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ENGINEERING THE EFFECT OF NANOMATERIALS ON BITUMEN AND ASPHALT MIXTURE PROPERTIES. A REVIEW

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Abstract. In recent years, several factors such as the increasing traffic loads and increasing number of vehicles have intensified the stress in pavement layers and thus reduced the service life of asphalt pavements. Today, with rising maintenance costs and traffic loads on asphalt pavements, researchers have paid more attention to diminishing defects such as cracks due to fatigue, temperature, moisture, and rutting as the most significant structural failures in asphalt pavements. The mentioned failures reduce road safety and service level during the operation period and impose huge costs on governments. In this study, we review recent research on nanotechnology applications to improve the performance of asphalt mixtures against these failures. Reviewing research suggests that different nanomaterials can improve the performance of bitumen and asphalt mixtures against cracking and rutting due to their structural properties.

Keywords: asphalt mixture, bitumen, cracking, fatigue, moisture susceptibility, nano materials, rutting.

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Introduction

The design and implementation of the asphalt layer should have the necessary conditions for asphalt pavements in terms of resistance to road traffic and weather factors. In addition, they must meet the requirements of comfortable, safe driving, and maintenance savings (Nazarko et al., 2015). However, research has shown that after 3 to 5 years of operation, most pavements will suffer from various distresses such as cracking and permanent deformation, which leads to a significant reduction in their service life (Cui et al., 2020). The occurrence of these failures, which lead to the loss of old pavement materials, is in complete contradiction with the concepts of environmental protection (Amiri, 2011). On the other hand, today, the maintenance costs of asphalt pavements have increased dramatically, accounting for a large part of the budget of countries (Hosseinian et al., 2020; Behbahani et al., 2019; Najafi Moghaddam Gilani et al., 2021).

With the increasing traffic loads that have increased the severity of failures in asphalt pavements, in recent years, researchers have paid special attention to improving the performance of this pavement against failures. One of these strategies which has long been considered is the application of additives in bitumen and asphalt mixture. In this regard, various modifiers and additives such as polymers, crumb rubbers, various waste materials, ashes, fibres, etc., have been evaluated with different purposes to change the physical and mechanical properties of bitumen and asphalt mixtures (Read & Whiteoak, 2003; Roberts et al., 1996).

Recently, however, a significant body of pavement scholars' research has focused on the impact and efficiency of applying nanomaterials on the performance of bitumen and asphalt mixtures (Faruk et al., 2014).

This application has received a lot of attention in recent years due to the ability of nanomaterials to improve the performance parameters of bitumen and asphalt mixtures (Teizer et al., 2012).

Owing to the morphological characteristics of the nanoscale, nanomaterials have unique properties such as high specific surface area, high surface free energy and high ability to disperse in bitumen and asphalt mixtures. Due to these properties, nanotechnology has had various applications in asphalt pavements, including application as an additive, crack repair, rejuvenator agent, etc. (Al-Mansoori et al., 2018; Xue et al., 2017; Ray & Okamoto, 2003). The most important compounds in bitumen are asphaltenes, oils and resins. Asphaltenes contain chemicals, which may be affected by polymers in the chemical chains. Nanomaterials can improve the behaviour of bitumen in the asphalt matrix by affecting the polymer chains at the nano level. In the literature, there are too many works that have been conducted on nano modification of bitumen. Scientists concentrate on nanotechnology knowledge to create new materials to meet better engineering requirements that conventional bitumen cannot meet these requirements (Saltan et al., 2019). Adding nanomaterials changes the rheological properties of the asphalt binder and also leads to changes in the intermolecular forces within the asphalt binder structures. This is due to the fact that as dimensions reach the nanometer level, interactions at phase interfaces become largely increased (Jamshidi et al., 2015). As the particle size decreases, a larger percentage of atoms and molecules appear on the surface. Accordingly, their surface properties become more important and dominant, and thus have a greater impact on the physical and chemical properties of the materials. Therefore, the application of these materials in various branches of civil engineering has received much attention (Iskender, 2016). This article analyses and evaluates their impact by looking at the latest efforts to improve the performance of bitumen and asphalt mixtures using nanomaterials. The article evaluates the most significant properties and failures of bitumen and asphalt mixture, including aging bitumen, which is the source of many deteriorations in asphalt mix, rutting, fatigue cracks, moisture, and low ambient temperature. Studies published from 1992 to 2021 have been reviewed, as detailed in the following sections.

1. Bitumen and asphalt mixture performance properties

Asphalt pavement as a surface that is exposed to frequent loads of heavy axes must have sufficient resistance to fatigue, cracking, creep and permanent deformations (Huang, 1992). Asphalt paving efficiency depends on various factors such as the type and grain size of aggregates, loading stresses and bitumen properties (Ameri et al., 2018). Most of the damage that asphalt pavement faces occurs mainly due to its poor performance at different temperatures. Asphalt mixture as a viscoelastic material cannot withstand high and low-temperature conditions and therefore deforms or cracks (Zhang et al., 2016). In many projects, despite the use of quality aggregates, desirable granulation, and mix design according to the regulations, we see premature failures in the asphalt mixture, mainly due to the bitumen used to construct the asphalt mixture. Bitumen has long been considered as an adhesive material in the manufacture of asphalt mix due to its good viscoelastic properties (Marandi et al., 2012). Despite the low weight percentage used in asphalt mix (4–6%), one of the main reasons for the failure of asphalt pavement is the loss of bitumen adhesion (Mansour & Vahid, 2016). After several years of use and repeated loading, bitumen becomes brittle and cracks (Yildirim, 2007). Moreover, the stiffness and strength of bitumen may decrease when exposed to frequent loads (Dai et al., 2013). The following sections examine some of the most important failures that occur in bitumen and asphalt mixtures, and reviews recent investigations conducted on the effects of nanomaterials on improving the performance of bitumen and asphalt mixtures against these failures.

2. Bitumen aging

Bitumen aging in asphalt mixture is a phenomenon that starts from the time of constructing the mix in the factory and progresses over time and eventually leads to the destruction and fragility of the mix. Aging is generally divided into short-term and long-term phases. Short-term aging is related to the amount of bitumen hardening in the asphalt construction stage when the mixture temperature is high, and long-term aging is related to the amount of bitumen hardening during the mixture service life. Oxidation-induced aging mainly includes breaking bonds in asphalt using ultraviolet (UV) radiation, destroying the molecular structure, fragility, and hardening of the asphalt, and thus affecting the performance of asphalt pavement (Chung et al., 2018).

Cadorin et al. (2021) have studied the effect of combining different percentages of nano-TiO_2 nanoparticles on the rheological properties of bitumen. Modified bituminous composites were tested with different percentages of produced TiO_2 nanoparticles (3, 6, 9, 12, and 15). The results showed that the addition of nano-TiO_2 to bitumen improved its rheological behaviour and showed more resistance to permanent deformation and fatigue. Moreover, the study showed that nano-TiO_2 created greater aging resistance of bitumen.

Akbas et al. (2021) have studied the effects of SB_2O_3 nanoparticles on the premature and long-term aging behaviour of bitumen. The results showed that adding 5% of this nanomaterial had a positive effect on short-term and long-term aging.

Shafabakhsh et al. (2020) studied the effect of SiO_2 and TiO_2 nanoparticles on the rheological behaviour of bitumen. The results showed that adding different percentages of nanomaterials to bitumen improved the rheological behaviour of bitumen and its aging resistance. The results showed that the addition of 1.2% nano-SiO_2 and 0.9% nano-TiO_2 had the best performance.

Long et al. (2020) studied the analysis of the properties of bitumen modified with nano-SiO_2. The results showed that nano-SiO_2 could reduce the sensitivity to aging and moisture damage, and the adhesion of bitumen with the addition of nano-SiO_2 due to the surface effects of its particles slightly increased.

Zheng et al. (2019) conducted a laboratory study on the performance of HEA asphalt modified with nano-TiO_2. Nano-TiO_2 additives could improve rheological performance at high temperatures and short-term thermal oxidation. Rheological properties and aging resistance of asphalt were evaluated using the dynamic shear rheometer (DSR), bending beam rheometer (BBR), and UV weathering test chamber. The results showed that the addition of Nano-TiO_2 could improve the high-temperature rheology and UV aging resistance of HEA asphalt without having an evident effect on the low-temperature rheology.

Xu et al. (2019) studied the physical properties and aging of bitumen modified with nano-ZnO powder. The addition of nano-ZnO significantly improved the anti-aging ability of bitumen against ultraviolet rays, and bitumen showed a reasonable viscosity during the aging process. In addition, nano-ZnO has strong UV absorption properties with an absorption rate of more than 95%, indicating its superiority as an anti-aging modifier for bitumen. Figure 1 illustrates how the nano-ZnO particles and bitumen react. The optimum content of nano-ZnO with modified bitumen after UV aging is reported 3%. Nano-ZnO particles possess a high surface area and many active groups on its surface. It is very likely to combine with asphalt molecules. The combination is promoted by the van der Waals force between nano-ZnO and asphalt molecules, which is a physical reaction. Surface-active groups readily

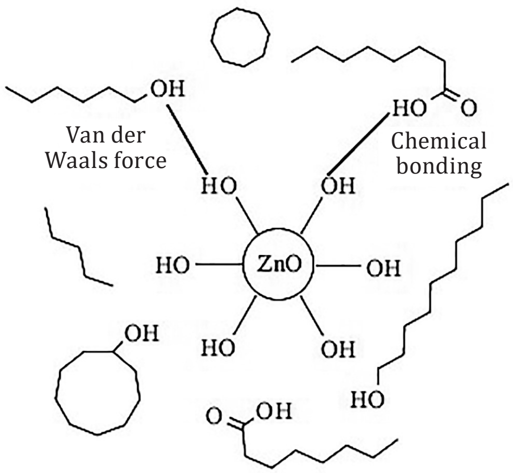

Figure 1. Action between ZnO nanoparticles and asphalt (Xu et al., 2019)

react with asphalt molecules. Then chemical bond is generated between nano-ZnO and asphalt molecules, which is a chemical reaction (Qing et al., 2018). Both physical and chemical effects increase the bonding strength between nano-ZnO and asphalt, which can increase stability of modified asphalt. Secondly, nano-ZnO particles in modified asphalt do not decompose and fill in asphalt molecules as small particles. They can increase the viscosity and decrease fluidity of asphalt by blocking asphalt molecule movement (Zhang et al., 2018).

Crucho et al. (2019a) studied the evaluation of bitumen modified with three types of nanomaterials of SiO_2, iron oxide, and clay. The results showed that bitumen modification with all three nanomaterials improved the behaviour of bitumen against aging. In terms of effectiveness, nano-clay can be selected as the most effective and then nano-SiO_2 and nano-Fe_2O_3.

Nazari et al. (2018) investigated the improvement of bitumen aging resistance using mineral nanoparticles. In this study, various mineral nanoparticles of SiO_2, TiO_2, and $CaCO_3$ were mixed with bitumen, and the chemical and microstructural properties of nanocomposites were evaluated using X-ray and electron microscopy, respectively. In addition, the effect of aging was investigated using infrared spectroscopy. The results suggested the improvement of aging resistance by adding TiO_2 and $CaCO_3$ nanoparticles to asphalt. Both rheological and chemical evaluations showed the antioxidant effect of TiO_2 and $CaCO_3$ nanoparticles, but no such behaviour was observed for bitumen containing SiO_2 nanoparticles.

Sun et al. (2017) studied bitumen modification using composite nanomaterials and polymers at high and low temperatures. The results show that nanomaterials attenuate the low-temperature performance of bitumen. Among all the tested compounds, the optimal modified bitumen formula was 0.5% SiO_2 + 5% SBR + 1PE, having the best performance of bitumen at high temperature, low temperature, and aging resistance.

Li et al. (2015) examined the effect of nano-ZnO on the aging properties of bitumen. The results of RTFO and PAV experiments showed that the mass change rate and aging index of bitumen viscosity were reduced by 3% nano-ZnO.

Yao et al. (2015) studied the analysis of bitumen aging. In this study, three nanomaterials including polymer modified nano-clay, unmodified nano-clay and Nano-SiO_2 were added to bitumen to investigate their effects on bitumen aging. The results of this study clearly suggest that these nanomaterials have anti-aging effects on bitumen.

Yusoff et al. (2014) studied the properties of polymer-modified asphalt (PMA) with adding nano-SiO_2 particles. The PG 76 PMA was mixed with nano-SiO_2 at concentrations of 0, 2 and 4wt% of bitumen. Based on the laboratory results in this study, the addition of 4% nano-SiO_2 showed the best performance to improve the aging resistance of polymer bitumen.

Conducted studies indicate that nanomaterials have had a significant impact on the rheological behavior of bitumen, improvement in aging index, increase in adhesion, positive and beneficial effects on short-term and long-term aging, decrease in moisture susceptibility and decrease in thermal sensitivity. Considering the results and beneficial effects of nanomaterials, it should be noted that the type of nanomaterials and the contents used can have significant effects on the final results. In the reviewed studies, according to the nanomaterials used, different contents, including 0.5% to 5%, have been used.

Table 1 summarises studies on the effect of nanomaterials on bitumen aging.

Table 1. Summary of studies on the effects of nanomaterials on bitumen aging

Article	Nano used	Results
Cadorin et al. (2021)	Nano-TiO_2	Nano increases aging resistance.
Akbas et al. (2021)	Nano-SB_2O_3	5% has a positive effect on short- and long-term aging.
Shafabakhsh et al. (2020)	Nano-SiO_2, nano-TiO_2	1.2% SiO_2 and 0.9% TiO_2 had the best results.
Long et al. (2020)	Nano-SiO_2	Nano reduces aging and moisture sensitivities.
Zheng et al. (2019)	Nano-TiO_2	It can increase high-temperature rheology and aging resistance.
Xu et al. (2019)	Nano-ZnO	3% can be the best performance against aging.
Crucho et al. (2019a)	Nano-SiO_2, nano-clay, nano-Fe_2O_3	The effect of nano-SiO_2 is better than nano-clay and nano-Fe_2O_3.
Nazari et al. (2018)	Nano-SiO_2, nano-TiO_2, nano-$CaCO_3$	SiO_2 had a greater effect on aging than TiO_2 and $CaCO_3$.
Sun et al. (2017)	Nano-SiO_2/polymers	Optimal formula for affecting aging: 0.5% SiO_2 + 5% SBR + 1PE.
Li et al. (2015)	Nano-ZnO	3% significantly reduces the aging index.
Yao et al. (2015)	Nano-SiO_2, nano-clay	Nanomaterials have shown good anti-aging effects.
Yusoff et al. (2014)	Nano-SiO_2/polymers	4% has the best performance.

3. Rutting in asphalt binder and mixture

One of the main failures of asphalt pavements, prevalent in hot climates, is rutting or permanent deformation (Shahin, 2006). Permanent deformation appears along the horizontal wheel path in the longitudinal surface, which decreases in pavement efficiency and makes the road surface uneven and dangerous (Golalipour, 2011). The potential of this failure in both bitumen and asphalt mix is evaluated separately by experiments. In the following, the effect of nanomaterials on the occurrence of this failure is investigated. Figure 3a shows the occurrence of this failure in asphalt pavement.

Kamboozia et al. (2021) studied the effect of using nano-SiO_2 modified bitumen on the rutting resistance of SMA mixtures. In this study, different contents of nano-SiO_2, 2, 4, and 6% were employed to modify the bitumen, and different contents of RAP, 20, 30, 40, and 50% were applied to construct asphalt samples. The results showed that the modified SMA mixtures with 6% nano-SiO_2 and 50% RAP had the best rank in terms of rutting.

Azarhoosh & Koohmishi (2020) studied the effect of styrene-ethylene/propylene-styrene (SEPS) nanocomposite as a bitumen modifier on bitumen rutting and asphalt mix using DSR and Dynamic Creep tests. The results of DSR tests showed that the addition of SEPS nanocomposites to bitumen could increase the complex modulus and decrease the phase angle. The application of SEPS nanocomposites reduced the rutting potential due to the reduction of non-recyclable creep adaptation and the increase in the percentage of elastic recovery in bitumen. Moreover, the results of the Dynamic Creep test indicated that the asphalt mixtures containing different percentages of SEPS nanocomposite showed more rutting resistance than the control mixture. In the meantime, 4% was selected as the optimal content of nano additive.

Karahancer et al. (2020) studied the effect of adding nano-Fe_2O_3 on improving the rheological properties of bitumen and the mechanical properties of HMA mixtures. The results showed that nano-Fe_2O_3 improved the rutting potential. Maximum rutting resistance was observed by 5% Fe_2O_3 modified bitumen. In addition, the HMA mixture modified with 5% Fe_2O_3 had the highest rutting resistance, with a rut depth of 6 mm.

Ghanoon et al. (2020) studied the effects of adding nano-clay, nano-lime, and SBS on the rutting of PG 64 22 bitumen. First, the bitumen was modified using 2, 4, and 6wt% of nano-clay. The results showed that the addition of nano-clay to bitumen improved the rheological properties and rutting resistance of bitumen, and the best value was obtained by 6% nano-clay. In addition, bitumen was modified using 4 and 6% nano-lime, 3% SBS, and 4 and 6% nano-clay and tested by the MSCR. Examination of the results of this composite showed that the combination of these additives (nano-clay and SBS with nano-lime) could significantly improve the rutting performance of bitumen. Furthermore, the effect of nano-lime was more than nano-clay. Finally, the best combination in this study was a combination of 3% SBS, 4% nano-clay, and 6% nano-lime.

Hu & Yu (2020) studied the extent of permanent deformations of HMA mixtures modified with thermochromic powder and nano-TiO_2 in different percentages by APA test. The results of APA test showed that the HMA mixture containing 1% red TC powder and 3% TiO_2 had the best performance in terms of rutting resistance.

Ameli et al. (2020a) studied the effects of montmorillonite nano-clay and the SBS polymer on the rutting behaviour of bitumen, as well as the effect of this modified bitumen on the rutting of SMA mixtures containing SBS polymer and fibres. The nano-clay in different percentages of 1, 2, 3, and 4wt% of bitumen was investigated. Bitumen rutting was evaluated by the MSCR test and the SMA mix rutting by wheel tracking apparatus. The results showed that the modified bitumen with 4% nano-clay and 5% SBS had the lowest Jnr and the highest recovery percentage, improving its resistance to rutting. Moreover, the modified SMA mixture with 3% montmorillonite nano-clay had the best cracking performance.

Amini et al. (2020) studied the performance of rutting, fatigue and rheological properties of Crumb Rubber Modified nano-clay (CRMN) bitumen. The results of the MSCR test showed that the addition of nano-clay to rubber bitumen decreased the permanent strain and increased the rutting resistance of rubber bitumen samples. The GTR15% + NC4% sample had the best rutting resistance, which improved the performance grade of bitumen from PG 58S 22 to PG 70H 28.

You et al. (2018) studied the evaluation of the distribution of hydrated lime nanoparticles in bitumen. The results of the DSR test showed that the high-temperature performance of the modified asphalt improved, and the rutting index increased. Nano-hydrated lime (NHL) particles stiffened bitumen and successfully introduced as a modifier to improve the bitumen performance against temperature and rutting.

Firouzinia & Shafabakhsh (2018) studied the effect of nano-SiO_2 on the thermal properties of asphalt mixtures. The contents of nanoparticles used in this study were 0, 0.2, 0.4, 0.7, and 0.9% of nano-SiO_2. The results showed that asphalt samples containing Nano-SiO_2 had a higher stiffness modulus than conventional asphalt samples. In addition, Nano-SiO_2 could improve the thermal sensitivity of asphalt mixtures, which were expected to have less rutting potential.

Al-Mansob et al. (2017) studied the performance of the epoxidized natural rubber (ENR) modified asphalt using nanoalumina (Al_2O_3) as an additive against rutting. Based on the results of the Dynamic Shear Rheometer test, the addition of nano-Al_2O_3 improved the bitumen resistance at high temperatures. The best results were recorded for modified bitumen with ENR containing 6% Al_2O_3.

Kordi & Shafabakhsh (2017) studied the effect of adding different percentages of nano-Fe_2O_3 on SMA asphalt resistance against rutting. At different temperatures and loading forces, the rut depth of asphalt samples containing 0.9% nano-Fe_2O_3 was reported about 25 to 40% lower than the rut depth of conventional ones.

Han et al. (2017) studied the evaluation of physical and rheological properties of bitumen modified with crumb rubber and nano-SiO_2. The results showed that the combination of nano-SiO_2 and crumb rubber increased the adhesion of bitumen at high temperatures. Nano-SiO_2 modified asphalt showed dynamic shear modulus, rutting parameter, and phase angle reduction, indicating that the rutting potential of the asphalt mixture could be minimized using such bitumen.

Sadeghnejad & Shafabakhsh (2017a) studied the effect of nano-SiO_2 and nano-TiO_2 on the rutting resistance of bitumen. Selected percentages of nanomaterials were 0.3, 0.6, 0.9, and 1.2wt% of bitumen. The results of DSR test showed that the modified bitumen had more rutting resistance than the standard 60 70 bitumen. Among the percentages used in this study, the use of 1.2% nano-SiO_2 and 0.9% nano-TiO_2 had the best results for the rutting resistance of bitumen. In addition, they evaluated the resistance of SMA asphalt mixtures constructed with bitumen modified with these nanomaterials. The results showed that adding nanoparticles to bitumen improved the rutting behaviour of SMA samples and the optimum content of nanoparticles to reduce the rutting in SMA mixtures was 0.9% nano-TiO_2 or 1.2% nano-SiO_2.

Ezzat et al. (2016) studied the effect of using nano-clay and nano-SiO_2 to modify the rutting behaviour of bitumen. The results showed that both nanomaterials used to improve the high-temperature performance of bitumen and especially rutting had favourable effects, and the maximum performance was achieved with 3% nano-clay and 7% nano-SiO_2.

Mubaraki et al. (2016) studied the properties of asphalt concrete modified with ASA polymer and Al_2O_3 nanoparticles at high temperatures with percentages of 3, 5, and 7%. The results showed that the addition of ASA polymer and Al_2O_3 nanoparticles had a great effect on the rheological properties of asphalt cement at high temperatures, and the mixture containing 5% Al_2O_3 had the highest value of rutting resistance among the samples.

Table 2. Summary of studies on the effects of nanomaterials on rutting

Article	Nano used	Results
Kamboozia et al. (2021)	Nan-SiO_2	6% of nano-SiO_2 and 50% RAP have the best rutting resistance.
Azarhoosh & Koohmishi (2020)	Styrene-ethylene/ propylene-styrene nanocomposite	4% has the most positive effect on the rutting resistance.
Karahancer et al. (2020)	Nano-Fe_2O_3	5% has the highest rutting resistance.
Ghanoon et al. (2020)	Nano-clay, nano-lime and SBS modifiers	4% of nano-clay, 6% nano-lime, and 3% polymer are the best combination to reduce rutting.
Hu & Yu (2020)	Thermochromic materials and nano-TiO_2	3% of nano-TiO_2 and 1% TC powder are the best combination to reduce rutting.
Ameli et al. (2020a)	SBS/montmorillonite nano-clay	The combination of 3% nano-clay and 5% SBS has shown the best rutting resistance.
Amini et al. (2020)	Nano-clay	The sample containing 4% nano-clay has had the best performance.
You et al. (2018)	Nano hydrated lime	Nano stiffens the bitumen and increases the rutting resistance.
Firouzinia & Shafabakhsh (2018)	Nano-SiO_2	Nano improves the stiffness module and rutting resistance of the mixture.
Al-Mansob et al. (2017)	Nano-Al_2O_3	The sample with 6% nano-Al_2O_3 has obtained the best result.
Kordi & Shafabakhsh (2017)	Nano-Fe_2O_3	0.9% nano-Fe_2O_3 reduces rut depth by 25 to 40%.
Han et al. (2017)	Nano-SiO_2	The use of this nano has shown the improvement of dynamic shear modulus and rutting resistance.
Sadeghnejad & Shafabakhsh (2017a)	Nano-SiO_2 and nano-TiO_2	Samples containing 1.2% nano-SiO_2 and 0.9 nano-TiO_2 have the best results.
Ezzat et al. (2016)	Nano-clay, nano-SiO_2	3% of nano-clay and 7% nano-SiO_2 are the best percentages to deal with rutting.
Mubaraki et al. (2016)	Nano-Al_2O_3	The mixture containing 5% nano-Al_2O_3 has the best rutting resistance.
Iskender (2016)	Nan- clay	This nano reduces rutting by an average of 50%.
Shafabakhsh & Ani (2015)	Nano TiO_2/SiO_2	Consumed nanomaterials significantly reduce rutting.

Iskender (2016) studied the rutting of modified asphalt mixtures with three different percentage nano-clays (5%, 3.5%, 2%) as a filler substitute. In general, it can be said that the nano-clay modified mixtures had on average 50% more rutting resistance than conventional mixtures.

Shafabakhsh & Ani (2015) used nano-TiO_2 and nano-SiO_2 particles to improve the performance of steel slag-modified asphalt mixtures (SSAM). Repeated Load Axial Test was used to determine the rutting resistance. The results showed that the addition of nano-TiO_2 and nano-SiO_2 significantly improved the rutting resistance of asphalt samples compared to conventional ones.

According to the results of studies, nanomaterials have brought positive effects, which include: increasing the complex modulus and reducing the phase angle, increasing the percentage of elastic recovery in bitumen, reducing permanent strain and increasing rutting resistance, stiffening bitumen and improving the performance of bitumen against temperature and rutting resistance, higher stiffness modulus, as well as increasing the elasticity and adhesion of bitumen compared to conventional specimens. According to the previous explanations, the contents used can be different depending on the nanomaterials used. In the conducted studies, the contents used are in the range of 0.3% to 7%.

Table 2 summarises the studies on the effect of nanomaterials on the rutting of bitumen and asphalt mixtures.

4. Fatigue

Fatigue cracks in asphalt mixes caused by frequent traffic loads are another important damage that will occur during the useful life of pavement and occur mainly at medium and low temperatures (Modarres & Hamedi, 2014). Fatigue in asphalt mixtures is a function of the chemical and rheological properties of bitumen and its aging, type, characteristics and granulation of aggregates, mix design, number and intensity of traffic loads, as well as environmental conditions (Hajj & Bhasin, 2018). In this section, the effect of nanomaterials on the fatigue resistance of bitumen and asphalt mixture is discussed. Figure 3b shows the occurrence of this failure in asphalt pavement.

Liu et al. (2021) studied the rheological changes of SBS-modified bitumen after the addition of SBS graphene nanoplatelets. In this research, bitumen modified with composite polyvinylpyrrolidone stabilized graphene nanoplatelets (PVP-GNPs) was prepared. The results showed that the optimal recommended percentage of PVP-GNPs for bitumen modification was 1.0–1.5% SBS. Adding the appropriate dose of PVP-GNP increased corrosion resistance and fatigue damage tolerance in SBS-modified asphalt.

Babagoli (2021) studied the functional properties of bitumen and stone mastic asphalt (SMA) mixtures modified with polyphosphoric acid (PPA), carbon nanotubes (CNT), and styrene-butadiene rubber (SBR). The results of LAS tests showed that the addition of SBR/PPA and CNT improved the fatigue life. In addition, the study of asphalt mixtures showed that the use of CNT improved the fatigue behaviour of the samples, and with an increase in its percentage, the fatigue life of samples also increased.

Kamboozia et al. (2021) also studied the effect of using nano-SiO_2 modified bitumen on the fatigue resistance of SMA mixtures. The results showed that modified SMA mixtures with 4% nano-SiO_2 had the best rank in terms of fatigue life. However, increasing the content of nano-SiO_2 from 4% to 6% reduced the fatigue life.

Das & Singh (2021) studied the effects of adding nano hydrated lime (NHL) as a filler on the fatigue damage rate of coarse-grained mastic asphalt mixture and compared its results with the mixture made with hydrated lime (HL) filler. The percentage of HL and NHL fillers was selected as 0, 5, 10, 15, and 20% by mass of bitumen AC-30. Fatigue factor showed that HL and NHL fillers could increase the fatigue resistance of the mixture. It can be said that the combination of two additives worked better in increasing the fatigue life of the asphalt mixture.

Badroodi et al. (2020) studied the fatigue performance of WMA mixture modified with 5% Nano-SiO_2 containing recycled asphalt pavement (RAP) materials with self-healing properties. The results showed that the addition of nano-SiO_2 could significantly improve the self-healing behaviour of WMA mixture and thus increase its fatigue life.

Amini et al. (2020) also studied the fatigue performance and rheological properties of Crumb Rubber Modified Nano-clay (CRMN) bitumen. The results of Linear Amplitude Sweep (LAS) test showed that at low strains, the presence of a polymer network in rubber bitumen increased the fatigue life; however, increasing the strain caused slipping and destruction of this polymer network. Moreover, adding nano-clay resulted in a delay in the oxidation process and improved the aging resistance, hence increasing the fatigue life of rubber bitumen.

Bhat & Mir (2020) studied the effect of using nano-Al_2O_3 as a modifier in SBS- modified bitumen. Different percentages of nano-Al_2O_3, 1, 2, 3, 4, and 5% were examined. The results showed that up to 4% application of Nano-Al_2O_3 could improve the fatigue performance of polymer bitumen. However, when the percentage of nano-Al_2O_3 increased to 5%, the polymer bitumen fatigue increased.

Motamedi et al. (2019) investigated fatigue in bitumen modified with nano-SiO_2 and synthesized polyurethane. The results showed that nano-SiO_2 and synthesized polyurethane increased the fatigue life of bitumen. Polyurethane performed better at low strain, while similar performance was reported between the two additives at high strain. The optimum amount of both additives was 7wt% of bitumen.

Kordi & Shafabakhsh (2017) also studied the impact of adding different percentages of nano-Fe_2O_3 on SMA asphalt resistance against fatigue. The results showed that the increase in fatigue life of modified samples with 0.9% nano-Fe_2O_3 was between 15 to 35% compared to conventional ones.

Sadeghnejad & Shafabakhsh (2017b) studied the indirect tensile fatigue test to estimate the fatigue life of modified SMA mixtures with different percentages of nano-SiO_2 and nano-TiO_2. The results showed that the modified SMA mixtures were more resistant to the fatigue cracking and indicated a significantly higher fatigue life than conventional ones. Based on the results of this study, it can be said that the use of 1.2% nano-SiO_2 and 0.9% nano-TiO_2 had the highest life for SMA mixtures at all temperatures and stresses.

Chelovian & Shafabakhsh (2017) studied the effect of adding nano-Al_2O_3 on the resistance of SMA asphalt mixture to fatigue. The results proved that among the percentages of 0.3, 0.6%, 0.9%, and 1.2% nano-Al_2O_3, 0.6% had the highest life for asphalt mix.

Sivakumar & Anjaneyulu (2016) studied the fatigue behaviour of nano-clay modified asphalt mixtures. According to the test results, the optimum percentage of nano-clay was 0.1wt% of bitumen. Nano-modified mixtures had a higher fatigue life for two reasons: First, the aggregates were coated with nano-modified bitumen, and this surface coating reduced the air void in the mix. Second, nano-clay modified the surface of the aggregate and caused better friction between the aggregates in the mixture. These two categories led to higher fatigue resistance.

Khattak et al. (2013) studied the fatigue of carbon nanofiber (CNF) modified asphalt mixtures. The results showed that CNF improved the adhesion between bitumen and aggregate, which led to improved fatigue life in these mixtures.

Studies in the fatigue section indicate that nanomaterials can significantly increase the corrosion resistance, the self-healing behaviour of hot asphalt mixtures, increase the friction between the aggregates in the mixture, which increases the adhesion between bitumen and aggregates and eventually increases the fatigue life. According to the results, it should be noted that the type of nanomaterials and the contents used can have significant effects on the final results, so that in most cases, increasing the contents of nanomaterials can have a negative effect. The contents used are in the range of 0.1% to 7%.

Table 3 summarises the studies on the effect of nanomaterials on the fatigue of bitumen and asphalt mixtures.

Table 3. Summary of studies on the effects of nanomaterials on fatigue

Article	Nano used	Results
Liu et al. (2021)	Graphene nanoplatelets	Nano has a favourable effect on the fatigue resistance of polymer bitumen.
Babagoli (2021)	Carbon nanotube	Nano increases the fatigue life.
Kamboozia et al. (2021)	Nano-SiO_2	4% increases fatigue life, but increasing it to 6% reduces it.
Das and Singh (2021)	Nano hydrated lime	Using this nano can increase the fatigue life.
Badroodi et al. (2020)	Nano-SiO_2	This nano increases self-healing and thus increases the fatigue life.
Amini et al. (2020)	Nano-clay	This nano increases the life of aging and fatigue.
Bhat & Mir (2020)	Nano-Al_2O_3	4% increases fatigue life, but 5% reduces it.
Motamedi et al. (2019)	Nano-SiO_2	To achieve the highest fatigue life, the optimal amount of this nano is 7wt% of bitumen consumed.
Kordi & Shafabakhsh (2017)	Nano-Fe_2O_3	0.9% increases the fatigue life by 15 to 35%.
Sadeghnejad & Shafabakhsh (2017b)	Nano-SiO_2 and nano-TiO_2	Samples containing 1.2% nano-SiO_2 and 0.9 nano-TiO_2 have the best results.
Chelovian & Shafabakhsh (2017)	Nano-Al_2O_3	0.6% is the best percentage to deal with fatigue.
Sivakumar & Anjaneyulu (2016)	Nano-clay	0.1% is the best percentage to deal with fatigue.
Khattak et al. (2013)	Carbon nanofiber	This nano increases the adhesion of bitumen and aggregates and increases the fatigue life.

5. Moisture susceptibility

One of the major causes of cracking in asphalt pavement is the loss of bitumen adhesion (Kök & Yılmaz, 2009), which depends on two environmental factors of temperature and moisture (Gorkem & Sengoz, 2009). Moisture damage in asphalt pavements is recognized as a widespread problem worldwide (Do et al., 2019). In this section, we evaluate the impact of nanomaterials on the resistance of bitumen and asphalt mixtures against these failures. Figure 3c illustrates the occurrence of this failure in asphalt pavement.

Hu & Yu (2020) studied the moisture sensitivity of HMA mixture modified with thermochromic powder and nano-TiO_2 in different percentages through the IDT test. The test results showed that the mixture of HMA modified with TC powder and nano-TiO_2 showed higher moisture resistance than the asphalt sample modified with single TC powder.

Ameli et al. (2020b) investigated the effect of adding polyphosphoric acid (PPA), carbon nanotube (CNT) and SBR on the moisture sensitivity of SMA samples. In this study, two types of bitumen, AC 60/70 and AC 85/100, were utilized. The percentages of additives in this study were 0wt%, 0.5wt%, 1wt%, 1.5wt%, and 2wt% of bitumen for CNT, 0.5wt% of bitumen for PPA, and 2wt% of bitumen for SBR. The results showed that the application of SBR/PPA increased the elastic modulus (M_r) and indirect tensile strength (ITS), measured as a criterion for improving the performance of asphalt mix against moisture sensitivity. Moreover, with the increase of CNT percentage, ITS energy continued to maintain its increasing trend, indicating the effect of this nano on the moisture sensitivity of the mixture.

Razavi & Kavussi (2020) studied the effects of various nanomaterials, including nano-$CaCO_3$, nano hydrated lime, nano-bentonite, and nano-SiO_2, on the moisture sensitivity of asphalt mixtures. The results showed that adding 4% of nano hydrated lime increased the TSR values of the mix by 61%. Furthermore, by adding 4% nano-$CaCO_3$, the TSR of the mixture increased by 59%. As a result, in this study, the application of low percentages of nanomaterials was suggested instead of conventional fillers such as hydrated lime to increase moisture resistance. In addition, among the nanomaterials, nano hydrated lime had the highest efficiency.

Taherkhani & Tajdini (2019) studied the comparison of the effects of nano-SiO_2 (NS) and hydrated lime (HL) on the asphalt concrete properties. The results showed that NS was more effective than HL in improving resistance to freeze-thaw cycles. Moisture resistance was higher in NS-modified mixtures than in mixtures containing HL.

Sezavar et al. (2019) investigated the moisture sensitivity of modified asphalt mixture with different percentages of SiO_2 nanoparticles (0, 0.2, 0.4, 0.7, and 0.9wt% of bitumen). The results showed that the use of SiO_2 nanoparticles significantly increased the tensile strength. In terms of bitumen type, the effect of SiO_2 nanoparticles on bitumen 85–100 was less than bitumen 60–70.

Omar et al. (2018) studied the effects of water infiltration on the asphalt mixture made with ordinary bitumen 60/70 and modified with 2 and 4% nano-clay. The results showed that the addition of nano-clay improved the resistance of mixes to moisture damage. In the meantime, bitumen containing 4% nano-clay had better performance.

Zakerzadeh et al. (2018) studied the effect of adding super hydrophobic nanomaterials to the asphalt mixture. The results of examining the contact angle of aggregates showed that hydrophobic nanomaterials had a significant effect on the contact angle of aggregates, and the reason for the increase of angles was the presence of nanomaterials on the surface of aggregates. Modified asphalt with nanomaterials reduced the moisture sensitivity of the mixture by an average of 85%. The cause of this phenomenon is the presence of nanomaterials that prevent water from penetrating the surface of the particles and prevent the formation of ice crystals in the mixture. Figure 2 illustrates the effect of nano on asphalt.

In the hydrophobicity industry, hydrophobic materials are divided into two general categories of water and solvent bases. The hydrophobic bases of water are dissolved in solutions that are soluble in water, and solvent bases of hydrophobic solvents are soluble in solutions whose solvents are hydrocarbon solvents (such as thinner and acetone) (Feng & Jiang, 2006). Hydrophobicity of the asphalt samples with hydrophobic

a) **b)**

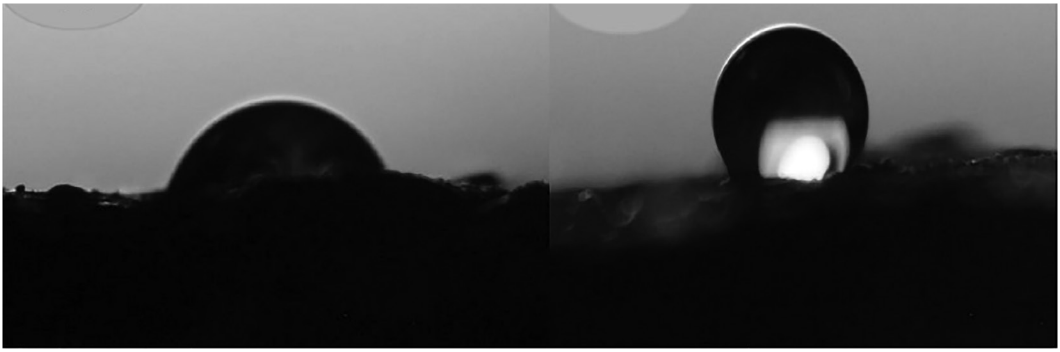

Figure 2. An example of droplets formed on the surface of an asphalt mix: a) Unmodified asphalt; b) Modified asphalt (Zakerzadeh et al., 2018)

coating materials is such that, first, they dissolve the raw material with a suitable solvent (water or hydrocarbon solvents); then, the obtained solution is sprayed on its surface. The hydrophobic process is such that after spraying hydrophobic materials on the surface, time should be allocated to the solvents to evaporate and form arrays of nanoparticles on the surface by chemical bonding. The hydrophobic surface should not be exposed to any material for 24 h until the chemical bond with the coating surface of the pavement is completed. The amount of hydrophobic material coating sprayed on the surface 12 g for asphalt sample and also the size of a droplet of one micro liter (using a Hamiltonian needle) for all asphalt samples were considered (Zakerzadeh et al., 2018).

Yang et al. (2018) investigated the effect of nano-clay and microfiber on the moisture failure of asphalt mixtures by non-chloride deicers. The percentages used for carbon microfiber were 1wt% of bitumen and for nano-clay at 3%, 0.6wt%, 1.2wt%, and 1.8wt% of bitumen. The results showed that the integration of nano-clay and carbon microfiber in asphalt concrete significantly improved the tensile strength of the mixture and reduced its moisture sensitivity, while the addition of carbon microfiber alone increased the moisture sensitivity.

Abandansari & Modarres (2017) studied the effects of using SBS nanocomposite on the moisture sensitivity of hot mix asphalt. The results showed that the addition of SBS nanocomposite increased the adhesion and cohesion of the mixture and did not allow the bitumen to move quickly on the surface of the aggregates in the presence of water and made the mixture more resistant against moisture after freeze-thaw cycles.

Golestani et al. (2015) studied the effect of polymer and nano-clay performance on moisture sensitivity of asphalt mixtures. The results of the moisture sensitivity test showed that modified asphalt mixtures with polymer and nano-clay were more resistant to moisture damage than conventional asphalt mixtures.

Ameri et al. (2013) investigated the moisture sensitivity of asphalt mixtures under freeze-thaw cycles before and after adding two additives, nano-zycosoil and hydrated lime slurry with two different types of aggregates. The results suggested the improvement of the indirect tensile strength of the mixtures containing both additives, which indicate the improvement of the resistance of these mixtures against moisture sensitivity. Meantime, nano-zycosoil increased the adhesion between aggregates and bitumen, reported to reduce the moisture sensitivity of the asphalt mixture.

Goh et al. (2011) studied the potential benefits of micro- to nano-sized materials for asphalt mixtures, especially when exposed to water or frost. The results showed that the addition of nano-clay and carbon microfiber reduced the moisture sensitivity of the asphalt mixture, and the modified asphalt mixtures with 1.5% nano-clay had the best performance in this regard.

According to the results of studies in the moisture section, nanomaterials have brought positive effects, which include: increasing the elastic modulus (Mr), indirect tensile strength (ITS), increasing the cohesion of the mixture, improving freeze-thaw resistance, a significant increase in tensile strength, preventing water penetration to the surface of the mixture particles, preventing freezing and reducing moisture susceptibility. The contents used are in the range of 0.2% to 4%.

Table 4 summarises the studies on the effect of nanomaterials on the moisture sensitivity of bitumen and asphalt mixtures.

Table 4. Summary of studies on the effects of nanomaterials on moisture sensitivity

Article	Nano used	Results
Hu & Yu (2020)	Thermochromic materials and nano-TiO_2	TC and nano-TiO_2 together have a good moisture resistance for the mixture.
Ameli et al. (2020b)	Carbon nano tube	5% had the best effect on the moisture sensitivity of the mixture.
Razavi & Kavussi (2020)	Nano $CaCO_3$, nano hydrated lime, nano bentonite and nano-SiO_2	The use of these nanomaterials with low percentage instead of fillers is recommended to reduce moisture sensitivity.
Taherkhani & Tajdini (2019)	Nano-SiO_2 and hydrated lime	Nano-SiO_2 is more effective than hydrated lime in moisture sensitivity.
Sezavar et al. (2019)	Nano-SiO_2	Significantly increases tensile strength (improves moisture resistance).
Omar et al. (2018)	Nano-clay	Samples with 4% of this nano had an acceptable performance against moisture sensitivity.
Zakerzadeh et al. (2018)	super hydrophobic nanomaterials	It is very effective in hydrophobicity of the mixture.
Yang et al. (2018)	Nano-clay and carbon microfiber	Both substances together reduce moisture sensitivity, but carbon microfiber alone increases moisture sensitivity.
Abandansari & Modarres (2017)	SBS nanocomposite	Increases the adhesion and cohesion of the mixture and reduces moisture sensitivity.
Golestani et al. (2015)	Polymer and nano-clay	The combination of polymer and nano-clay reduces moisture sensitivity.
Ameri et al. (2013)	Nano-organosilane	Increases adhesion and decreases moisture sensitivity.
Goh et al. (2011)	Nano-clay and carbon microfiber	1.5% nano-clay has the best performance.

6. Thermal cracking

Climate change and environmental deterioration have led to severe temperature fluctuations over the past years (Han et al., 2018). One of the failure types in asphalt pavements, especially in cold areas, is low-temperature cracks, which vast sums are spent on pavement repair after their occurrence. The two main parameters in the onset and propagation of cracks are traffic loads and thermal stresses (Wei et al., 2020). In the following, we review several articles evaluating the impact of nanomaterials against this significant failure. Figure 3d shows the occurrence of this failure in asphalt pavement.

Mahani et al. (2021) studied the effect of nano calcium carbonate (NCC) on the fracture behaviour of asphalt mixtures in different fracture modes. In this experiment, two types of bitumen PG 64-22 and PG 58-28 modified with 1%, 3%, 5%, and 7% NCC were employed. The results showed that NCC had a significant effect on fracture toughness. A closer examination revealed that the values of 3.62, 4.17 and 6.29% of NCC had the best values for the fracture toughness of the asphalt mixture for different loading modes.

Fakhri & Mottahed (2021) investigated the improvement of fracture resistance of warm mix asphalt (WMA) mixture containing RAP and nano-clay additive. The fracture toughness of semicircular samples with 0, 20, 30, 40% RAP, and 0 and 3% nano-clay were studied at 10 and 20 °C by the SCB test. The results showed that the use of 40% RAP with 3% nano-clay in WMA not only brought environmental and financial benefits, but also improved the performance of the mixture against low-temperature cracking.

Shafabakhsh et al. (2021) studied the effect of nano-SiO_2 on the occurrence of low-temperature cracks in asphalt mixtures. In this study, the semicircular bending (SCB) test was performed at different temperatures of 5 °C, 15 °C, and 25 °C. The results showed that vertical and angular cracks were significantly improved by adding 1.2% Nano-SiO_2 at all temperatures.

Hasaninasab et al. (2019) studied the performance improvement of modified asphalt mixtures with Forta fibres and nano-zycotherm against thermal cracking through the SCB test. The results of this study showed that the resistance of the modified asphalt mixture with nano-zycotherm additives and Forta fibres against this failure was much higher than the mixture without them. The reason is that the presence of nano-zycotherm increases the density, and the presence of Forta fibres increases the tensile strength of asphalt. Simultaneous application of these two substances has a great effect on increasing the self-healing ability of the mixture.

The results of studies and investigations in this section indicate that nanomaterials improve the low-temperature cracking performance of the mixture, increase the tensile strength of asphalt, and increase the fracture toughness of the mixture. As mentioned in the previous sections, the contents used can be different according to the type of nanomaterials. The contents used are in the range of 1% to 7%.

Table 5 summarises studies on the effect of nanomaterials on thermal cracking of asphalt mixtures.

Table 5. Summary of studies on the effects of nanomaterials on thermal cracking

Paper	Nano used	Results
Mahani et al. (2021)	Nano-$CaCO_3$	3.62%, 4.17%, and 6.29% are the best percentages of this nano for the fracture toughness parameter.
Fakhri & Mottahed (2021)	Nano-clay	3% nano-clay and 40% RAP improve low-temperature crack resistance.
Shafabakhsh et al. (2021)	Nano-SiO_2	1.2% nano-SiO_2 improves the cracking performance of the mixture.
Hasaninasab et al. (2019)	Nano-zycotherm/ Forta fibres	Simultaneous application of these two materials improves the self-healing ability of the mixture.

7. Cost considerations

The production of nanomaterials and the asphalt binder modification are important phases where exposure can be significant that one of the most likely routes for exposure is inhalation, ocular, and dermal adsorption (Ramachandran et al., 2011). There are concerns about the safety of nanoparticles and the potential health risks associated with them. For example, inhalation of nano-TiO2 with a concentration of 8.8 mg/m3 causes lung inflammation. Due to their nano dimensions, they can easily pass through biological systems such as human skin and cell membranes and accumulate in undesirable locations to toxic levels (Grassian et al., 2006). The safety handling protocols should be implemented accordingly: modified asphalt binder in laboratory using a fume hood cabinet and individual protections; nitrile gloves at least 0.5 mm thick, mask for eye protection, breathing mask with particle-filter FFP3, and protection suit.

Regarding the cost of nanomaterials, it should be noted that although the use of nanomaterials has a higher cost than normal asphalt, it can

Figure 3. Occurrence of failure in asphalt mixtures

be very useful and effective in the long run due to the reduction of pavement maintenance costs. Generally, due to the costs associated with their synthesis, the price of the nanomaterials is highly dependent on the particle size range and specific surface area. Products with high purity and narrow size range and high specific surface may demand higher processing efforts, thus having higher final costs (Crucho et al., 2019b).

Conclusion

In recent years, the constant increase in traffic and vehicle traffic and the high cost of road maintenance have led to large sums of money being allocated from the national budget to road maintenance. In recent decades, specialists and road engineers with ongoing studies, tests, and efforts have sought to increase the pavement life and reduce road maintenance costs. One of the most significant and well-known additives used to improve bitumen and asphalt mixtures are nanomaterials, the most prominent of which are nano-SiO_2, nano-clay, nano-lime, nano-carbon. This research has conducted a comprehensive review of many investigations done in recent years on nanomaterials and their impact on various failures such as bitumen aging, rutting, fatigue of bitumen, mixtures, and the mixture resistance to moisture sensitivity and temperature. The main results of these studies are summarised as follows:

- The use of nanomaterials has had a very high impact on improving the rheological behaviour of bitumen and its resistance to aging. The most effective types of nanomaterials in this section are nano-SiO_2, nano-TiO_2, nano-clay, and nano-ZnO.
- The use of nanomaterials in paving significantly increases the strength and adhesion between bitumen and aggregates and can increase the pavement resistance against rutting and fatigue. Nanomaterials such as nano-clay, nano-SiO_2, nano-lime and nano-Al_2O_3 have had the most positive effect on the rutting and fatigue of bitumen and asphalt mixtures.
- Another positive feature of nanomaterials has been their high impact on the moisture resistance of asphalt mixtures. The use of nanomaterials can reduce the moisture sensitivity of the mixture and increase its life. Nano-SiO_2, nano-clay, and nano-TiO_2 alone and in combination with polymers have had the best performance in this area.
- Another advantage of nanomaterials is the improvement of the low-temperature performance of asphalt mixture, which decreases performance of bitumen and aggregates at these

temperatures lead to cracking in the asphalt mixture. Effective nanomaterials in this field include nano-SiO_2, nano-clay, and nano-$CaCO_3$.

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