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ORIGINAL RESEARCH PAPER

# Evaluation the Rheological Behavior of Asphalt Binder, Fracture Resistance and Moisture Susceptibility of Asphalt Mixtures: Before and After Adding Nano $Fe_2O_3$

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**Abstract**
The occurrence of distresses such as rutting, fatigue, thermal and moisture cracks in asphalt pavements has caused that a significant part of the country's budget is spent annually on the repair and rehabilitation of road pavements. Since it has been proven that the combination of bitumen and aggregates alone cannot provide optimal performance in all loading and weather conditions, the need to modify the asphalt mixture is felt more than ever. In recent years, nanotechnology has become very popular due to its unique properties in improving the properties of bitumen and asphalt mixtures. To this end, the objective of this research is the application of nano-$F_2O_3$ in improving the performance of bitumen and asphalt mixture against common distresses such as rutting, fatigue, thermal and moisture cracks to suggest a suitable solution based on the results for reducing the probability of the occurrence of these failures in asphalt pavements. In order to achieve the objectives of this research, bitumen was mixed with different percentages of nano-$F_2O_3$ and subjected to rheological tests such as Dinamic shear rheometer (DSR), Bending beam rheomether (BBR), Muiliple stress creep recovery (MSCR), and Linear amplitude sweep (LAS). The results of this section indicated that the application of 1% nano-$F_2O_3$ compared to other percentages brought the greatest improvement for the rheological behavior of bitumen. Hence, to measure the resistance of asphalt mixture against thermal and moisture cracking, control asphalt specimens (bitumen without additives) and specimens modified with 1% nano were manufactured, and these specimens were subjected to semicircular bending (SCB) and indirect tensile strength (ITS) tests. The results of this research indicated that adding 1% nano-$F_2O_3$ can improve the fracture toughness of asphalt mixtures from about 8 to 19%. Moreover, replacing 1% of bitumen with nano-$F_2O_3$ improves the moisture susceptibility of asphalt mixtures by 9% compared to control specimens.

**Keywords** HMA · Binder · Rheology · Nano $Fe_2O_3$ · Fracture toughness · Moisture

## 1 Introduction

The development and increase in the amount of public transportation cause more load on the axis and also more pressure on the roads [1]. Attention to this point increases the demand for the application of stronger and more durable asphalt mixes, which creates more demand for effective research and investigation in this field and the application of new technologies and materials produced with this technology [2]. On the other hand, the weaknesses in the components of the asphalt mixture, especially asphalt binder, cause it to experience a drop in performance in different temperature and weather conditions, which can lead to distresses such as rutting, fatigue, thermal and moisture cracks in asphalt mixture [3]. This study has focused on the asphalt binder properties, fracture resistance and moisture susceptibility of asphalt mixture.

One of the major causes of cracking in asphalt pavement is the loss of bitumen adhesion, which depends on two environmental factors of temperature and moisture. Also, climate change and environmental deterioration have led to severe temperature fluctuations over the past years [4].

One of the failure types in asphalt pavements, especially in cold areas, is low-temperature cracks, which vast sums

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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 [5].

Moisture damage in asphalt pavements is recognized as a widespread problem worldwide. This process is divided into physical and mechanical processes. The physical processes that are included as important contributors to moisture induced damage are (I) weakening of the mastic and the aggregate–mastic bond due to molecular diffusion of moisture and (II) weakening of the mastic from an erosion process of the mastic, caused by water flow (Fig. 1a). The mechanical damage process that is identified as a contributor to moisture damage is the occurrence of intense water pressure fields inside the mix caused by traffic loads and referred to as 'pumping action' (Fig. 1b) [6].

Given the failures in asphalt binder and asphalt used in roads, it is necessary to conduct comprehensive research on the modification of asphalt binder behavior, which is a determining factor in the behavior of asphalt mixtures.

## 2 Literature Review

Many studies have been carried out on the application of nano additives to modify the behavior of asphalt binder and asphalt mixture.

Akbari et al. [7] have studied the effect of different contents of nanographene oxide (GO) to improve the low-temperature cracking and high-temperature rutting resistance of hot-mix asphalt (HMA).Outcomes of the SCB fracture test indicated that nanoGO enhances the stress intensity factor (SIF) of the asphalt mixture and improves the low-temperature cracking resistance of asphalt specimens. Moreover, the addition of nanoGO to the asphalt mixture increased rutting resistance and decreased rutting depth.

Fakhri and Shahryari [8] investigated the effects of Nano Zinc Oxide (ZnO) and Nano Reduced Graphene Oxide (RGO) on moisture susceptibility resistance of Stone Mastic Asphalt (SMA) mixtures. The pure asphalt binder was modified with three content of Nano ZnO and Nano RGO (0.2%, 0.4%, and 0.6% by weight of asphalt binder). The mechanical test results showed that an increase in the percentage of Nano ZnO and RGO lead to a rise in Marshall Stability, indirect tensile strength, accumulated strain, pull-off adhesion strength, and fracture energy and improve the asphalt binder coating on aggregates in SMA mixtures. According to calculated moisture susceptibility indices of the mentioned tests above, the Nano ZnO and Nano RGO can significantly improve the resistance of stone mastic asphalt mixtures against moisture susceptibility.

Cadorin et al. (2021) [9] have explored the effect of the combination of different percentages of $TiO_2$ nanoparticles on the rheological properties of asphalt binder. Modified bituminous composites with different percentages of nano-$TiO_2$ composition (3, 6, 9, 12, and 15) were produced and tested. The results indicated that adding nano-$TiO_2$ to asphalt binder improves its rheological behavior and shows more resistance to permanent deformation and fatigue. Moreover, it was proven in this research that nano-$TiO_2$ provides higher aging resistance of asphalt binder.

Long et al. (2020) [10] have studied and analyzed the properties of asphalt binder modified with nano-$SiO_2$. The results indicated that nano-$SiO_2$ can reduce the aging sensitivity and moisture damage. Moreover, with the addition of nano-$SiO_2$, the adhesion of asphalt binder increases slightly due to the surface effects of its particles.

Karahancer et al. (2020) [11] have examined the effect of adding nano-$Fe_2O_3$ on improving the rheological properties of asphalt binder and the mechanical properties of the HMA mixture. The results indicated that nano-$Fe_2O_3$ improves the rutting potential. Maximum rutting resistance was observed in asphalt binder modified with 5% nano-$Fe_2O_3$. Moreover, the HMA mixture modified with 5% nano-$Fe_2O_3$ was reported to have the highest rutting resistance with a 6 mm rut depth.

Shafabakhsh et al. (2020) [12] have investigated the effect of nano-$SiO_2$ and nano-$TiO_2$ on the rheological behavior of asphalt binder. The results indicated that adding different

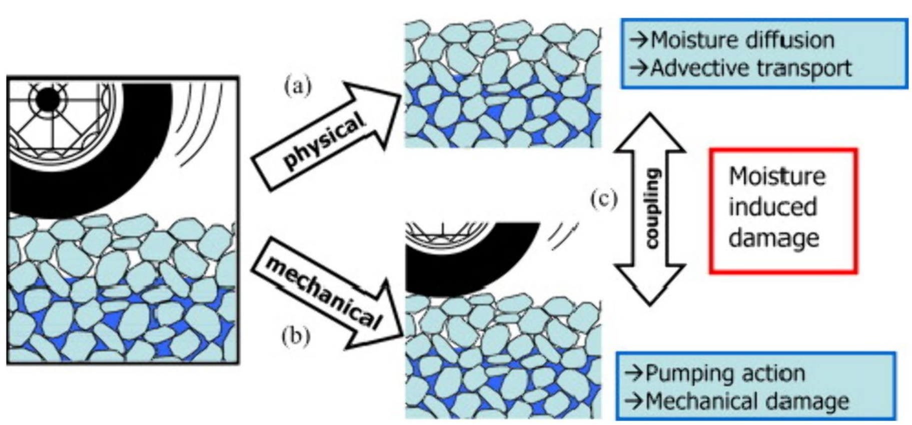


**Fig. 1** Schematic moisture damage in asphalt mixture [6]

percentages of nanomaterials to asphalt binder has improved the rheological behavior and aging resistance of asphalt binder. The obtained results revealed that the addition of 1.2% nano-$SiO_2$ and 0.9% nano-$TiO_2$ have had the best performance.

Xu et al. (2019) [13] have evaluated the physical properties and aging of asphalt binder modified with nano zinc oxide (ZnO) powder. The addition of nano-ZnO can improve the anti-aging ability of asphalt binder against UV rays and make the asphalt binder show a reasonable viscosity during the aging process. Furthermore, nano-ZnO has strong UV absorption properties with an absorption rate of more than 95%, indicating its superiority as an anti-aging modifier for asphalt binder. The results of this research indicated that the reasonable percentage of nano-ZnO with modified asphalt binder after UV aging is 3%.

Crucho et al. [14] have studied the evaluation of the asphalt binder modified with three types of nanomaterials: nano-$SiO_2$, nano-$Fe_2O_3$ and nano-clay. The results indicated that the asphalt binder modification with all three nanomaterials improved the behavior of asphalt binder against aging, and in terms of effectiveness, Nano clay can be selected as the most effective, followed by nano-$SiO_2$ and nano-$Fe_2O_3$.

Firouzinia and Shafabakhsh [15] examined the effect of Nano-$SiO_2$ on the thermal properties of asphalt mixture. The nano percentages employed in this research are 0, 0.2, 0.4, 0.7, and 0.9% of Nano-$SiO_2$. The results indicated that the asphalt specimens containing Nano-$SiO_2$ have a higher stiffness modulus than conventional specimens. Furthermore, Nano-$SiO_2$ can enhance the temperature susceptibility of asphalt mixtures, which are expected to have less rutting potential.

Kordi and Shafabakhsh (2017) [16] have investigated the effect of nano-$Fe_2O_3$ additive on the mechanical properties of stone mastic asphalt mixture. They added different percentages of nano-$Fe_2O_3$ to asphalt binder 60–70 by the wet method and utilized the modified asphalt binder in producing stone mastic asphalt mixtures. The results indicated that utilizing 0.9% nano-$Fe_2O_3$ can significantly improve the life of asphalt mixtures and their stiffness modulus. Another result of their research was a significant reduction in the rate of permanent deformation of stone mastic asphalt mixtures due to the addition of different percentages of nano-$Fe_2O_3$.

Hamedi et al. [17] have evaluated the effect of nanoparticles as an antistrip agent on the moisture damage of HMA. Two types of aggregates were evaluated in this study with different sensitivities against moisture damage (limestone and granite aggregate) and the asphalt binder with 60/70 penetration grade and nano zinc oxide (ZnO) in two different percentages by weight of the asphalt binder. The results showed that the ratio of wet/dry values of indirect tensile strength for the mixtures containing nano ZnO for two types of aggregate were higher than the control mixtures. In addition, the results of the SFE method showed that adding nano ZnO increased the total SFE of the asphalt binder, which led to better coating of the aggregate with asphalt binder.

Shafabakhsh et al. [18] added different percentages of nano-$TiO_2$ to asphalt binder and asphalt mixtures with a diameter and height of 40 and 100 mm by the wet method. They performed a fatigue test by the indirect tension method at different temperatures and stresses. They found that nano-$TiO_2$ prevented tensile and vertical cracks caused by tensile stresses and prevented their propagation and expansion. The results obtained from this research indicate that replacing 1% of the weight of asphalt mixtures with nano-$TiO_2$ is the best amount of this material, improving the fatigue performance of asphalt mixtures even at high temperatures and stresses.

Sezavar et al. [19] have investigated the moisture susceptibility of asphalt mixture modified with different percentages of nano-$SiO_2$ (0, 0.2, 0.4, 0.7, and 0.9% by weight of asphalt binder). The results indicate that using nano-$SiO_2$ significantly increases the tensile strength. In terms of asphalt binder type, the effect of nano-$SiO_2$ on asphalt binder 85–100 has been less than on asphalt binder 60–70.

The results of previous investigations have indicated that oxidizers are employed to increase the stiffness of asphalt binder and asphalt mixture and, as a result, prevent the kind of damage that occurs due to the softness of asphalt at high temperatures (such as rutting under the wheels). However, the stiffening of asphalt as a result of the addition of oxidizers increases the risk of increasing thermal cracks and fatigue. Therefore, in the present research, to improve the rutting resistance of asphalt binder, as well as to prevent the occurrence of the stiffening phenomenon and reduce the fatigue life, the oxidizer with the size of nanoparticles (Nano-$Fe_2O_3$) is used to improve the asphalt binder performance problem at different low, medium, and high temperatures. Therefore, different percentages of Nano-$Fe_2O_3$ were used in this research. These percentages were mixed with asphalt binder 60–70 under the same mixing conditions and subjected to physical and rheological tests. Investigating the effect of Nano-$Fe_2O_3$ on thermal and moisture cracks of asphalt mixture is one of the other parts of this research.

## 3 Material and Methods

### 3.1 Material

The aggregates used in this research are limestone aggregates. The gradation is the average of the aggregate size recommended in the Iran Highway Asphalt Paving Code (No. 234) with a maximum nominal size of 19 mm for the Topeka layer, which is presented in Table 1. The gradation test of aggregates was conducted according to the AASHTO-T27

**Table 1** Aggregate gradation used in this study

| Sieve size | Minimum | Maximum | Percent Passing |
|---|---|---|---|
| 19 mm | 100 | 100 | 100 |
| 12.5 mm | 90 | 100 | 93.3 |
| 4.75 mm | 44 | 74 | 46.8 |
| 2.36 mm | 28 | 58 | 34.5 |
| 0.3 mm | 5 | 21 | 10.5 |
| 0.075 mm | 2 | 10 | 4.3 |

standard. The consumed asphalt binder is asphalt binder 60–70. Its specifications are mentioned in Table 2. In addition, the specifications of nano-$Fe_2O_3$ used in this research are illustrated in Table 3. Moreover, the image of nanomaterial used can be seen in Fig. 2.

**Fig. 2** Nano $Fe_2O_3$ used in this study

### 3.2 Laboratory Program

In the present research and in the continuation of the investigations on adding nanomaterials to asphalt binder and asphalt mixture carried out by the researchers of this research [12, 16, 18, 20, 21], common mixing process has been employed. In order to mix, asphalt binder was first heated to 150 °C, and the nanoparticle-solvent mixture was slowly poured into the mixer for half an hour with regular time intervals. The mixing process continued at 4000 rpm until a homogeneous mixture of asphalt binder and nanoparticles was obtained. The selection of the most optimal homogeneous mixture was based on the physical experiments conducted on the control and modified asphalt binder. In order to more clearly show the evolution of the nano particles effects on asphalt binder microstructures, Fig. 3 illustrate the Atomic Force Microscopy (AFM) view of the unmodified and nano modified sample. Due to the same mix design, any change in their surface roughness would be a result of the dispersion of nano $Fe_2O_3$ particles. Of note, incorporation of nano particles resulted in reduced surface roughness of asphalt binder. Therefore, it can be concluded that nano $Fe_2O_3$ has a considerable effect on the stiffness property of the modified asphalt binder.

In many studies, the percentage of nanomaterials used in asphalt binder has varied between 0 and 1.5wt% of asphalt binder, because using more than this amount makes the design uneconomical. As a result, based on the experiences from past studies in the present research, first, three different percentages of nano-$Fe_2O_3$ (0.5%, 1%, and 1.5wt% of asphalt binder) were used for asphalt binder modification, and asphalt binder rheology tests such as DSR, BBR, MSCR, and LAS tests were performed on them to determine the most optimal percentage for nano-$Fe_2O_3$ with these three percentages by examining the results of these tests. Then, asphalt mixture was constructed with this optimal percentage to determine the optimal asphalt binder percentage. After that, SCB and ITS specimens were employed to identify the effect of this nano amount on the fracture toughness and moisture susceptibility of the asphalt mixture.

**Table 2** Asphalt binder properties used in this study

| Parameter | Percent purity % | Weight loss % | Flash point ℃ | Ductility cm | Softening point ℃ | Penetration grade 0.1 mm | Density at 25 ℃ |
|---|---|---|---|---|---|---|---|
| Standard limit | > 99 | < 0.2 | > 230 | > 100 | 49–56 | 60–70 | 1.01–1.06 |
| Value | 99.6 | 0.2 | 308 | 102 | 50 | 68 | 1.013 |

**Table 3** Nano $Fe_2O_3$ properties used in this study

| Chemical formula | Bulk specific gravity (g/cm$^3$) | Particle size (Nm) | Color | Morphology | Specific surface area (m$^2$/g) | Purity (%) | Water absorption (%) |
|---|---|---|---|---|---|---|---|
| $Fe_2O_3$ | 0.25 | 40 | Red Brown | Spherical | 60 | 99.0 | 0.2 < |

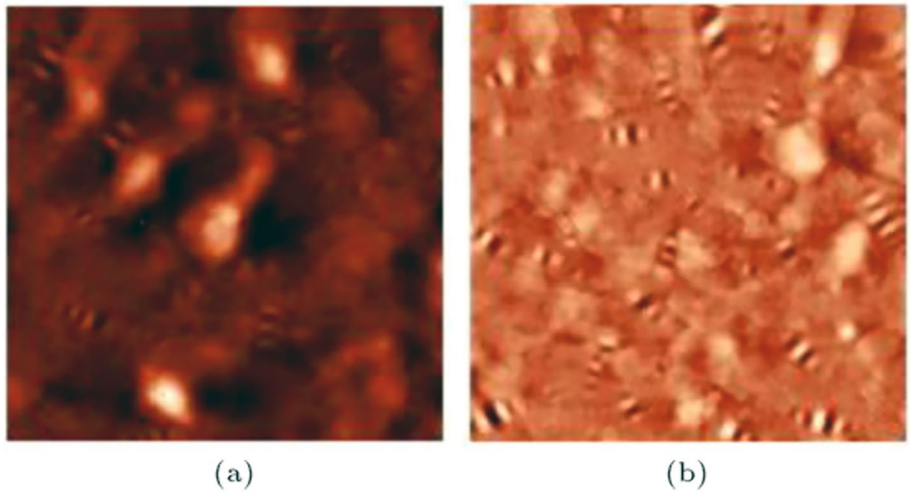


**Fig. 3** Two-dimensional surface topography of **a** origin asphalt binder **b** nano modified asphalt binder

**Table 4** Details of asphalt mixture tests

| Tests | Variables | Value |
|---|---|---|
| SCB test | Crack geometry | Vertical and angular |
| | Temperature (℃) | −5, −15, −25 |
| | Me parameter | 0, 0.5, 1.0 |
| | Nano $Fe_2O_3$ (%) | 0% and the optimal percentage of asphalt binder test results |
| Modified Lottman test | Temperature (℃) | 25 |
| | Nano $Fe_2O_3$ (%) | 0% and the optimal percentage of asphalt binder test results |

The details of the research and its flowchart are showed in Table 4 and Fig. 4.

## 3.3 Test Methodology

### 3.3.1 Asphalt Binder Tests

In order to investigate the effect of Nano-$Fe_2O_3$ on the rheological performance of asphalt binder and its resistance against rutting, fatigue, and cracking that occurs in high, medium, and low temperature ranges, respectively, in the present research, Dynamic shear rheometer tests were conducted based on ASTM standard D7175, Bending beam rheometer based on ASTM D6648 standard, Multiple Stress Creep and Recovery based on AASHTO T350 standard, and Linear Amplitude Sweep Test based on AASHTO TP 101–14 standard. According to the results of these tests, the optimal nano percentage was identified among the investigated percentages.

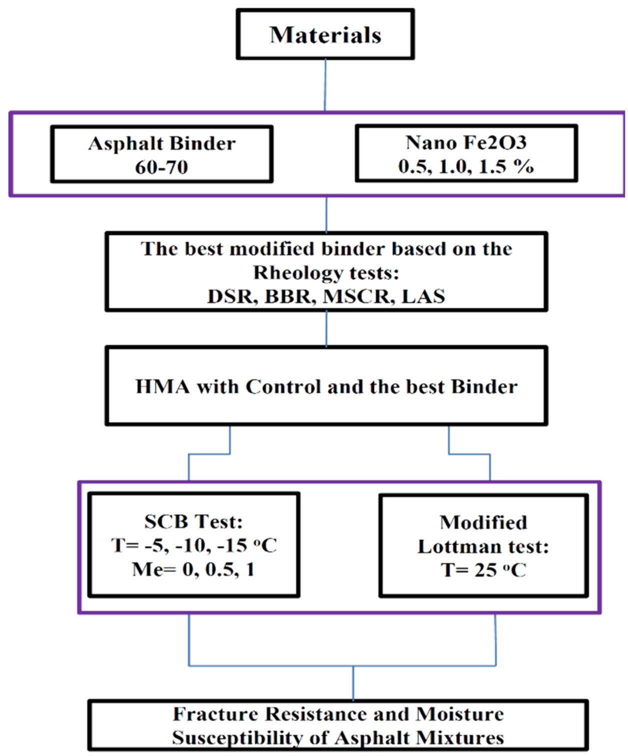


**Fig. 4** Flowchart of the research

### 3.3.2 Marshal Test

Marshal Test procedure was conducted according to ASTM D1552 Standard to calculate the optimum asphalt binder content of asphalt mixtures. The sample size was 101.6 mm in diameter by 76.2 mm in height. Three cylindrical sample were prepared and compacted according to the standard test method. It should be noted that in this research, in order to investigate more precisely the impact of nano-$Fe_2O_3$ on the mixture resistance against cracking and moisture sensitivity, only the optimal asphalt binder percentage of the control specimens was calculated and the nano-modified specimens were also constructed with this asphalt binder percentage. According to the results of Marshall's tests, the optimal percentage of asphalt binder has been calculated as 5%.

#### 3.3.3 SCB Test

In the present research, the SCB section was selected as a suitable laboratory specimen to perform fracture tests in asphalt mixtures. To construct SCB specimens, cylindrical specimens with a diameter of 15 cm and a height of about 12 cm are prepared first. All of the specimens were made and compacted according to Marshal method based on ASTM D1559. Then, to construct SCB-shaped specimens from cylindrical specimens, it is necessary to first cut the cylindrical specimens into discs with a thickness of about 1/3 cm. To construct SCB specimens, it is necessary to cut the cut specimens in half. After preparing SCB specimens, it is time to create cracks in these specimens. This stage of the work was performed using a waterjet cutting machine. Two different types of vertical and angular cracks were created in the specimens.

In conducting fracture toughness tests, the crack length is determined in such a way that the location of the crack tip is far enough from the location of the supports, the place of force application, and the specimen boundaries so that the stress area created around the crack tip is not affected by the concentration of the stresses created in the supports. Therefore, the crack length was considered 22.5 mm in all the manufactured sections, which, according to the radius of the SCB specimens, the a/R ratio in the specimens is approximately equal to 0.3. Figure 5 depicts a view of the SCB specimens constructed in this research.

**Fig. 5** SCB Sample made in this study

#### 3.3.4 Modified Lottman Test

One of the most popular methods for evaluating the moisture susceptibility of asphalt mixtures is the modified Lottman test. This test is performed to determine the moisture susceptibility of dense mixtures after exposing the specimens to saturated conditions. Cylindrical specimens with a diameter of 150 mm and a thickness of 57.5 mm were compacted. Then according to the AASHTO T283, cured and uncured were subjected to the indirect tensile test between two rigid metal strips in the diagonal direction of the specimens with a loading rate of 50 mm/min at a temperature of 25 ℃ until the specimens were fractured. The force value required to fracture the specimens is recorded. The indirect tensile strength (ITS) value is calculated from the following equation:

$$ITS = \frac{2 \times P}{\pi \times t \times d} \quad (1)$$

where $P$ the maximum value of the applied force to fracture the specimens (kN), t is the thickness of the specimens (mm) and d is the diameter of the specimens (mm). The Tensile strength ratio (TSR) value is typically expressed as a percentage and as the average ITS of saturated to unsaturated specimens in Eq. 2:

$$TSR = \frac{ITS_{Conditioned}}{ITS_{unConditioned}} \times 100 \quad (2)$$

Lottman stated that asphalt mixtures with a content higher than 70% are generally less susceptible to moisture damage that occurs in high rainfall areas. Meanwhile, the Iran Highway Asphalt Paving Code (No. 234) suggests the appropriate ratio of 75% in accordance with AASHTO. Therefore, asphalt specimens whose TSR ratio is more than 75% will not be susceptible to moisture damage.

## 4 Results and Discussions

### 4.1 Asphalt binder Rheological Tests Results

#### 4.1.1 DSR Test Results

The DSR test was performed to evaluate the fatigue and rutting resistance of asphalt binder under normal and aged conditions at different temperatures. One of the main outputs of this test is G*/Sinδ (rutting resistance) and G*.Sinδ (fatigue resistance). To investigate the effect of nano-$Fe_2O_3$ on the rutting behavior of asphalt binder, the DSR test was conducted at different temperatures of 58, 64, and 70 ℃ on

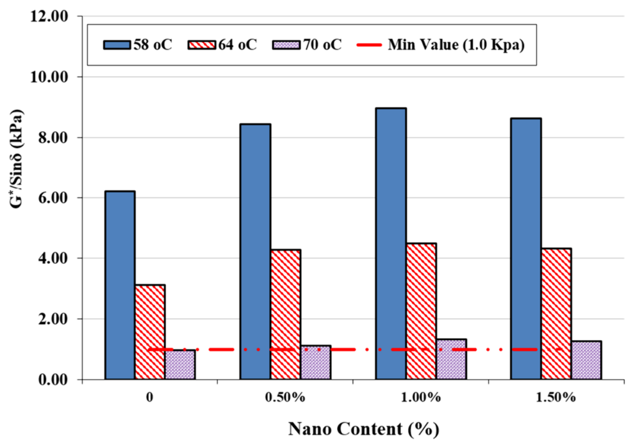


**Fig. 6** G*/Sinδ value of unaged asphalt binder containing different percentages of nano-$Fe_2O_3$

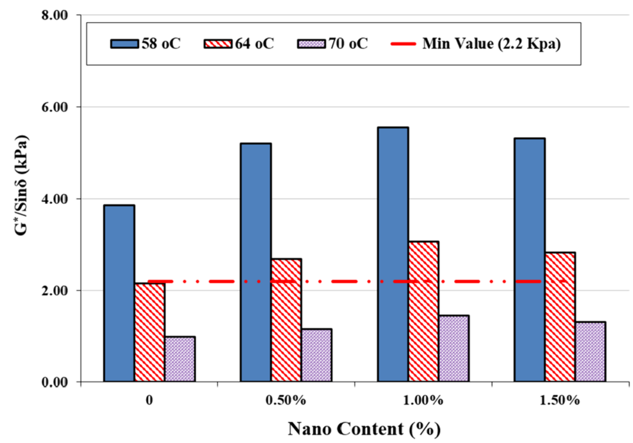


**Fig. 7** G*/Sinδ value of short-term aged asphalt binder containing different percentages of nano-$Fe_2O_3$

two different types of asphalt binder (control and modified). The value of G*/Sinδ for different specimens in two conditions of normal asphalt binder (unaged) and short-term aged asphalt binder are shown in Figs. 6 and 7. The minimum 1 kPa and 2.2 kPa are determined for unaged asphalt binder and short-term aged asphalt binder.

As it is clear from the test results in Fig. 6, in control asphalt binder (unaged) at all temperatures, the value of G*/Sinδ, which is the main parameter for measuring the rutting resistance of asphalt binder, is more than the value of 1 kPa determined by SHRP, and it is only at 70 °C that the value of G*/Sinδ is less than 1. This issue is justified by the fact that the asphalt binder used in the laboratory process of this research is asphalt binder 60–70, which in terms of performance is equivalent to asphalt binder 64–22, and at temperatures higher than 64 ℃, the performance will drop. This issue is evident in the results of this study. However, after adding nano-$Fe_2O_3$, the value of G*/Sinδ at 70 ℃ is more than the minimum regulation value (1 kPa), and the rutting resistance has increased with the increase in nano percentage at all temperatures.

Figure 7 depicts the G*/Sinδ values for the short-term aged bituminous specimens. As it can be deduced from this Figure, in the asphalt binder modified with different percentages of nano-$Fe_2O_3$ and at all temperatures except 70 ℃, the G*/Sinδ value is more than the minimum SHRP value (2.2 kPa). The noteworthy point is that this value has also increased after adding different percentages of nano-$Fe_2O_3$, which can be a good sign for improving the behavior of asphalt binder against permanent deformations and rutting. As the results show, at 58 ℃, the G*/Sinδ value in asphalt binder modified with 1% nano-$Fe_2O_3$ is about 45% more than its value without nano, which indicates the significant effect of nano-$Fe_2O_3$ on the improvement of the rheological behavior of asphalt binder. However, with the increase in temperature from 64 to 70 ℃, although the trend is still increasing with the addition of nanoparticles, it has not been able to increase the G*/Sinδ value more than the minimum regulation value (2.2 kPa). The main reason for this can be the high asphalt binder susceptibility to temperature.

The results indicate that aging affects the chemical composition of asphalt binder. Asphalt binder is a colloidal mixture constituted of large molecules called asphaltene that forms the dispersed phase and saturates, aromatics, and resins that form the continuous (liquid) phase. Based on the studies conducted [22, 23], carbonyl compounds and sulfoxides increase due to the aging process, while aromatics decrease. The amount of resin and asphaltene has also increased, and the amount of saturates has also changed slightly. In the short-term aging stage, the reduction in volatile substances in bituminous mixtures causes the asphalt binder to become stiffer, which can improve the asphalt binder's performance in the short term, and it has more brittle properties in the long term due to the stiffening of the asphalt binder.

The high effect of nano-$Fe_2O_3$ additive is determined at 64 °C, where the value of G*/Sinδ has decreased significantly with increasing temperature and has become less than 2.2. Meanwhile, at the same temperature, after adding different percentages of this nano value, the G*/Sinδ value has increased, and the rheological performance of asphalt binder has improved against rutting failure leading to significant damage to the pavement at this temperature.

As it is clear from the results, in the conditions of unaged and short-term aged, the value of G*/Sinδ is improved by adding different percentages of nano-$Fe_2O_3$, and the highest value is obtained by adding 1% of nano-$Fe_2O_3$. The results indicate that the variation of G*/Sinδ has decreased with the increase of nano percentage from 1 to 1.5%.

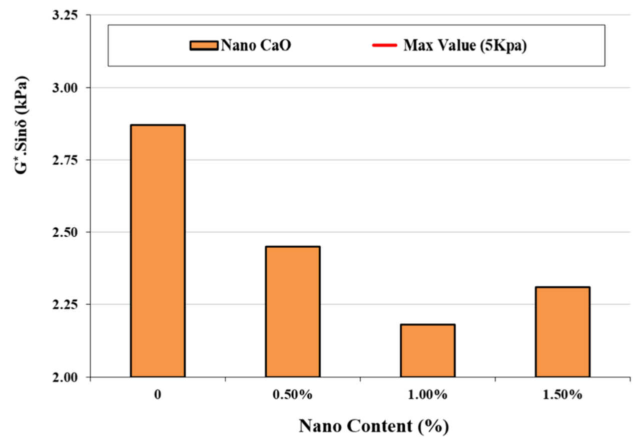


**Fig. 8** G*/Sinδ value of long-term aged asphalt binder modified with different percentages of nano-$Fe_2O_3$

Since fatigue cracks typically occur at low to intermediate pavement temperatures and after the pavement was subjected to loading for a while, a dynamic shear rheometer test was performed to determine the fatigue at 20 °C on aged asphalt binder in two Rolling thin film oven (RTFO) and Pressure Aging Vessel (PAV) devices. The control factor of fatigue cracks is G*.Sinδ. SHRP Institute considers the maximum value of 5 kPa for G*/Sinδ. Figure 8 depicts the values of G*.Sinδ for asphalt binders modified with different percentages of nano-$Fe_2O_3$. The results of this experiment indicate that the addition of different percentages of nano-$Fe_2O_3$ can improve the fatigue performance of asphalt binder, and the G*.Sinδ value in asphalt binder containing 1% nano-$Fe_2O_3$ is about 24% lower than asphalt binder without nano. However, after increasing the amount of nano-$Fe_2O_3$ from 1 to 1.5%, this amount starts to decrease. As a result, 1% of nano-$Fe_2O_3$ can be the best choice from the perspective of the present research results.

In this research, an oxidizer ($Fe_2O_3$) was used with the aim of increasing the hardness of asphalt binder and preventing the kind of damage that occurs due to the softness of asphalt at high temperatures (such as rutting), and the results showed its positive effect. However, the hardening of asphalt as a result of the addition of oxidizers increases the risk of increasing thermal cracks and fatigue. In order to prevent the occurrence of hardening phenomenon and reduce the fatigue life, an nano oxidizer (Nano $Fe_2O_3$) was used, so that the high specific surface of the nano particles makes the bonds of the bitumen particles stronger and prevents them from separating in the high repetition of loads (occurrence of the fatigue phenomenon) and start cracking.

### 4.1.2 BBR Test Results

When the pavement temperature decreases, the asphalt mixture shrinks. Since the friction in the sub-layers of the pavement prevents their movement, tensile stresses arise in the pavement. When this stress exceeds the tensile strength of the asphalt mixture, low-temperature cracks occur. A Bending Beam Rheometer test is employed to determine the creep stiffness of asphalt binder. If the creep stiffness is very high, asphalt binder will behave brittle, and low-temperature cracks are likely to occur. Since low-temperature cracks typically occur after a period of the pavement operation time, the tests are performed on the aged asphalt binder in the RTFO and PAV devices. The variation rate of asphalt binder stiffness over time at low temperatures is known as the m-value. A higher m-value is appropriate; because as the temperature decreases and the pavement begins to shrink, the asphalt binder will react like a low-stiffness material. This decrease in stiffness (stress-to-strain ratio) leads to less tensile stresses in the asphalt binder; therefore, there will be less possibility of low-temperature cracks. The minimum m-value after 60 s of loading is equal to 0.3 according to the requirements of Superior Performing Asphalt Pavements.

Figure 9 depicts the results of the BBR test on asphalt binders that were first modified with different percentages of nano-$Fe_2O_3$ and then aged by the RTFO and PAV devices. As it is clear from the results, the value of creep stiffness (m) obtained is higher than the minimum value determined by the SHRP (0.3).

It can be inferred from the results of this test that up to 1% nano, the more nano-$Fe_2O_3$ percentage increases, the more the creep stiffness increases, which can significantly contribute to the asphalt mixture, especially in cold weather. As it is evident, the possibility of low-temperature cracks in the asphalt mixture increases in these weather conditions. Meanwhile, the higher the rate of creep stiffness variations,

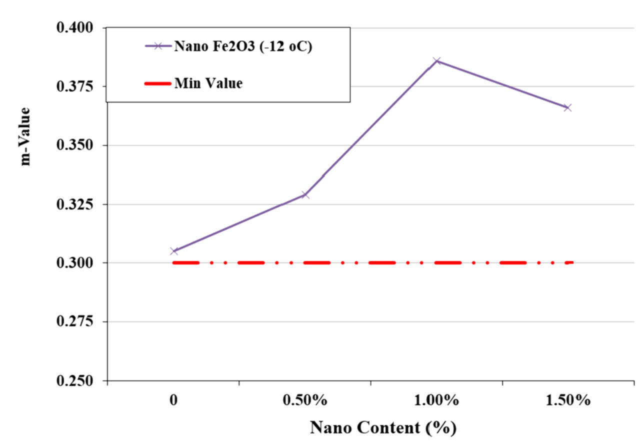


**Fig. 9** m-value of asphalt binder modified with different percentages of nano-$Fe_2O_3$ at -12 °C

the higher resistance of asphalt binder and asphalt mixture against the occurrence of these cracks. However, with the increase of nano from 1 to 1.5%, this parameter has started to decrease. Although its value is still more than the minimum of 0.3, it is expected that with the further increase of nano, its value will be less than the minimum value of the standards. As a result, based on the results of this test at – 12 ℃, adding 1% nano-$Fe_2O_3$ has brought the best result, increasing the low-temperature crack resistance of asphalt binder by about 27%.

## 4.2 MSCR Test Results

According to the AASHTO T350 standard, the MSCR test is performed on RTFO specimens at PG temperature (64 ℃ in this research) and includes 1 s of loading and 9 s of unloading at the stress levels of 0.1 and 3.2 kPa in 10 cycles for each stress level. Non-recoverable creep compliance ($J_{nr}$) and percent recovery (R) are obtained in each cycle to determine the high temperature performance of the asphalt mixture.

Figures 10 and 11 illustrate the MSCR test results at 64 °C. As shown in Fig. 10, pure asphalt binder has the highest $J_{nr}$ value in both stress levels, which indicates that the base asphalt binder is softer. As the percentages of additives increase, the asphalt binder gradually becomes stiffer, and in both stress levels, the non-recoverable creep compliance ($J_{nr}$) value decreases, and the percent recovery (R) increases. The stiffening of asphalt binder in this test is in agreement with the results of the BBR test, which indicates that adding nano-$Fe_2O_3$ has been able to increase the stiffness of asphalt binder. This increased stiffness leads to improving the rutting performance of asphalt binder.

One of the evaluation criteria in this test is the percentage difference of $J_{nr}$ at two stress levels of 0.1 and 3.2 kPa, which according to the AASHTO T350 standard, the percentage difference of $J_{nr}$ should not be more than 75%. This

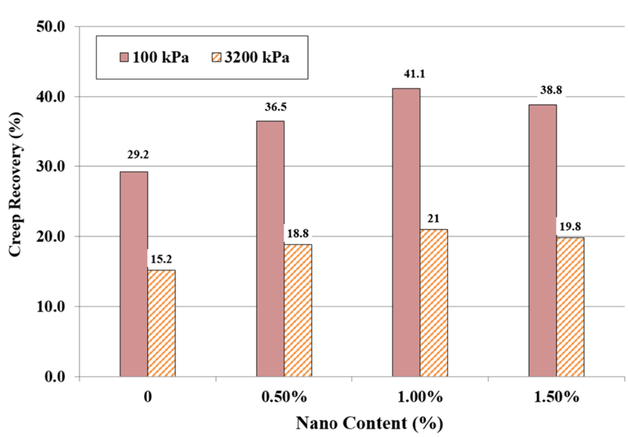


**Fig. 10** The strain recovery percentage in different stresses versus nano-$Fe_2O_3$ percentages

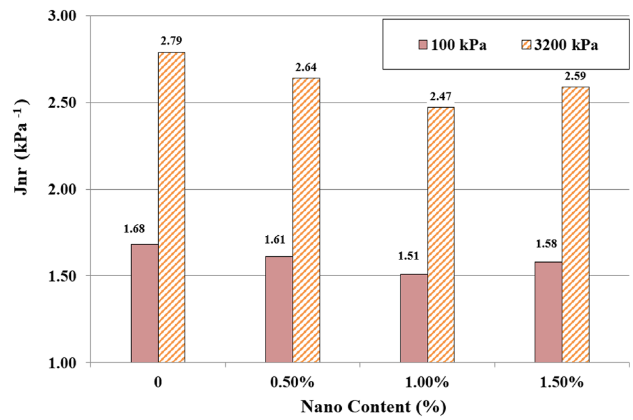


**Fig. 11** Creep compliance of bituminous specimens versus nano-$Fe_2O_3$ percentages

parameter expresses the susceptibility of the material to the stress level. In other words, by changing the stress level from 0.1 to 3.2 kPa, $J_{nr}$ should not vary more than 75%. As the results indicate, this value was less than 75% in the specimens examined in this test and all conditions. However, the significant point is the effect of adding additives that, as its value increases, the value of $J_{nr\text{-}diff}$ decreases, indicating that adding additives will reduce the stress sensitivity of asphalt binder. However, this decrease continued until the nano percentage of 1%, and with the increase of the nano percentage, it will be an increasing trend again, which can be a negative point.

### 4.2.1 LAS Test Results

Figure 12 depicts the results of the LAS test at strain levels of 2.5 and 5% at 25 °C. As the results show, the fatigue life of bituminous specimens increases gradually with the addition of different percentages of nano-$Fe_2O_3$ and continues up to 1% nano. These results, extracted from the LAS test, confirm how G*/Sinδ varies in the DSR test. The results of the LAS test indicate that the fatigue life of asphalt binder and consequently the asphalt specimens modified with 1% nano-$Fe_2O_3$ is about 33% higher than the asphalt binder specimens without nano. However, after the nano percentage addition from 1 to 1.5%, the fatigue life of the specimens will start to decrease. As a result, based on the results of this test, 1% nano-$Fe_2O_3$ can be selected as the best percentage.

## 4.3 Asphalt Mixture Test Results

The results of asphalt binder tests in the previous section proved that the specimens containing 1% nano-$Fe_2O_3$ had the best performance among the three options. Thus, in the asphalt mixture tests, only the control specimen (without

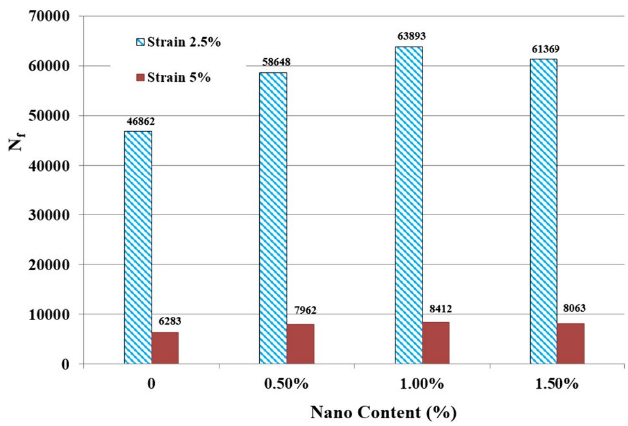


**Fig. 12** The number of tolerable loading cycles of asphalt binder versus nano-$Fe_2O_3$ percentages

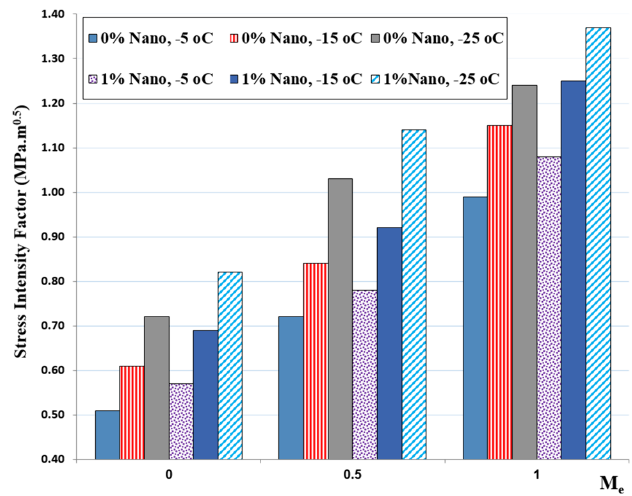


**Fig. 14** Fracture toughness values of asphalt specimens with angular cracks

additives) and the asphalt specimen constructed with asphalt binder containing 1% nano-$Fe_2O_3$ will be compared.

#### 4.3.1 SCB Test Results

Figures 13 and 14 illustrate the results of the fracture toughness test on control and modified asphalt specimens with 1% nano-$Fe_2O_3$ at different temperatures and in two conditions of specimens with vertical and angular cracks, in shear ($M_e = 0$), tensile loading modes ($M_e = 1$), and combined mode ($M_e = 0.5$). As the results indicate, the stress intensity factor ($K_{eff}$) has increased by decreasing the temperature from – 5 ℃ to – 15 ℃, – 25 ℃, and in both conditions of specimens with vertical and angular cracks. This indicates that the asphalt specimens will perform better at lower temperatures, and at lower temperatures, the cracking resistance increases as the asphalt binder stiffens. Given the viscoelastic behavior of asphalt binder, this section is more elastic at lower temperatures, and it will gradually show plastic behavior at higher temperatures. However, in comparison between – 25 ℃ and – 5 ℃ temperatures, it will be – 25 ℃ that asphalt binder and asphalt mixture can have more elastic behavior, and as a result, their resistance to fracture initiation will be higher. On the other hand, the worrying point in this regard is the level under the load–displacement diagram at lower temperatures, the increase of which is actually equivalent to the increase of the fracture energy, and it can be stated that the specimens treat harder at low temperatures; however, they will endure and experience harder fracture. The above items have the same process but different values in each loading mode. As an example, the results indicate that in the condition of vertical crack and pure tension loading mode ($M_e = 1$), the value of the fracture resistance of the control specimens at – 25 ℃ will be approximately 8 and 19% higher than at – 15 ℃ and – 5 ℃.

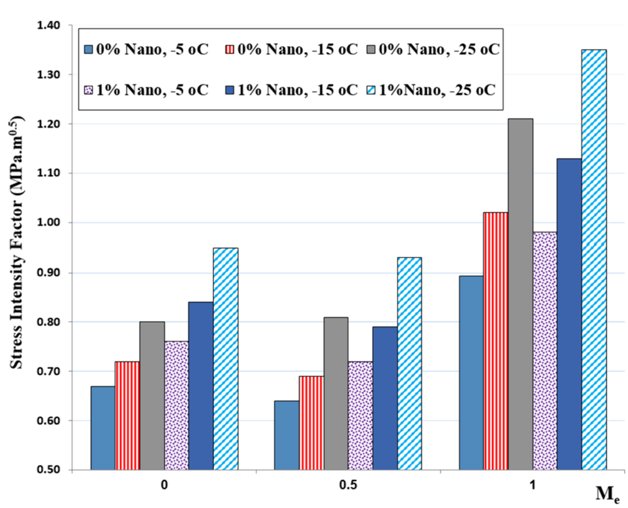


**Fig. 13** Fracture toughness values of asphalt specimens with vertical cracks

To investigate the effect of variations in crack geometry on the stress intensity factor under different temperatures, as the results indicate, at a given temperature, the fracture toughness of specimens with vertical cracks in the combined loading mode ($M_e = 0.5$) has reached the lowest value. Meanwhile, in specimens with angular cracks, this lowest value of resistance has occurred in the shear loading mode II ($M_e = 0$). In general, it can be stated that the lowest crack resistance occurs in the condition of angular cracks and the shear loading mode ($M_e = 0$). The comparison of the results in two conditions of vertical and angular crack geometry indicates that the closer the loading mode is to shear ($M^e = 0$), the higher the resistance value of the specimens with vertical cracks is than the specimens with angular cracks. However, with the increase of the $M_e$ value, the strength of specimens with angular cracks increased. And finally, in the tensile loading mode ($M_e = 1$), specimens with angular cracks have more resistance than specimens with vertical cracks.

Figure 15 examines the variations of cracking resistance in the modified specimens compared to the control ones at different temperatures. This Figure illustrates well the effect of adding 1% nano-$Fe_2O_3$ on the cracking resistance of asphalt specimens. The results indicate that this effect starts from about 8% and continues to about 19%. The least impact has occurred on the shape factor of 0.5, angular crack at – 5 ℃, and the most impact has occurred on the shape factor of 0, vertical crack at – 25 ℃. Due to being an oxide, nano-$Fe_2O_3$ makes asphalt binder stiffer, and this asphalt binder shows more cracking resistance at low temperatures.

### 4.4 Asphalt Mixture Moisture Susceptibility

According to the results of the Marshall test and after achieving the optimal asphalt binder percentage, specimens were prepared to evaluate the moisture susceptibility of asphalt mixtures using the indirect tensile test. The TSR ratio, which is an effective index for evaluating the moisture susceptibility of asphalt mixtures, is obtained by dividing the tensile strength of saturated specimens by dry specimens. According to the Iran Highway Asphalt Paving Code, this ratio should not be less than 75%. Figures 16 and 17 show the full results of the indirect tensile test.

The results of the indirect tensile test indicate that adding the optimal percentage of nano-$Fe_2O_3$ (1%) to asphalt binder and then using it in asphalt mixture will improve their moisture susceptibility. One of the reasons for this improvement is that nano-$Fe_2O_3$ makes the asphalt binder stiffer and improves its adhesion. The use of this modified asphalt binder with better adhesive properties in asphalt mixtures can improve the resistance of the asphalt mixture against water penetration and prevent its adverse effects. Hence, using this asphalt mixture in areas with considerable amount of rain and snowfall can substantially prevent the occurrence of moisture damage in asphalt mixtures. As the results of this research indicate, by replacing 1% of asphalt binder with nano-$Fe_2O_3$, the moisture susceptibility of asphalt mixtures has improved by 9% compared to control specimens.

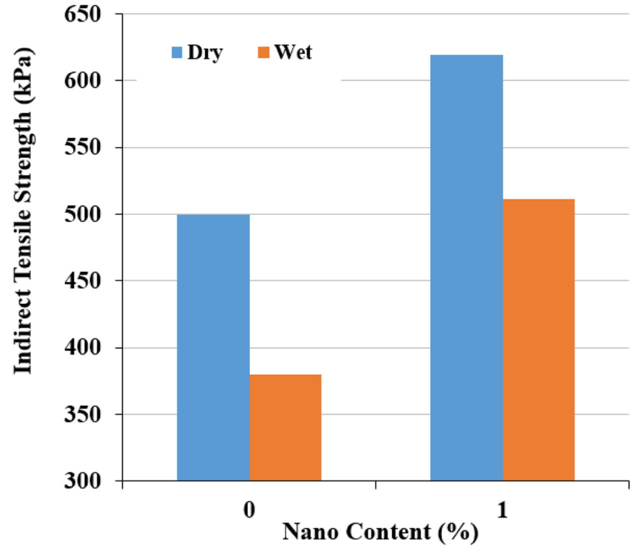


**Fig. 16** Indirect tensile strength of asphalt mixtures

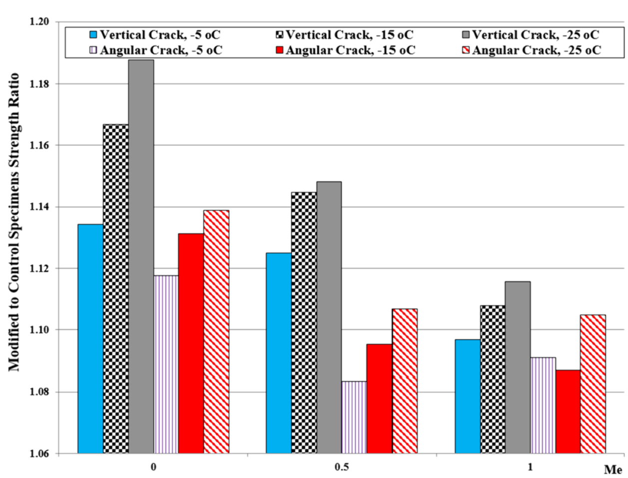


**Fig. 15** Resistance variations of modified specimens to control specimens at different temperatures and crack types

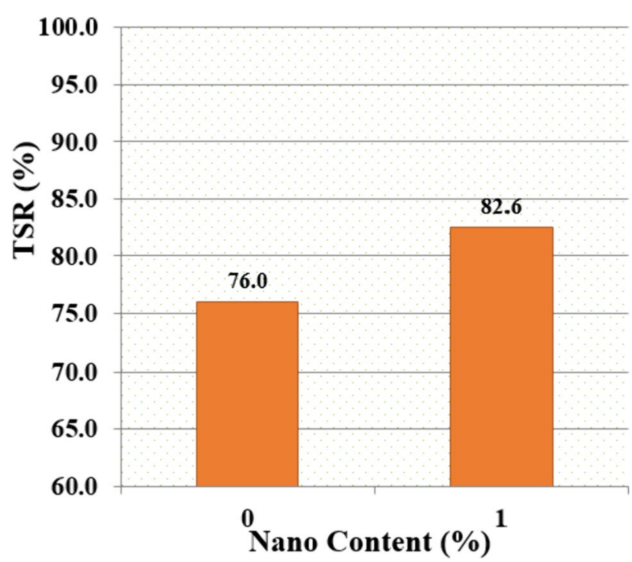


**Fig. 17** TSR of control and modified asphalt mixtures

## 5 Summary and Conclusion

In the present research, a laboratory program was developed to investigate the effect of nano-$Fe_2O_3$ on the rheological characteristics of asphalt binder, and different tests were conducted on normal asphalt binder and modified asphalt binder with different percentages (0.5, 1.0, and 1.5%) of nano-$Fe_2O_3$. The main results of the asphalt binder tests in this research are as follows:

- As it is clear from the results, in the unaged and short-term aged conditions, the value of G*/Sinδ is improved by adding different percentages of nano-$Fe_2O_3$, and the highest value is obtained by adding 1% of nano-$Fe_2O_3$.
- The values of G*/Sinδ in this research indicate that adding nano-$Fe_2O_3$ can increase asphalt binder resistance to fatigue failure that leads to alligator cracking in the asphalt mixture.
- The results of the BBR test indicate that at – 12 ℃, adding 1% nano-$Fe_2O_3$ increases the low-temperature crack resistance of asphalt binder by about 27%.
- From the results of the MSCR test, it is concluded that asphalt binder without additives has the highest value of creep compliance. This matter confirms the softness of the control asphalt binder. Meanwhile, the asphalt binder becomes stiffer with the addition of nano-$Fe_2O_3$, and the strain recovery percentage (R) increases.
- From the results of the LAS test, it can be deduced that adding 1% nano-$Fe_2O_3$ improves the number of tolerable loading cycles of asphalt binder and, accordingly, asphalt mixtures by about 33%.

Generally, based on the results of the asphalt binder tests in the present research, it was found that the best performance occurs in asphalt binders with 1% nano-$Fe_2O_3$. Therefore, in order to investigate the effect of this modified asphalt binder on the resistance of asphalt binder against thermal cracking as well as moisture in comparison with the control specimen, asphalt specimens were constructed with control asphalt binder (without additives) and asphalt binder modified with 1% nano. The results of SCB and ITS tests on asphalt specimens are as follows:

- This research indicated that adding 1% nano-$Fe_2O_3$ can improve the fracture toughness of asphalt mixtures from about 8 to 19%. The least impact has occurred on the shape factor of 0.5, angular crack at – 5 ℃, and the most impact has occurred on the shape factor of 0, vertical crack at – 25 ℃.
- The results of this research indicate that replacing 1% of asphalt binder with nano-$Fe_2O_3$ improves the moisture susceptibility of asphalt mixtures by 9% compared to control specimens.

## Declarations

**Conflict of Interest** The authors declare that they have no conflict of interest.

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