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# New achievement in moisture sensitivity of nano-silica modified asphalt mixture with a combined effect of bitumen type and traffic condition

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## Abstract

Moisture damage is a critical problem of asphalt pavements. Some factors affecting the moisture sensitivity of asphalt mixtures include bitumen properties, air void content, and the type of anti-stripping additive. This study investigated the use of nano-silica for improving the properties of different bitumens and to increase the moisture resistance of asphalt mixtures at different traffic loading conditions. Two types of bitumen (60-70 and 85-100 pen grade) were modified with various percentages of nano-silica (0, 0.2, 0.4, 0.7, and 0.9% by the weight of the bitumen), and physical and rheological tests were conducted to determine the optimum nano-content. Then, the optimal content was used to prepare asphalt mixtures at three different air void contents (4, 5, and 6%) to simulate the heavy, medium, and low traffic loading conditions. Indirect tensile strength (ITS) and compressive strength tests were performed on the specimens in dry and wet conditions, and the effect of air void content and the type of bitumen on the moisture sensitivity and compressive strength of unmodified and nano-silica-modified asphalt concrete was investigated. The results showed that nano-silica improved the moisture sensitivity of asphalt mixtures with 60-70 and 85-100 pen grade bitumens at all air void contents and the effect of nano-silica was higher on the moisture sensitivity of asphalt mixtures with 60-70 than 85-100 pen grade bitumen.

*Keywords*: Asphalt mixtures; Moisture sensitivity; Nano-silica; Air voids; Modified bitumen; Compressive strength

## 1. Introduction

To achieve the optimal performance of asphalt pavements, asphalt must have good durability and sustainability during service life, a feature that depends on asphalt properties such as adhesion and cohesion between aggregates and the bitumen as well as various environmental factors such as temperature, air, and water. Moisture in asphalt mixture may cause the loss of adhesion between aggregates and the asphalt binder (the composition of bitumen and filler) or the loss of cohesion of bitumen that leads to stripping and bleeding in asphalt pavements [1,2]. Although the mechanism of moisture sensitivity is not completely understood yet, factors such as the properties of bitumen, aggregates, asphalt mixtures, and antistripping admixture play an essential role in causing moisture sensitivity and increasing damages [3]. Several features of aggregates, including surface texture, porosity, type of mineralization, and surface coatings tremendously affect the sensitivity of asphalt mixes to moisture. Dry aggregates with coarse and rough textures and the alkali-free properties of clay minerals and surface coatings are more resistant to stripping and moisture damages [4-6]. One of the most influential parameters on the moisture sensitivity of the bitumen is the type of bitumen used in asphalt mixes. The rheological properties of bitumen, the amount of asphaltene in the bitumen, and bitumen chemical functional groups are highly influential on the sensitivity of asphalt mixtures to moisture. For instance, bitumen chemical groups readily dissolved in water have carboxylic acids and water-resistant groups such as ketones and phenolics [7,8]. An important mechanism in moisture sensitivity is mechanical and physical adhesion and the chemical bonding between the bitumen and aggregates, and water can cause early breakdowns in the chemical bonding between the bitumen and aggregates in asphalt [9,10].

There are several solutions for reducing the moisture sensitivity of asphalt mixtures. A significant technique for managing the moisture sensitivity of asphalt mixtures is using anti-stripping agents [11]. Two approaches are adopted for measuring the effect of anti-stripping agents on asphalt moisture sensitivity: experiments carried out for loose (e.g. static immersion test) or compacted mixtures (e.g. the modified Lottman) [12,13]. Anti-stripping agents are incorporated to coat the surface of the aggregates to overcome the prevailing electrical loads on the surface and reduce the surface energy of the aggregates. Another approach is to enhance the adhesion and cohesion properties of bitumen through a liquid anti-stripping additive. These additives are chemical surfactants that improve the surface coatings of aggregates by reducing surface tension, and hydrated lime and the

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amine family materials are the most important of them [14,15]. Using liquid anti-stripping additives has numerous drawbacks such as lack of considerable variations in the resistance of different mixtures against moisture damage [16,17] and a negative effect on rutting, aging, and fatigue cracking of asphalt mixtures [17-19]. In addition to liquid anti-stripping additives, polymer modified bitumen is used to improve different properties of asphalt like rutting, moisture damage, etc. Also, different other performance test can also be performed to measure the effectiveness against these properties like rutting, low temperature cracking, bleeding, and stripping etc [20-22]. However, it seems that the application of nanomaterials is one of the best methods for enhancing the bitumen and aggregate properties and increasing the moisture resistance of asphalt mixtures.

Nano-materials can induce changes in the electrical load of aggregate surfaces and improve bitumen coating by coating the surfaces of aggregates [15]. Due to the specific surface area above nanometer levels, nanomaterials are capable of improving and controlling some of the properties of bitumens and asphalt mixtures [23]. Previous studies have utilized various nanomaterials in order to modify bitumen properties and reduce the moisture sensitivity of asphalt mixes. For instance, Behbahani et al. [24] used nano-zycosoil to evaluate the moisture sensitivity of glass asphalt mixtures through the indirect tensile test on wet and dry specimens. It was revealed that the addition of this material increased the tensile strength ratio (TSR) and the moisture resistance of the specimens. In order to modify the moisture properties of bitumens, Hamedi and Moghadas Nejad [15] used nano-calcium-carbonate ($CaCO_3$) because of its calcareous origin and hydrophobic properties. Two types of granite and limestone aggregates were utilized to prepare asphalt specimens. The results of surface free energy method and modified Lottman test demonstrated that nano-$CaCO_3$ increases the wettability of the bitumen on the aggregate surface, increases the adhesion between the bitumen and aggregates, and enhances the moisture resistance of the specimens. Azarhoosh et al. [25] evaluated the effect of nano-$TiO_2$ on the bitumen-aggregate bond by using 85-100 penetration-grade (pen grade) bitumen and two types of aggregates (limestone and granite). According to the surface free energy of aggregates and the asphalt binder, they concluded that with an increase in nano-$TiO_2$, the polar component of the bitumen surface energy is decreased, the bitumen acidity is decreased, and thus the adhesion of bitumen and acidic aggregates is enhanced. Other nanomaterials that have been used to reduce moisture sensitivity include nano-zinc oxide (ZnO) [26], nano-zycosoil [27], and nano-clay [28].

One of the important nanomaterials useful for modifying the properties of bitumen and asphalt mixtures is nano-silica. High-performance properties and low manufacturing costs are the important benefits of nano-silica. Nano-silica has many valuable features such as a suitable dispersing ability, strong absorption, a high specific area, high stability, and a high purity percentage [29]. In concrete structures, silica is used to raise the adhesion and fill the high-workability concrete [29]. Consequently, it seems that the use of nano-silica improves the performance of bitumens and asphalt mixtures as well as the adhesion between the bitumen and aggregates. Researchers have utilized nano-silica to modify bitumen properties and observed that the bitumen modified with nano-silica has a lower penetration grade and higher softening point than virgin bitumen [30,31] with an increased complex modulus (G*) and reduced phase angle ($\delta$) of bitumen [32,33]. In addition, the dynamic shear rheometer (DSR) experiments demonstrated that the fatigue resistance of bitumens is increased through modification with nano-silica [29,34]. However, the bending beam rheometer (BBR) test showed a higher creep resistance and lower m-value [32]. Researchers have also evaluated the effect of nano-silica on the performance of asphalt mixtures. For example, Yao et al. [29] utilized nano-silica to modify bitumen properties. By adding this material to the polymer acrylonitrile butadiene styrene (ABS) containing bitumen, they concluded that bitumen viscosity decreased at high temperatures and, in general, nano-silica improved the rutting and fatigue performance of the asphalt binder. Nur Izzi et al. [35] used polymer bitumen modified with nano-silica to study the effect of moisture sensitivity and different aging conditions on the properties of the asphalt mixture and observed that nano-silica decreases the moisture sensitivity and improves the mechanical properties of the asphalt mixture. Also, it was observed that the tendency of the bitumen to aging decreases with an increase in the nano-silica content. Based on these studies, it can be stated that the simultaneous influence of bitumen type, air void content of the mixture, and nano-silica on the moisture sensitivity of asphalt mixtures has not been investigated yet.

The aim of this study was to improve the properties of various bitumens and the moisture sensitivity of asphalt mixtures at different traffic loading conditions with nano-silica. The effect of nano-silica on the moisture sensitivity and compressive strength of asphalt mixtures prepared under different traffic loading conditions was investigated. To this end, the physical and rheological properties of two types of bitumen (60-70 and 85-100 pen grade) modified with low amounts of nano-silica (<1%) were investigated, and the optimal percentage of nano-silica was determined. Then, the optimal content was used to prepare asphalt mixtures at three air void contents (4, 5, and 6%) to simulate the heavy, medium, and low traffic loading conditions. Indirect tensile strength (ITS) and compressive strength tests were performed on the specimens in dry and wet conditions, and the influence of air void content was examined on the moisture sensitivity and compressive strength of the mixtures prepared with nano-silica-modified bitumen.

## 2. Experimental plan and methodology

### 2.1. Materials

### 2.1.1. Bitumen and aggregate

Two types of bitumen were used to manufacture the samples with 60-70 and 85-100 pen grades prepared from Isfahan and Tabriz refineries (Iran), and their properties are provided in Table 1. Limestone aggregates were utilized in this study. The physical properties and chemical compositions of the aggregates are listed in Table 2 and 3, respectively. The aggregates were graded using the continuous type IV scale of the AASHTO standard [36] which is presented in Table 4.

### 2.1.2. Nano silica

Silica is an abundant compound found worldwide that is used in the production of silica gel, colloidal silica, fumed silica, etc. [37]. The chemical formula of silica is $SiO_2$, it is structurally similar to diamond, and is found in nature in both crystalline and amorphous shapes. This material is white, with relatively high melting and boiling points [38]. Nano-silica has functional properties such as a high specific surface area, strong adsorption, good dispersal ability, high chemical purity, and high stability [29]. In the industry, nano-silica is employed as a rheological material to reinforce elastomers. In the pavement, it can also be used to increase the adhesion and mechanical strength of asphalt mixtures [39,40]. In the present study, nano-silica was incorporated to modify the bitumen properties; the chemical composition and physical properties of nano-silica are given in Tables 5 and 6, respectively.

### *2.2. Sample preparation and laboratory setup*

A high shear mixer and a temperature control chamber were used to produce bitumen modified with nano-silica to distribute nanoparticles homogeneously within the bitumen. Initially, the bitumen was heated to become fluid. Then, the nanoparticles were added to the bitumen at different percentages (0.2, 0.4, 0.7, and 0.9%

Table 1
Properties of the base bitumen.

| Test | Standard | Binder type | |
|---|---|---|---|
| | | 60-70 | 85-100 |
| Penetration (100 g, 5 s, 25°C), 0.1 mm | ASTM D5 | 61 | 86 |
| Ductility (25°C, 5 cm/min), cm | ASTM D113 | 112 | 115 |
| Rotational Viscosity at 135°C (Pa.s) | ASTM D4402 | 0.512 | 0.321 |
| Softening point (°C) | ASTM D36 | 51 | 49 |
| Flash point (°C) | ASTM D92 | 262 | 235 |

Table 2
Physical properties of the aggregate.

| Test | Standard | Lime stone | Specification limit |
|---|---|---|---|
| Apparent Specific gravity (coarse agg.) | ASTM C 127 | 2.63 | - |
| Apparent Specific gravity (fine agg.) | ASTM C 128 | 2.69 | - |
| Water absorption (%) (coarse agg.) | ASTM C 127 | 1.52 | - |
| Water absorption (%) (fine agg.) | ASTM C 128 | 1.68 | - |
| Los Angeles abrasion (%) | ASTM C 131 | 21.3 | Max 45 |
| Crushed particles of No. 4 (%) | ASTM D5821 | 95 | >90 |

Table 3
Chemical composition of aggregate.

| Properties | Lime stone |
|---|---|
| pH | 8.7 |
| Silicon dioxide, $SiO_2$ (%) | 4.5 |
| Calcium oxide, CaO (%) | 52.3 |
| $R_2O_3(Al_2O_3+Fe_2O_3)$ (%) | 19 |
| Aluminum oxide, $Al_2O_3$ (%) | 1.3 |
| Ferric oxide, $Fe_2O_3$ (%) | 0.6 |
| Magnesium oxide, MgO (%) | 1.6 |

Table 4
Gradation of aggregates used in the study.

| Sieve size (mm) | 19 | 12.5 | 4.75 | 2.36 | 0.3 | 0.075 |
|---|---|---|---|---|---|---|
| Lower–upper limits | 100 | 90-100 | 44-74 | 28-58 | 5-21 | 2-10 |
| Percent passing (%) | 100 | 95 | 59 | 43 | 13 | 6 |

Table 5
Ingredients of Nano silicon.

| Elements | $SiO_2$ | Ti | Ca | Na | Fe |
|---|---|---|---|---|---|
| content | ≥99% | <120ppm | <20ppm | <50ppm | <200ppm |

Table 6
Physical properties of the Nano silicon.

| Properties | Nano $SiO_2$ |
|---|---|
| Diameter (nm) | 20-30 |
| Surface volume ratio ($m^2/g$) | 130-600 |
| Density ($g/cm^3$) | 2.1 |
| Melting point(°C) | 1600 |

by the weight of the bitumen) and mixed at a shear influence of 4000 rpm and 150 °C. In order to achieve homogeneous samples, the bitumen and nano-silica were mixed for ~30 minutes.

The optimal amount of nano-silica was obtained from the rheological test results on the modified bitumen. In order to determine the optimum bitumen for asphalt mixtures, the Marshall mix design method (ASTM D1559) was employed, and the asphalt specimens were prepared with the nano-silica optimum amount and optimum amount of bitumen. To simulate different traffic loading conditions (heavy, medium, and light) on the specimens, the percentage of the target air void was set as 4, 5, and 6%, respectively. Accordingly, to reach the air void content of 4, 5, and 6 %, the required number of Marshall hammer blows was obtained as 60, 50, and 40, respectively, through experiments and trial and error, and the asphalt specimens were prepared with these air void contents. Finally, the modified Lottman and compressive strength tests were carried out on the specimens at 25°C. The experimental program adopted in this study is presented in Fig. 1.

### 2.3. *Laboratory tests*

#### 2.3.1. *Empirical rheological tests on bitumen*

In order to determine the optimal amount of nano-silica, rheological tests were conducted on unmodified and modified bitumen with different percentages of nano-silica (0.2, 0.4, 0.7, and 0.9% by the weight of the bitumen). This range was selected in order to examine the effect of low levels of nano-silica on the physical-rheological properties of different bitumens and reduce the cost of construction of asphalts modified with nano-silica. The rheological tests performed on the unmodified and modified bitumen included: penetration grade, softening point, ductility, rotational viscometer (RV), and flash point. The penetration test was carried out to determine the consistency (hardness) of the bitumen at 25°C according to the ASTM D5 standard. Moreover, the softening point of the bitumen was measured according to the ASTM D36 standard. In order to determine the bitumen ductility,

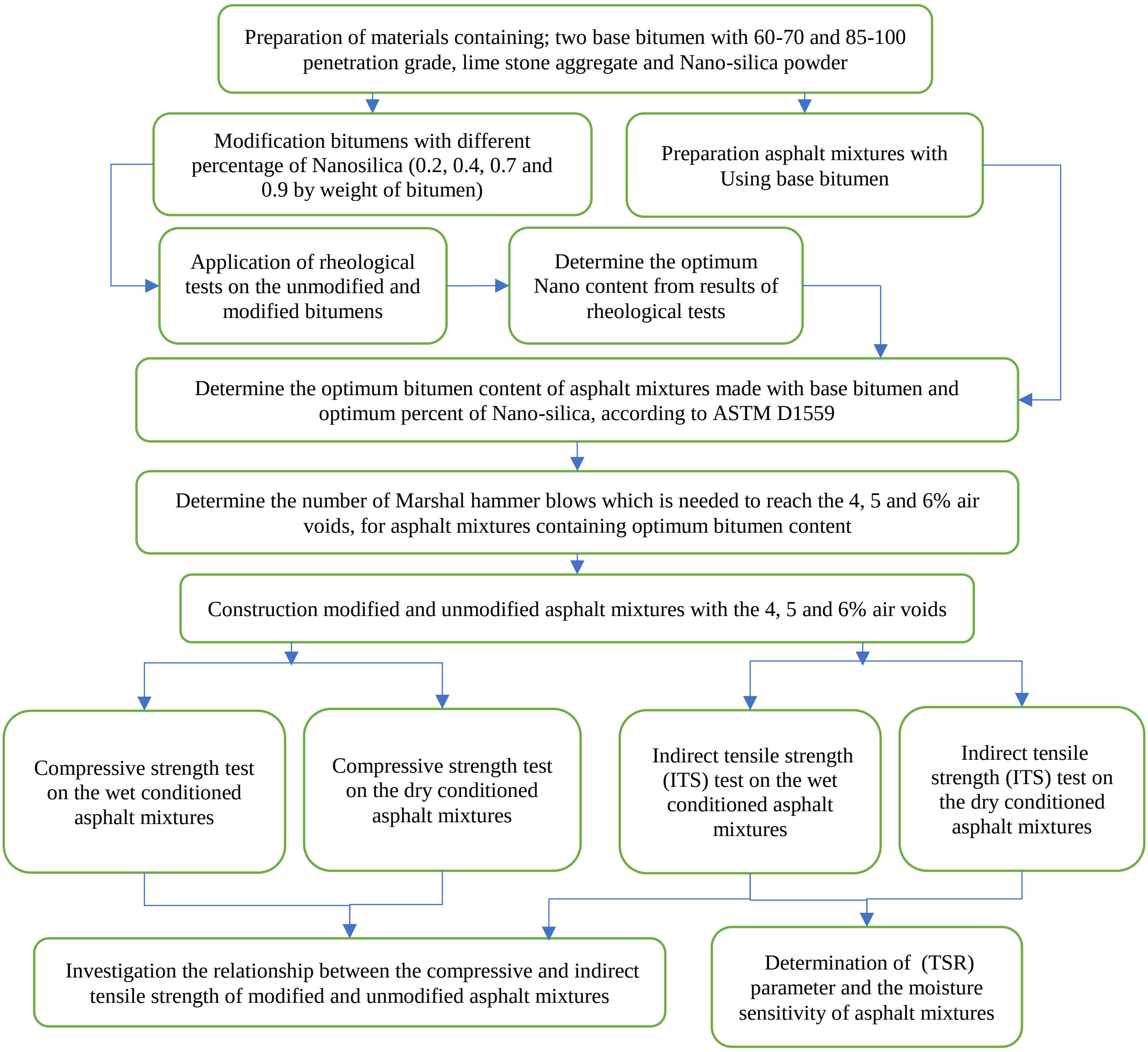

Fig. 1. Experimental program in this study.

which is an indicator of the ultimate tensile strength of the bitumen the ASTM D113 standard was used. In addition, the RV test was performed to measure the apparent viscosity of the bitumen at high temperatures and to ensure bitumen usability according to ASTM D4402. An RV measures the rotational torque required to provide a constant rotational velocity (20 rpm) of a cylindrical spindle submerged in the bitumen binder at a constant temperature. In this study, spindle SC4-27/SD, with a diameter of 11.7 mm, was used, and the rotational viscosity test was performed at 135°C. Also, the flash point test is used to determine the safe mixing and laying temperature for unmodified and modified bitumen with nano-silica according to the ASTM D92 standard.

### 2.3.2. *Modified Lottman Indirect Tension Test*

The AASHTO Standard Method of Test T283, ‘‘Resistance of Compacted Bituminous Mixture to Moisture Induced Damage,” was used to determine the moisture sensitivity of asphalt specimens. In this method, 6 cylindrical asphalt concrete specimens prepare with 7% ± 1 air void. Half of the specimens were selected as conditioned specimens and saturated with water in the vacuum condition of 70 to 80%. These vacuum and saturated specimens are placed in the freezer with temperature -18°C for 16 hours and then placed in a hot bath of 60°C for 24 hours. In the end, to determine the indirect tensile strength, both groups of specimens (wet conditioned and dry conditioned) were loaded until failure, at a loading rate of 50.8 mm/min at 25° and the maximum force to break the sample is measured. The indirect tensile strength is calculated from Eq. (1).

$$ITS = \frac{2\,P_{max}}{\pi\,Dt} \tag{1}$$

where, ITS = indirect tensile strength (KPa). $P_{max}$= total applied vertical load at failure (kg). $t$ = height of specimen (cm). $D$ = diameter of specimen (cm).

Indirect tensile strength ratio (TSR) is obtained from Eq. (2). The ITS value in Eq. (2) is the average value for specimens in wet and dry conditions. The TSR value should be at least 80%, and the higher TSR value indicates the higher resistance to moisture sensitivity [13,41,42].

$$TSR = \frac{ITS\ wet}{ITS\ dry} \times 100 \tag{2}$$

### 2.3.3. *Compressive strength test*

Compressive strength was determined in accordance with ASTM D1074. In this method, the compressive strength of the compacted asphalt mixtures was measured with uniaxial compression testing, and the specimens were loaded until failure, at a loading rate of 50.8 mm/min.

### 2.4. *Statistical analysis*

Multivariate analysis of variance (MANOVA) is an extension of the univariate analysis of variance (ANOVA) that used to analyze data that involves more than one dependent variable at a time. Numerous researchers have utilized MANOVA to analyze effect parameters on mechanical properties of asphalt mixture, cement mortar, and concrete [43-46]. In this study, the MANOVA was conducted to evaluate the effect of the factors bitumen type (60-70 and 85-100 pen grade), modification (modified and unmodified), air void content (4, 5 and 6%), and the interaction between them as independent variables on the ITS and $F_c$ values of asphalt mixtures as dependent variables. The significance level and confidence value were selected α = 0.05 and 95% in the statistical study, respectively. Factors with a p-value lower than 0.05 are construed as statistically significant, and the factors are effected on the results.

## 3. Results and discussions

### 3.1. *Physical-rheological properties of bitumen samples*

Figs. 2-6 illustrates the results of the penetration grade, softening point, ductility, RV, and flash point of the modified bitumen samples for the two types of bitumen (60-70 and 85-100 pen grade), respectively. As observed in Fig. 2, the penetration grade decreases in both types of bitumen, with an increase in the nano-silica content from 0 to 0.7%. This can be due to the high strengthening property of nano-silica, which increases the asphaltene part of the bitumen against the maltene phase [47,48]. Also, Azarhoosh et al. [25] evaluated the effect of nano-$TiO_2$ on physical-rheological properties of 85-100 pen grade bitumen and reported the 6% nano-$TiO_2$ modification increases 7% the penetration grade of bitumen 85-100. The results of the softening point tests are depicted in Fig. 3. Overall, according to this figure, it can be concluded that by increasing the amount of nano-silica from 0 to 7%, the softening point of the 60-70 and 85-100 pen grade bitumens increase 8 and 6 %, respectively. In other words, the temperature sensitivity of the bitumen has been decreased by increasing nano-silica. Azarhoosh et al. [25] reported the 6% nano-$TiO_2$ modification increases 10% the softening point of bitumen 85-100. It can be seen in Fig. 4, the stiffness of the bitumen increased with increasing nano-silica content in bitumen, and this reduced the elasticity and ductility of the bitumen. But, Azarhoosh et al. [25] reported the 6% nano-$TiO_2$ modification increases 5% ductility of bitumen 85-100. It is also observed in Fig. 5 that the viscosity of modified bitumen decreases but has an increasing trend with an increase in nano-silica content from 0 to 0.7%. According to Fig. 5, the 0.7% nano-silica modification increases 41 and 59% the viscosity of the 60-70 and 85-100 pen grade bitumens, respectively. Also, Azarhoosh et al. [25] reported the 6% nano-$TiO_2$ modification increases 33% the viscosity of bitumen 85-100. The results of the presented study showed that the viscosity of the 85-100 pen grade bitumen increased more than the study performed by Azarhoosh et al. [25]. Fig. 6 shows the results of the flash point test on unmodified and modified bitumen samples. The results show that the flash point of both types of bitumen increased with increasing nano-silica content from 0 to 0.7%, and the 0.7% nano-silica modification increases 4.5 and 4% the flash point of the 60-70 and 85-100 pen grade bitumens, respectively. Also, Azarhoosh et al. [25] reported the 6% nano-$TiO_2$ modification increases 8% the flash point of bitumen 85-100.

Overall, based on Figs. 2-6, the penetration grade and ductility decreased, while the RV, softening point, and flash point increased with increasing the nano-silica content from 0 to 0.7%. Thus, in this study, the optimal amount of nano-silica was 0.7% and was used to prepare the asphalt mixes.

### 3.2. *Results of the modified Lottman test (AASHTO T283)*

The ITS results of unmodified and modified asphalt mixtures with nano-silica in dry and wet conditions for the two types of bitumen (60-70 and 85-100 pen grade) are shown in Fig. 7(a) and 7(b), respectively. Evidently, the highest ITS value occurred at void = 4% for all the specimens (unconditioned and moisture-

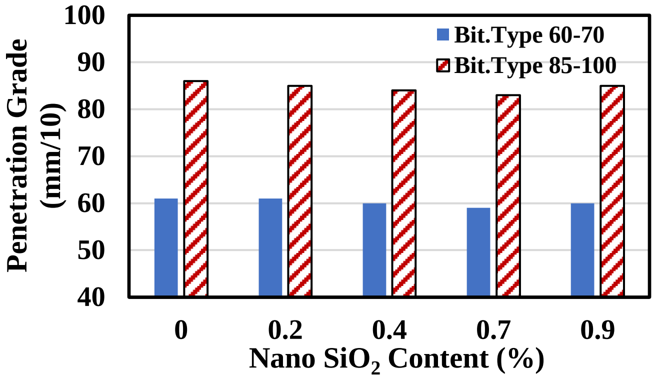

Fig. 2. Penetration test results on unmodified and modified bitumen samples

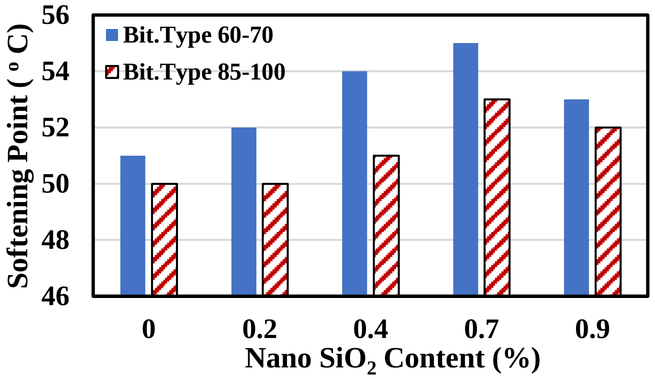

Fig. 3. Softening point test results on unmodified and modified bitumen samples.

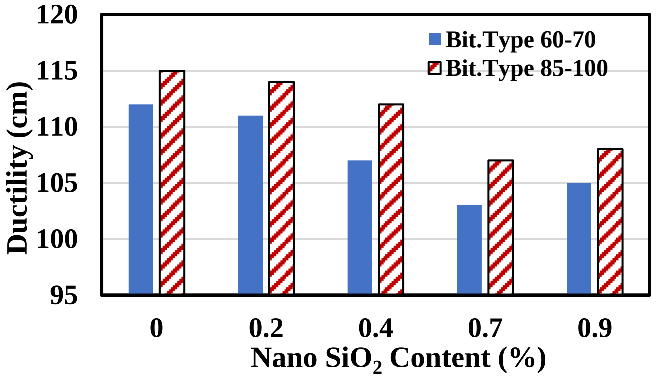

Fig. 4. Ductility test results on unmodified and modified bitumen samples.

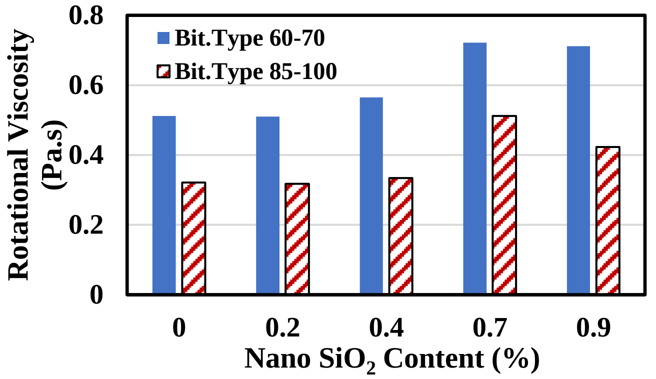

Fig. 5. Rotational viscometer test results at 135 ℃ on unmodified and modified bitumen samples.

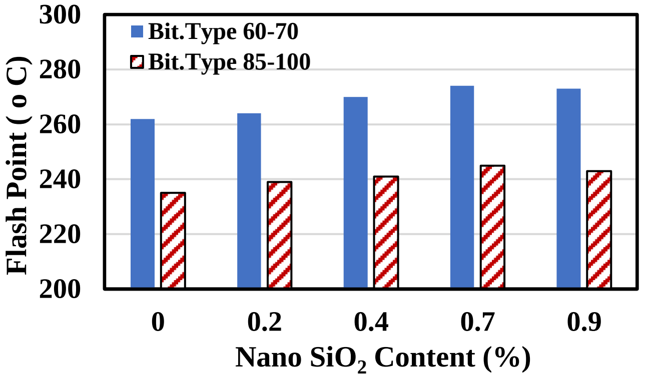

Fig. 6. Flash point test results on unmodified and modified bitumen samples.

conditioned). This finding is observed because the stiffness of the mixture increases with the decrease of air void content of asphalt mixture. As a result, the ITS values of the mixture increase at void = 4%. Also, it can be observed from Fig. 7 that, regardless of the type of moisture condition applied to the specimen (dry and wet-conditioned) and the bitumen type (unmodified and modified), with an increase of air voids in the mixtures, a decrease is observed in ITS values for all the mixes. This can be due to an increased air void content in the mixture, which decreases the cohesion and strength of the mixture and increases the penetration of water. This penetrating water decreases the adhesion between the bitumen and aggregates and separates the bitumen layer from the aggregate surface. It also changes the bitumen properties and decreases bitumen cohesion [49,50].

It can be observed from Fig. 7 that the modified mixtures had higher ITS values than the unmodified mixtures at the same air void, and the Mod-Dry and Mod-Wet mixtures had the highest strength, respectively. This means that the addition of nano-silica increased the tensile strength of the mixture due to the increased viscosity and bitumen hardness and, consequently, increased the adhesion between the bitumen and aggregates [51]. Moreover, the modified mixtures in the wet condition (Mod-Wet) had a higher ITS value than the unmodified mixtures in the dry condition (Unmod-Dry), at almost all air voids. This illustrates the significant role of nano-silica in increasing the tensile strength of the mixture in wet conditions, and it can be concluded that the addition of nano-silica increases the adhesion and cohesion of the bitumen, leading to more hard separation of aggregates from the asphalt binder in the presence of water.

Overall, in Fig. 7a and b, the ITS values of asphalt mixtures made with 60-70 pen grade bitumen are higher than those of asphalt mixtures made with 85-100 pen grade bitumen, which may be due to the higher viscosity and hardness of the 60-70 than 85-100 pen grade bitumen. Also, the ITS results of all the specimens showed that the modification of bitumen with nano-silica significantly increased the ITS strength of the specimens in dry and wet conditions.

Using the ITS results shown in Fig. 7 and Eq. (2), the TSR of the mixtures was calculated and is presented in Fig. 8. TSR is one of the important parameter in the evaluation of the moisture sensitivity of asphalt mixtures, and its higher values indicate the

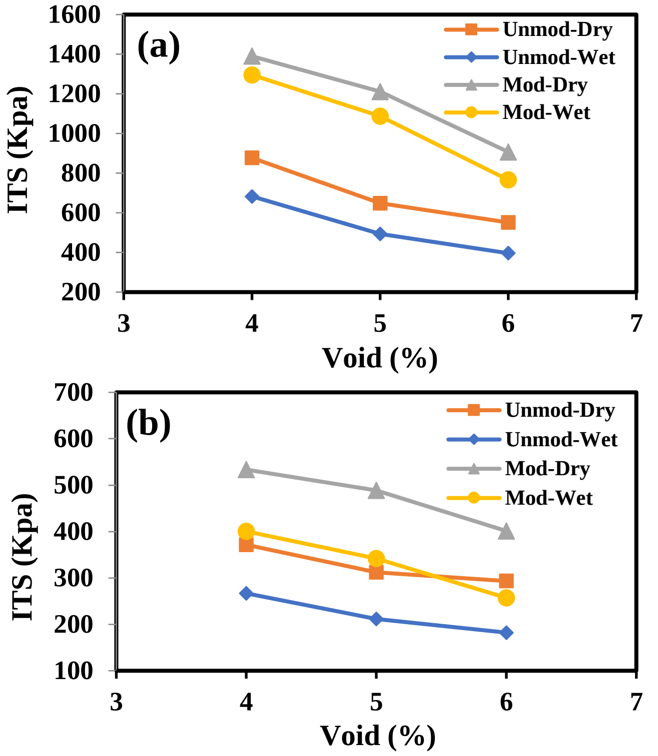

Fig. 7. Variation of ITS with air void in modified and unmodified specimens with, (a) 60-70 bitumen, and (b) 85-100 bitumen.

higher moisture resistance of asphalt mixtures. The minimum acceptable TSR is 80-70% [13,41].

Based on Fig. 8, the TSR values decreased in all the mixtures by an increase in the air void content from 4 to 6%. This is due to the increased water penetration into the mixture, which decreased the adhesion of bitumen-aggregate and the bitumen cohesion through the formation of water emulsions in the bitumen [52]. It can also be observed in Fig. 8 that the modification of asphalt with nano-silica improved the moisture sensitivity of the asphalt mixture with 60-70 and 85-100 pen grade bitumen. Because, as observed in Section 3.1, nano-silica decreases the penetration grade and increases the viscosity and hardness of the bitumen, which can reduce the water diffusion into the asphalt binder, increase the bitumen cohesion, and make bitumen harder to remove from the aggregate surface [15,51]. The results also demonstrated that the TSR values of the unmodified and modified mixture for the 60-70 pen grade bitumen are higher than those of the 85-100 pen grade bitumen, which is due to the higher viscosity of the 60-70 than 85-100 pen grade bitumen as well as the slower movement of the asphalt binder from the aggregate surface.

Fig. 9 shows the percentage of increase of TSR from the unmodified to the modified mixture at various voids. The percentage of increase of TSR is calculated from Eq. (3).

$$\%\ increase\ of\ TSR = \frac{A_2 - A_1}{A_1} \times 100 \qquad (3)$$

where, $A_2$ = TSR of modified mixture. $A_1$= TSR of unmodified mixture.

According to Fig. 9, the percentage of increase of TSR for mixtures with the 60-70 is higher than the mixtures with the 85-100 pen grade bitumen. This indicates that nano-silica is more effective on the 60-70 than 85-100 pen grade bitumen. Also, the results revealed that the highest percentage of increase of TSR occurred at void = 4%, and the percentage of increase of TSR decreased with an increase in the air void content (more than 4%) for all the specimens.

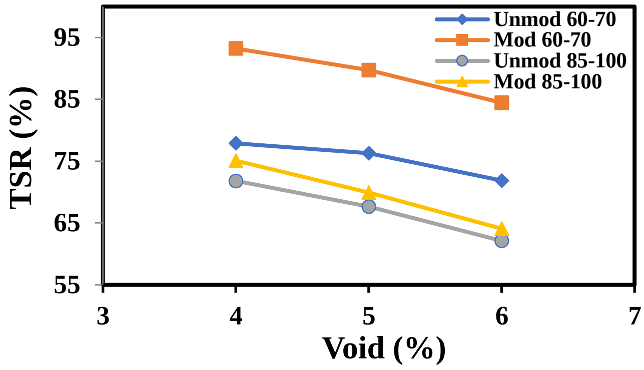

Fig. 8. Variation of TSR with air void in modified and unmodified mixtures.

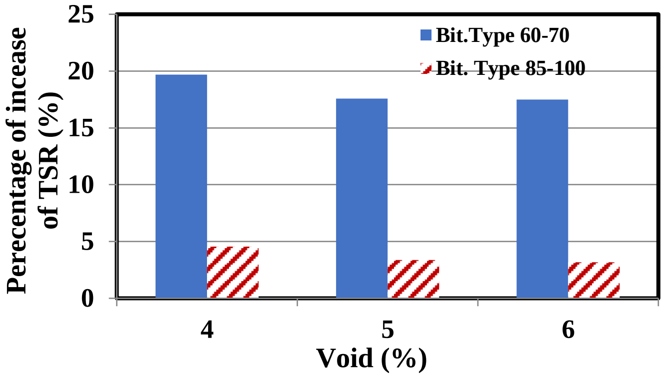

Fig. 9. The percentage of increase of TSR from unmodified to the modified mixtures at various air voids.

### 3.3. *Compressive strength test results*

Fig. 10 illustrates the compressive strength results of unmodified and modified asphalt mixtures with nano-silica in dry and wet conditions for two bitumens, 60-70 and 85-100 pen grade. As observed in Fig. 10, upon modifying the bitumen by nano-silica, the compressive strength increased. This increase was to the extent that the modified mixture in the wet condition (Mod-Wet) had a higher compressive strength at almost all air void percentage compared to the unmodified mixture in the dry condition (Unmod-Dry). In other words, nano-silica through increasing the stiffness of bitumen increases the compressive strength of the specimens in dry and wet conditions. As observed in Fig. 10, the compressive strength of all the specimens decreased by increasing the air void content from 4 to 6%, which is due to the decreased density and cohesion of the asphalt mixture. Moreover, comparing the results of Fig. 10a and b, it can be concluded that the compressive strength values of the bitumen prepared with the 60-70 are higher than the 85-100 pen grade bitumen in the same conditions, which can be due to the higher stiffness and viscosity of 60-70 than the 85-100 pen grade bitumen. Overall, according to Fig. 10, it can be concluded that using optimum amount of nano-silica increases the compressive strength of asphalt mixtures, and the increased air void content negatively affects the compressive strength of asphalt mixtures.

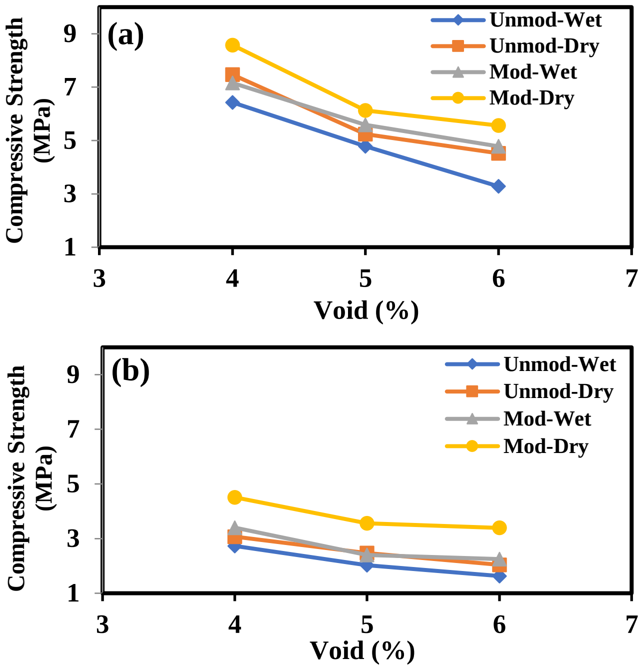

Fig. 10. Variation of compressive strength with air void in unmodified and modified specimens with, (a) 60-70 bitumen, and (b) 85-100 bitumen.

### 3.4. *Relationship between ITS and Compressive strength results*

According to Fig. 11, the relationship between the ITS and the compressive strength of unmodified and modified asphalt mixtures was investigated by the use of the experimental data obtained by the study. The non-linear regression model with power function has been used to develop Eqs. (4) and (5). Eqs. (3) and (4) are presented for the unmodified and modified mixtures with a regression of $R^2 = 0.9528$ and $R^2 = 0.943$, respectively.

$$ITS_{Unmod} = 119.69(F_{CUnmod})^{0.9741} \tag{4}$$

where, $ITS_{Unmod}$= indirect tensile strength of umodified asphalt mixtures (KPa). $F_{CUnod}$= compressive strength of umodified asphalt mixtures (MPa)

$$ITS_{Mod} = 87.432(F_{CMod})^{1.3531} \tag{5}$$

where, $ITS_{Mod}$= indirect tensile strength of modified asphalt mixtures (KPa). $F_{CMod}$= compressive strength of modified asphalt mixtures (MPa)

Fig. 11 showed the ITS and $F_c$ of unmodified and modified asphalt mixtures are related with a high relationship coefficient. It is also observed from Fig. 11 that the ITS value of both asphalt mixtures (modified and unmodified) increased with an increase in the compressive strength. Also, the ITS values of the modified mixture was higher than the unmodified mixture. This indicates the positive effect of nano-silica on improving the mechanical properties of asphalt mixtures.

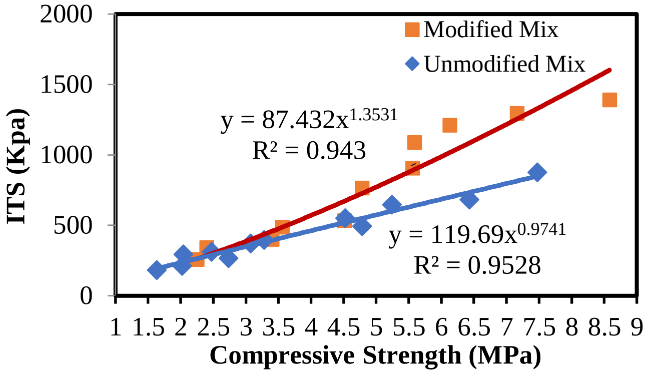

Fig. 11. The relationship between ITS and Compressive strength for unmodified and modified mixtures.

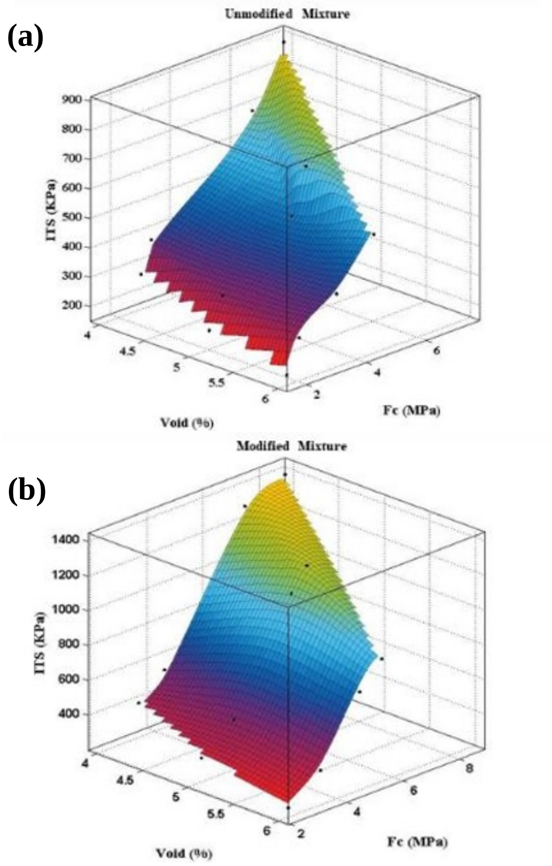

Fig. 12. 3D interpretation of ITS vs. air void and compressive strength in (a) unmodified mixtures, and (b) modified mixtures.

### 3.5. *3D interpretation of different variables variations used in this study*

Fig. 12(a) and 12(b) display a three-dimensional (3D) schematic diagram of the ITS changes, based on the air void content and compressive strength of the unmodified and modified asphalt mixtures, respectively. Evidently, the air void percentage of the mixture strongly affected the ITS value of the unmodified and nano-silica-modified asphalt mixture. By increasing the percentage of air voids in the mixture, the ITS of the specimen decreased. Furthermore, based on Fig. 12, the higher the compressive strength of the asphalt specimens, the higher their ITS value, and at higher compressive strengths, by decreasing the air void content, the ITS rate increased. It can be observed from Fig. 12(b) that the modified mixture has a higher increase rate in compressive strength and ITS value than the unmodified mixtures at each air void.

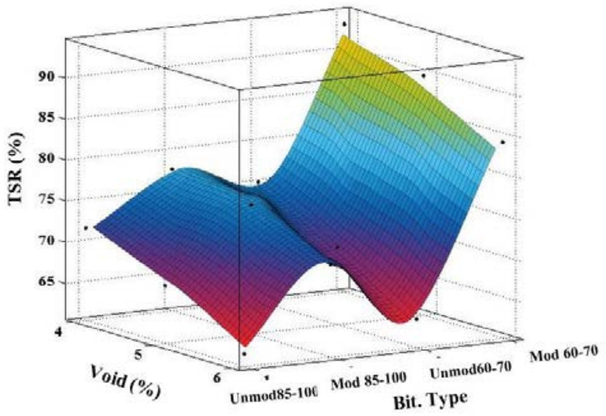

Fig. 13. 3D interpretation of TSR vs. air void and bitumen type in asphalt unmodified and modified mixtures.

Fig. 13 shows a 3D interpretation of the moisture sensitivity of the asphalt mixture vs. bitumen type and air void content. The moisture sensitivity of the asphalt mixture was highly dependent upon the air voids content in the asphalt mixture and the type of bitumen used. Moisture sensitivity decreased with decreasing the air void content. Moreover, for both types of bitumen (60-70 and 85-100 pen grade), the modification of bitumen with nano-silica increased the TSR value. In other words, the TSR of the mixture modified with the 85-100 pen grade bitumen (Mod 85-100) was higher than that of both mixtures prepared with unmodified 60-70 and 85-100 pen grade bitumens. Also, the mixture made with modified 60-70 pen grade bitumen had the highest TSR value and the best moisture sensitivity performance.

### 3.6. Results of Statistical Analysis

The results of the MANOVA analysis are presented in Table 7. The results showed that bitumen type, modification and air void content had significant effects on variables of ITS and $F_c$ because their P-values are less than the $\alpha = 0.05$. Also, the interaction between bitumen type and modification had a significant effect ($p < 0.05$) on the variable of ITS. However, do not significantly affect ($p > 0.05$) on $F_c$. But, the interaction between modification and air void content, and the interaction between bitumen type, modification and air void content was not significant on the variables of ITS and $F_c$.

## 4. Conclusion

The main objective of this study was to investigate the use of nano-silica in improving the properties of different bitumens and increasing the moisture resistance of asphalt mixtures at different traffic loading conditions. After physical and rheological testing on nano-silica modified bitumen, ITS, and compressive strength tests were performed on the asphalt mixtures, and the results of the experiments were compared. The following results were obtained:

1. Generally, the penetration grade and ductility decrease, while RV, softening point, and flash point increase with increasing the nano-silica content in the bitumen, and the optimal amount of nano-silica is 0.7%.
2. Nano-silica improves the moisture sensitivity of asphalt mixtures with 60-70 and 85-100 pen grade bitumens at all air void contents (4, 5, and 6%). In other words, nano-silica can be used to decrease the moisture sensitivity of asphalt mixtures at different traffic loading conditions.
3. The ITS values of asphalt mixtures made with the 60-70 pen grade bitumen are higher than the asphalt mixtures made with the 85-100 pen grade bitumen. Also, the ITS results of all the specimens showed that the modification of bitumen with nano-silica significantly increases the ITS values of the specimens in dry and wet conditions. Thus, the modified mixtures in the wet condition (Mod-Wet) has a higher ITS value than the unmodified mixtures in the dry condition (Unmod-Dry) at almost all air void contents.

Table 7
MANOVA analysis results on main and interaction effects on ITS and $F_c$.

| Source of variation | Dependent Variable | Type III Sum of Squares | DF | Mean Square | F | Sig. |
|---|---|---|---|---|---|---|
| A= Bitumen Type | ITS | 1626302.3 | 1 | 1626302.3 | 174.915 | 0 |
| | $F_c$ | 54.15 | 1 | 54.15 | 128.537 | 0 |
| B = Modification | ITS | 599610.09 | 1 | 599610.09 | 64.49 | 0 |
| | $F_c$ | 5.636 | 1 | 5.636 | 13.378 | 0.003 |
| C= Air void | ITS | 266081.52 | 2 | 133040.76 | 14.309 | 0.001 |
| | $F_c$ | 16.659 | 2 | 8.33 | 19.772 | 0 |
| A * B | ITS | 206035.07 | 1 | 206035.07 | 22.16 | 0.001 |
| | $F_c$ | 0.012 | 1 | 0.012 | 0.028 | 0.87 |
| A * C | ITS | 87990.187 | 2 | 43995.094 | 4.732 | 0.031 |
| | $F_c$ | 3.25 | 2 | 1.625 | 3.857 | 0.051 |
| B * C | ITS | 23861.482 | 2 | 11930.741 | 1.283 | 0.313 |
| | $F_c$ | 0.113 | 2 | 0.057 | 0.134 | 0.876 |
| A * B * C | ITS | 7394.351 | 2 | 3697.175 | 0.398 | 0.68 |
| | $F_c$ | 0.045 | 2 | 0.022 | 0.053 | 0.949 |
| Error | ITS | 111571.93 | 12 | 9297.66 | - | - |
| | $F_c$ | 5.055 | 12 | 0.421 | - | - |
| Total | ITS | 11529652 | 24 | - | - | - |
| | $F_c$ | 526.876 | 24 | - | - | - |

Note: * denotes interaction.

4. The TSR value decreases with the increases of the voids from 4 to 6% in all the mixtures. The results also show that the TSR values of the unmodified and modified mixture with 60-70 are higher than the 85-100 pen grade bitumen, which is due to the higher viscosity of 60-70 than the 85-100 pen grade bitumen and the slower movement of the asphalt binder from the aggregate surface.
5. The percentage increase of TSR for mixtures with the 60-70 pen grade bitumen is higher than the mixtures with the 85-100 pen grade bitumen. This suggests that nano-silica is more effective on the 60-70 than 85-00 pen grade bitumen.
6. Nano-silica enhances the compressive strength of asphalt mixtures, and an increase in the air void content has a negative impact on the compressive strength of asphalt mixtures.
7. The results of MANOVA analysis indicated that the bitumen type, modification and air void content had significant effects on variables of ITS and $F_{c.}$

## References

[1] A.E.A.E.-M. Behiry, Laboratory evaluation of resistance to moisture damage in asphalt mixtures, Ain Shams Eng. J. 4 (3) (2013) 351-363.

[2] F. Xiao,, W. Zhao,, T. Gandhi,, S. N. Amirkhanian, Influence of antistripping additives on moisture susceptibility of warm mix asphalt mixtures, J. Mater. Civ. Eng. 2010. 22(10) 1047-1055.

[3] B.M. Kiggundu, F.L. Roberts, Stripping in HMA mixtures: State-of-the-art and critical review of test methods, National Center for Asphalt Technology Auburn, AL, USA, 1988.

[4] P.S. Kandhal,, Moisture susceptibility of HMA mixes: identification of problem and recommended solutions. Quality Improvement Publication (QIP) No. 119. National Asphalt Pavement Association, Washington DC, USA, December 1992.

[5] S. Kim, B.J. Coree, Evaluation of hot mix asphalt moisture sensitivity using the Nottingham asphalt test equipment, Iowa State University, Center for Transportation Research and Education, IA, USA, 2005.

[6] R.L. Terrel, T. V. Scholz, A. Al-Joaib, S. Al-Swailmi, Validation of binder properties used to predict water sensitivity of asphalt mixtures (with discussion), J. Assoc. Asphalt Paving Technol. 62 (1993) 172-222.

[7] H. Plancher, S. Dorrence, J. Petersen, Identification of chemical types in asphalts strongly adsorbed at the asphalt-aggregate interface and their relative displacement by water [Moisture damage to roads], Energy Research and Development Administration, Laramie Energy Research Center, Laramie, WY, USA, 1977.

[8] W.G. Craig,Variables of the Static Asphalt Stripping Test, Proceedings, The Assocation of Asphalt Paving Technologists, Transportation Research Board, Washington DC, USA, Vol. 27, 1958, pp. 519.

[9] D.N. Little, A. Bhasin, Exploring mechanism of H ealing in asphalt mixtures and quantifying its impact, in Self healing materials, Springer, Dordrecht, Netherlands, 2007, p. 205-218.

[10] J. Ekblad, R. Lundström, E. Simonsen, Water susceptibility of asphalt mixtures as influenced by hydraulically active fillers, Mater. Struct. 48 (4) (2015) 1135-1147.

[11] B.V. Kok, M. Yilmaz, The effects of using lime and styrene–butadiene–styrene on moisture sensitivity resistance of hot mix asphalt, Constr. Buil. Mater. 23 (5) (2009) 1999-2006.

[12] R. Gubler, M. N. Partl, F. Canestrari, A. Grilli, Influence of water and temperature on mechanical properties of selected asphalt pavements, Mater. Struct. 38 (5) (2005) 523-532.

[13] M. Solaimanian, R.F. Bonaquist, V. Tandon, Improved conditioning and testing procedures for HMA moisture susceptibility, Transportation Research Board, Washington DC, USA, Vol. 589, 2007.

[14] P.E. Sebaaly, Comparison of lime and liquid additives on the moisture damage of hot mix asphalt mixtures, National Lime Association, Arlington, VA, USA, 2007.

[15] G.H. Hamedi, F. Moghadas Nejad, K. Oveisi, Investigating the effects of using nanomaterials on moisture damage of HMA, Road Mater. Pavement Des. 16 (3) (2015) 536-552.

[16] P. Tohme, P. E. Sebaali, E. Y. Hajj, D. Johnston, Effectiveness of antistrip additives for bituminous mixtures. Inter. J. Pavements 3 (2) (2004).

[17] P.E. Sebaaly, D. Little, E. Y. Hajj, A. Bhasin, Impact of lime and liquid antistrip agents on properties of Idaho hot-mix asphalt mixture, Transp. Res. Rec. 1998 (1) (2007) 65-74.

[18] K.P. Thomas, J.F. McKay, J.F. Branthaver, Surfactants in aged asphalt and impact μ on moisture susceptibility of laboratory-prepared mixes, Road Mater. Pavement Des. 7 (4) (2006) 477-490.

[19] M.C. Won, M.K. Ho, Effect of antistrip additives on the properties of polymer-modified asphalt binders and mixtures, Transp. Res. Rec. 1436 (1994) 108-114.

[20] G.H. Hamedi, A. Sahraei, M.R. Esmaeeli, Investigate the effect of using polymeric anti-stripping additives on moisture damage of hot mix asphalt, European J. Environ. Civ. Eng. (2018) https://doi.org/10.1080/19648189.2018.1517697

[21] C. Gorkem, B. Sengoz, Predicting stripping and moisture induced damage of asphalt concrete prepared with polymer modified bitumen and hydrated lime, Constr. Buil. Mater. 23 (6) (2009) 2227-2236.

[22] M. Vamegh, M. Ameri, S.F.C. Naeni, Experimental investigation of effect of PP/SBR polymer blends on the moisture resistance and rutting performance of asphalt mixtures, Constr. Buil. Mater. 253 (2020) 119197.

[23] J. Shen, B. Huang, X. Shu, B. Tang, Size effect of sub nano-scaled hydrated lime on selected properties of HMA, Inter. J. Pavement Res. Technol. 4 (4) (2011) 252-257.

[24] F.M. Nejad, A. R. Azarhoosh, G. H. Hamedi, M. J. Azarhoosh, Influence of using nonmaterial to reduce the moisture susceptibility of hot mix asphalt, Constr. Buil. Mater. 31 (2012) 384-388.

[25] A. Azarhoosh, F. Moghaddas Nejad, A. Khodaii, Evaluation of the effect of nano-$TiO_2$ on the adhesion between aggregate and asphalt binder in hot mix asphalt, European J. Environ. Civ. Eng. 22 (8) (2018) 946-961.

[26] G.H. Hamedi, F.M. Nejad, K. Oveisi, Estimating the moisture damage of asphalt mixture modified with nano zinc oxide, Mater. Struct. 49 (4) (2016) 1165-1174.

[27] H. Behbahani, H. Ziari, N. Kamboozia, A. M. Khaki, S. M. Mirabdolazimi, Evaluation of performance and moisture sensitivity of glasphalt mixtures modified with nanotechnology zycosoil as an anti-stripping additive, Constr. Buil. Mater. 78 (2015) 60-68.

[28] M. Ameri, M. Vamegh, R. Imaninasab, H. Rooholamini, Effect of nanoclay on performance of neat and SBS-modified bitumen and HMA, Petrol. Sci. Technol. 34 (11-12) (2016) 1091-1097.

[29] Yao, H., et al., Rheological properties and chemical bonding of asphalt modified with nanosilica, J. Mater. Civ. Eng. 25 (11) (2012) 1619-1630.

[30] H. Ezzat, S. El-Badawy, A. Gabr, E. S. I. Zaki, T. Breakah, Evaluation of asphalt binders modified with nanoclay and nanosilica, Proc. Eng. 143 (2016) 1260-1267.

[31] E.H. Fini, P. Hajikarimi, M. Rahi, F. Moghadas Nejad, Physiochemical, rheological, and oxidative aging characteristics of asphalt binder in the presence of mesoporous silica nanoparticles, J. Mater. Civ. Eng. 28 (2) (2015) 04015133.

[32] A. Onochie, E. Fini, X. Yang, J. Mills-Beale, Z. You, Rheological characterization of nano-particle based bio-modified binder, in Transportation Research Board 92nd Annual Meeting, Washington DC, USA, 2013.

[33] F.M. Nejad, H. Nazari, K. Naderi, F. Karimiyan Khosroshahi, M. Hatefi Oskuei, Thermal and rheological properties of nanoparticle modified asphalt binder at low and intermediate temperature range, Petrol. Sci. Technol. 35 (7) (2017) 641-646.

[34] A. Baldi-Sevilla, M. L. Montero, J. P. Aguiar, L. G. Loría, Influence of nanosilica and diatomite on the physicochemical and mechanical properties of binder at unaged and oxidized conditions, Constr. Buil. Mater. 127 (2016) 176-182.

[35] N.I.M. Yusoff, A. A. S. Breem, H. N. Alattug, A. Hamim, J. Ahmad, The effects of moisture susceptibility and ageing conditions on nano-silica/polymer-modified asphalt mixtures, Constr. Buil. Mater. 72 (2014) 139-147.

[36] American Association of State Highway Transportation Officials, AASHTO guide for design of pavement structures. Vol. 1. Washington DC, USA, 1993.

[37] J. Yang, S. Tighe, A review of advances of nanotechnology in asphalt mixtures, Proc. Soci. Behavioral Sci. 96 (2013) 1269-1276.

[38] L.D. Fernández, E. Lara, E.A. Mitchell, Checklist, diversity and distribution of testate amoebae in Chile, European J. Protistology 51 (5) (2015) 409-424.

[39] G. Quercia, H. Brouwers, Application of nano-silica (nS) in concrete mixtures, in 8th fib PhD Symposium in Kgs, , Lyngby, Denmark; 2010, pp. 431-436.

[40] K. Chrissafis, K. M. Paraskevopoulos, G. Z. Papageorgiou, D. N. Bikiaris, Thermal and dynamic mechanical behavior of bionanocomposites: fumed silica nanoparticles dispersed in poly (vinyl pyrrolidone), chitosan, and poly (vinyl alcohol), J. Appl. Polym. Sci. 110 (3) (2008) 1739-1749.

[41] M. Solaimanian, J. Harvey, M. Tahmoressi, V. Tandon, Test methods to predict moisture sensitivity of hot-mix asphalt pavements. in Moisture Sensitivity of Asphalt Pavements-A National SeminarCalifornia Department of Transportation; Federal Highway Administration; National Asphalt Pavement Association; California Asphalt Pavement Alliance; and Transportation Research Board, San Diego,CA, USA, 2003, pp. 77-110.

[42] American Association of State Highway Transportation Officials, Standard specifications for transportation materials and methods of sampling and testing, AASHTO, Washington DC, USA, 2011.

[43] O. Sirin, D. K. Paul, M. S. Khan, E. Kassem, M. K. Darabi, Effect of Aging on Viscoelastic Properties of Asphalt Mixtures, J. Transp. Eng., Part B: Pavements 145 (4) (2019) 04019034.

[44] G.P. He, W.G. Wong, Laboratory study on permanent deformation of foamed asphalt mix incorporating reclaimed asphalt pavement materials, Constr. Buil. Mater. 21 (8) (2007) 1809-1819.

[45] G. Medina, I. S. del Bosque, M. Frías, M. S. de Rojas, C. Medina, Durability of new recycled granite quarry dust-bearing cements, Constr. Buil. Mater. 187 (2018) 414-425.

[46] J. Chi, R. Huang, C. C. Yang, J. J. Chang, Effect of aggregate properties on the strength and stiffness of lightweight concrete, Cem. Concrete Compos. 25 (2) (2003) 197-205.

[47] H. Taherkhani, S. Afroozi, The properties of nanosilica-modified asphalt cement, Petrol. Sci. Technol. 34 (15) (2016) 1381-1386.

[48] G. Shafabakhsh, O.J. Ani, Experimental investigation of effect of Nano $TiO_2/SiO_2$ modified bitumen on the rutting and fatigue performance of asphalt mixtures containing steel slag aggregates, Constr. Buil. Mater. 98 (2015) 692-702.

[49] P. Pell, I. Taylor, Asphaltic road materials in fatigue. in Association of Asphalt Paving Technologists Proc., Transportation Research Board, Washington DC, USA, Vol 381, 1969, pp. 371-422.

[50] S. Caro, E. Masad, A. Bhasin, D. N. Little, Moisture susceptibility of asphalt mixtures, Part 2: characterisation and modelling, Inter. J. Pavement Eng. 9 (2) (2008) 99-114.

[51] J.C. Sprague, Laboratory investigation of anti-stripping admixtures used for promoting wetting power and adhesion between bitumens and aggregates-discussion, in Proceedings-American Society for Testing and Materials, W Conshohocken, PA, USA, 1946,19428-2959.

[52] H.J. Fromm, The mechanisms of asphalt stripping from aggregate surfaces, Ministry of Transportation and Communications, Ontario, Canada, Vol 43, 1974, pp 191-223.