of the gilsonite-modified binder at high temperatures and at low temperatures were DSR and BBR, respectively. Based on the available literature, the gilsonite improved the performance of asphalt binder in high temperatures through increased shear strength of the modified binder [12,15,17,28,37,39,41]. However, the reviewed studies confirmed the weakness of gilsonite as a modifier for asphalt binder in terms of its properties at low temperature, due to the stiffness values of gilsonite-modified asphalts increasing with increasing gilsonite concentration [15,38,39]. It is suggested that gilsonite can be used in regions with a tropical environment to improve the asphalt binder performance. At the same time, it is recommended to avoid the use of gilsonite-modified binders in cold regions due to the brittleness of the asphalt binder resulting in pavement cracking.

The Marshall tests are considered the basic unit and the universally acceptable measurement to identify the quality of asphalt mixtures as indicated by common standards. Regarding Marshall tests, the use of gilsonite in asphalt mixtures can improve the Marshall properties of mixtures by increasing Marshall stability [24,27,30,38,42]. Moreover, the stability value continued to increase when the rate of gilsonite increased up to 10% [25,27], enhancing the resistance against deformation of the asphalt mixture [24].

Other tests were also used to assess the performance of the asphalt mixtures. For example, the indirect tensile strength (ITS) test was carried out for most of the resulting asphalt mixtures, as this test indicates the mechanical properties and moisture sensitivity of asphalt mixtures. According to the reviewed studies [18,23,30,34,38,42], mixtures with gilsonite have a greater ITS value compared to mixtures without gilsonite. This is due to the improved adhesion of aggregates to the binder as a result of the better adhesion of gilsonite-modified asphalt binder to aggregate [24]. Moreover, based on the TSR index, adding gilsonite to asphalt binder enhanced the moisture sensitivity to asphalt mixtures [24,27,30,38,42]. However, gilsonite content of more than 5% in asphalt binder did not have a significant effect on TSR index; thus, there is no effect on moisture sensitivity [18,26]. At the same time, adding more than 5% gilsonite reduced the TSR index, which had a negative impact on moisture damage due to the increase in the mixture brittleness [42]. Generally, adding up to 5% gilsonite improved the resistant moisture damage; therefore, using gilsonite to modify asphalt binder is recommended in humid regions.

The main important outcome obtained from the creep test is to show the permanent deformation which somewhat relies on the rutting resistance of asphalt mixture [27,45]. The findings from the DCT, MSCR and WTT tests showed that the asphalt mixtures prepared by binder modified with gilsonite improved the rutting resistance [17,24,27–29,31]. This improvement is suggested to be due to the high content of asphaltenes in gilsonite [14], which increased the rutting resistance of the asphalt-modified mixtures. Likewise, adding gilsonite improved the stiffness [17,26] and decreased the elasticity of asphalt-modified mixtures at low temperatures, which increased the rutting resistance. Interestingly, based on the literature reviewed, it should be noted that results from dynamic tests on asphalt mixtures and rheological tests on asphalt binders are consistent and verified similar trends seen in rutting resistance as gilsonite content increases.

According to the reviewed studies, the resilient modulus test was considered by many of the researchers [24,26,27,30,33,42] to evaluate the asphalt-modified mixtures in terms of resilient deformation. The results showed that the resilient modulus parameter increased

as the content of gilsonite in the asphalt mixtures increased. This reflects the positive influence of gilsonite on asphalt binder. However, resilient modulus test findings showed a significant drop in the flexibility property of asphalt mixtures made of binder modified with gilsonite [27]. Considerably, an overdose of gilsonite in asphalt binder led to excessive resilient modulus values, which resulted in brittle asphalt mixtures.

Since gilsonite is recognized as an additive material which contributes towards to modification of the asphalt binder and consequently improves the asphalt mixture properties, some of the research projects [37–39,41,42] have carried out further investigations combining other additives alongside gilsonite to develop the asphalt binder. Most of these investigations have decided to select SBS as an extra additive with gilsonite, due to SBS improving the HMA performance properties, and also used gilsonite to replace SBS due to SBS being an expensive additive [37,38,41]. Based on the rheological evaluation of the modified binders, using gilsonite with SBS in the same mixture decrease the viscosity of binder [37,38], which helps to increase the workability of the asphalt mixture by decreasing the mixing and compaction temperatures. Based on the reviewed studies, modification of the asphalt binder with gilsonite and SBS improved stability, ITS, moisture sensitivity and fatigue life [37,38,41]. It also improved the storage stability of asphalt-modified binder [41], thus, extending the service lives of HMA.

However, the gilsonite has a negative effect on asphalt binder at low temperatures [15,23,34,38,39]. Here, in order to improve the low temperature performance, SBR was selected as an additive to asphalt binder modified with gilsonite [39]. Based on the DSR test results obtained, the rheological properties of asphalt binder modified with gilsonite improved with addition of SBR [39]. Significantly, the addition of SBR improved the fatigue resistance performance of asphalt binder modified with gilsonite [39]. Furthermore, the results indicated that the addition of SBR increased the penetration, SP and ductility values of asphalt binder modified with gilsonite [39]. Hence, SBR enhanced asphalt binder modified with gilsonite, making it more flexible and cracking resistant which improved its low temperature properties. However, there is a negative effect that needs to be considered; the SBR increased the temperature sensitivity for gilsonite-modified asphalt binder [39]. However, it is interesting to note that adding wax improved the temperature sensitivity of asphalt binder [46–48] and can overcome this problem.

Once the amount of gilsonite that increased the mixing temperature and compaction temperature had been determined [31] (thus, avoiding this negative impact of gilsonite), Sasobit was nominated to modify the asphalt binder accompanied by gilsonite [42]. Sasobit has been used in practice mostly to decrease the construction temperatures (mixing and compaction temperatures) of asphalt modified mixture [46–48]. However, Sasobit has a negative effect on the moisture sensitivity of asphalt mixtures [42]. According to laboratory binder tests [42], it is clear that Sasobit content has a positive effect on the performance and rheological properties of WMA modified by gilsonite. It has improved the performance properties of WMA, including Marshall stability, resilient modulus, DCT and permanent deformation. Significantly, Sasobit decreased the high construction temperatures related to the use of gilsonite, whereas gilsonite increased the resistance to moisture damage compared to using Sasobit alone [42]. Considering the reviewed studies, results indicate that the application of a combination of gilsonite and other additives such as SBS, SBR or Sasobit provides more benefits than using gilsonite alone.

Most of the reviewed studies [24,27,30,38,42] focused on the Marshal mix design specifications, while few studies [12,23] were relying on the Superpave specifications test. The outcomes for both procedures indicated that modifying asphalt binder with gilsonite improved the rutting resistance of asphalt mixtures; however, it reduced fatigue life and cracking resistance at low temperatures. As a suggestion, another mix design method, “European mix design”, can be implemented. Most studies in the field of modified asphalt gilsonite have only focused on the wet process, despite the dry process being exceedingly cost-effective. According to reviewed papers, the studies focus on FTIR and TLC–FID techniques, while the XRD and SEM will be more useful for understanding the

chemical structure after modification due to the SEM images indicating the dispersion of asphalt-modified binder [49]. As a result, it can certainly support understanding of the physiochemical behaviour of the asphalt-modified binder. Many laboratory-based experiments have been accomplished to appraise the performance of the gilsonite on asphalt binder and mixture, and essentially improve the service life of asphalt pavements. However, the applications of the modified binder on the site need additional study. Although most of the available literature aimed to improve the rheological and mechanical properties, and the durability of asphalt binder and mixtures by modifying it with gilsonite, only a few research studies [27,42] mentioned the economic and environmental benefits of using gilsonite.

4. Economic and Environment Assessment

The criteria considered in this study for the assessment of economic and environmental aspects are the use of gilsonite alone and in combination with other additives in the asphalt industry.

From the economic perspective, the better quality and longer life of pavements satisfy both safety and economical aspects [35]. The use of gilsonite improves asphalt mixture resistance against tensile stresses, resulting in an increase in the service life of the pavement and a reduction in repair and maintenance costs [27]. Likewise, gilsonite is an appropriate alternative and economical choice for enhancing the properties of the binders [29]; for example, gilsonite is less expensive compared with other modifiers such as SBS or EVA (Ethylene-Vinyl-Acetate) [27]. According to the resilient modulus results, gilsonite demonstrated higher performance compared to asphalt mixtures made with pure asphalt binder [24,26,27,30,33,42]. This outcome could have a significant influence on the economic design of roads because of the need in asphalt pavements to have resilient modulus [27]. Moreover, the increased resilient modulus of the mixtures containing gilsonite may reduce the asphalt layer thickness, resulting in improved resistance to heavy traffic loads, and extended service life of the pavement.

From an environmental perspective, the asphalt industry process plays an important role in contributing to the causes associated with global greenhouse gas emissions [50]. Therefore, asphalt industries are actively searching for alternative materials and techniques in order to move towards sustainable development. Considering the amount of recycled material included in the asphalt mixture, gilsonite increases the performance of the porous HMA when using 100% recycled concrete as aggregate [25]; therefore, it is a potential material for sustainable pavement systems. However, adding gilsonite to asphalt binder decreases the penetration value and increases the SP and viscosity [17,24,30,31]. This leads to increased mixing and compaction temperatures [31,42], resulting in increased energy consumption and associated CO₂ emissions. This can be prevented by using gilsonite with an asphalt binder modified with Sasobit [42]. Furthermore, the DSR test indicated that use of gilsonite resulted in lower viscosity than SBS, and thus gilsonite offers energy consumption advantages [38]. Ultimately, the evaluation of the environmental impacts and economic feasibility of asphalt mixtures containing gilsonite indicates that adding gilsonite in combination with other selective additives provides more economic and environmental advantages. However, more research is needed to consider the economic and environmental perspectives fully; the cost-effectiveness and carbon footprint of using gilsonite as an asphalt modifier needs further investigation, since only a few publications considered this aspect.

5. Future Orientation

As the utilisation of gilsonite negatively affects the properties of asphalt mixtures at low temperatures, adding other modified materials with gilsonite is required. For instance, the polyphosphoric acid additive to asphalt binder modified with polymers increases the asphalt mixture strength by 15–18% at the temperature of −20 °C, compared with the conventional non-modified asphalt binder [51]. In addition, modification of the asphalt

binder with polyphosphoric acid and in combination with other additives has improved the resistance to low temperature cracking [52]. Furthermore, waste toner (up to 8% of the neat asphalt binder's weight) has been considered as a useful modifier to increase asphalt binder performance at low temperatures [53]. Hence, polyphosphoric acid or waste toner could significantly advance the performance of asphalt mixtures at low temperatures. Here, it is recommended that further research should be undertaken to improve the properties of asphalt mixtures modified with gilsonite at low temperatures. According to FTIR test results, there were no chemical reactions between the neat asphalt binder (asphalt binder) and gilsonite powder [18]; thus, activating the gilsonite chemically by other additives is of great importance. Based on the reviewed studies, further investigation needs to be conducted regarding ternary blended additives such as gilsonite, SBR and Sasobit, as they have improved the asphalt properties as solo or binary additives. Furthermore, adding some additive to change SARA fractions to be similar or close to the asphalt binder could lead to the discovery of a new asphalt binder in powder form, as an alternative for pelletised asphalt binder currently used in road maintenance. Moreover, gilsonite as a stable powder could be used as a coating material in the asphalt binder pelletised industry to improve the storage and stability of asphalt binder pellets. Future work should concentrate on enhancing the quality of gilsonite in asphalt mixtures by replacing the traditional limestone filler with another material, with the consideration that the gilsonite appears very poor in mineral clays [14].

6. Conclusions

According to the review of the research studies on asphalt mixtures and binder modified with gilsonite, the main conclusions are as follows:

- It was very clear that gilsonite is an important factor to improve the properties of asphalt binder and asphalt mixtures. However, according to the rheological properties of asphalt binder and the mechanical properties of asphalt mixture, gilsonite decreases the cracking resistance at low temperatures.
- According to rheological properties, gilsonite increased the construction temperatures by increasing the viscosity of the modified binder. Conversely, adding gilsonite to asphalt modified with SBS decreased the construction temperatures by decreasing the viscosity of modified binder. This indicated that using gilsonite in combination with other additive materials resulted in better performance; for instance, the best results indicated for asphalt binder and asphalt mixtures recorded using gilsonite in combination with SBR or Sasobit.
- The combination with gilsonite improves the low temperature properties (by adding SBR) and decreases the construction temperatures related with the use of gilsonite (by adding Sasobit); at the same time, the gilsonite improved the moisture sensitivity of asphalt mixtures modified with SBR and Sasobit. Therefore, these findings suggest the following opportunity for future research to include further experimental investigation about ternary blended additives (gilsonite, SBR and Sasobit).

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Abbreviations

BBR	bending beam rheometer
DCT	dynamic creep test
FM	fluorescence microscopy
FTIR	Fourier transform infrared
HMA	Hot Mix Asphalt
ITS	indirect tensile strength
LAS	linear amplitude sweep
MSCR	multiple stress creep and recovery
RV	rotational viscosity
SARA	saturate, aromatic, resin and asphaltenes
SBS	styrene–butadiene–styrene
SP	softening point
TLC-FID	thin layer chromatography–flame ionization detector in an Iatroscan instrument
WMA	Warm Mix Asphalt
WTT	wheel tracking test

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