

Modification of Stone Matrix Asphalt with Nano-SiO₂

Mojtaba Ghasemi^a, Seyed Morteza Marandi^{a,b,*}, Majid Tahmooresi^b, Reza Jalal kamali^a,
Reza Taherzade^c

^aCivil Engineering Department, Shahid Bahonar University, Kerman, Iran

^bInternational Center for science, High Technology & Environmental Sciences, Kerman, Iran

^cDepartment of Materials Science and Engineering, Sahand University of Technology, Iran

ABSTRACT

In the present paper, the potential benefits of nano-SiO₂ powder and SBS were reported for the asphalt mixtures used on pavements. A primary study was conducted for determining the physical properties of asphalt cement and modifiers. Five asphalt binder formulations were prepared using various percentages of SBS and nano-SiO₂ powder. Then, Marshall samples were prepared by the modified and unmodified asphalt binders. Additionally, workability tests (Marshall stability, indirect tensile strength, tensile strength ratio and indirect tensile stiffness modulus) were conducted. The results of this investigation indicated that the asphalt mixture modified by 5% SBS plus 2% nano-SiO₂ powder could give the best results in the tests carried out in the current study so that this modification can increase physical and mechanical properties of asphalt binder and mixtures.

KEY WORDS: SMA, Nanotechnology, ITS, TSR, Resilient Modulus.

1. INTRODUCTION

Asphalt is an organic mixture that is widely used in road pavements because of its good viscoelastic properties [1]. Unfortunately, asphalt becomes brittle and liquid at lower and higher temperatures, respectively, which can result in low temperature cracking of pavement and high temperature rutting. Its application is limited by this temperature susceptibility. Therefore, to expand the aggregate performance of asphalts, it is essential to modify asphalts by adding modifiers like polymer, rubber and clay [2–6].

Use of styrene-butadiene-styrene (SBS) as an asphalt modifier was developed by the Shell Chemical Company [7] and it was recognized that adding SBS can improve the physical and mechanical properties and rheological behavior of conventional asphalt compositions [8]. However, the storage stability of SBS-modified asphalt is usually poor at elevated temperatures due to the poor compatibility between SBS and asphalt. A number of methods have been developed for preparing practical storage stable SBS-modified asphalt [9–11]. Since the rediscovery of nanomaterials in 1985 at the Toyota Central R & D laboratory [12] and the subsequent development of polymer nanocomposites, these materials have generated a great deal of interest in the academic and industrial communities. Basically, polymer nanocomposites consist of a blend of one (or more) polymer(s) with various nanomaterials such as nanoclays, carbon nanotubes, etc. [13,14].

Goh, S.W. et al. (2011) found that, in most cases, addition of nanoclay and carbon microfiber improves the mixture's moisture susceptibility performance or decreases the moisture damage potential [15].

Sureshkumar, M.S. et al. (2010) studied the effect of adding clay (as a third component) in polymer modified asphalts in a system which was composed of base asphalt, EVA copolymer and either an organomodified cloisite or dellite clays. It was found that clay had a compatibilizing effect on asphalt and polymer and that the high compatibility between clay and polymer can lead to the better dispersion of the polymer in the asphalt; therefore, it influences the final rheological properties of the systems under study [16].

Baochang, Z. et al. (2009) studied the effect of styrene-butadiene-rubber/montmorillonite (SBR/MMT) modification on the properties of asphalt and found that SBR/MMT modified asphalts were formed an an ideal fine network structure. Their research indicated that the modified asphalts were very stable in a number of SBR/MMT content ranges. Additionally, the modified asphalts exhibited higher complex modulus (G^*) and lower damping factor ($\tan \delta$), which implies that SBR/MMT displays improved viscoelastic properties and resistance to rutting at high temperature [17].

*Corresponding Author: Seyed Morteza Marandi, International Center for science, High Technology & Environmental Sciences, Kerman, Iran,
Email: marandi@uk.ac.ir

Polacco, G. et al. (2008) studied the effect of adding clay as a third component in polymer modified asphalts in a system composed of soft base asphalt and radial SBS block copolymer. This study concluded that the mixing procedure can significantly affect the final rheological properties [18].

In the current study, SBS/nano-SiO₂ modified bitumens were prepared by incorporating nano-SiO₂ into SBS and mixing this into the bitumen. The properties of the modified bitumens were evaluated using the conventional test methods. The effects of SBS/nano-SiO₂ on the properties of modified asphalt mixtures were also investigated.

2. MATERIALS AND METHODS

2.1. Materials

The 60/70 penetration grade bitumen was obtained from Isfahan Mineral Oil Refinery, Isfahan, Iran. Table 1 shows the physical properties of the bitumen.

Table (1) The physical properties of the bitumen

Property	Test method	Quantity
Penetration at 25 °C, 100g, 5 s (deci-millimetre, d-mm)	ASTM D-5	64
Softening Point, ring and ball (°C)	ASTM D36	48
Flash Point, Cleveland open cup (°C)	ASTM D-92	284
Ductility at 25 °C at 5 cm/min (cm)	ASTM D-113	100+
Specific gravity at 25 °C (gr/cm ³)	ASTM D-70	1.021
Loss on heating, wt (%)	ASTM D-6	0.06

SBS was produced by the Yueyang Petrochemical Co., Ltd., China and is a linear polymer, containing 30 wt% styrene with the average molecule weight of 110,000 g/mol. SiO₂ nanopowder with the average diameter of 15 nm was used in the present research. Table 2 demonstrates the properties of the utilized nanoparticles.

Table 2 The properties of nano-SiO₂.

Diameter (nm)	Surface volume ratio (m ² /g)	Density (g/cm ³)	Purity (%)
15±3	160±12	<0.14	>99.9

Aggregate was excavated from a mine located in EkhtiarAbaad, Kerman site, south-east of Iran. In Table 3, the suggested gradation for the SMA mixtures developed by the FHWA-sponsored SMA Technical Working Group (TWG) in publishing IS 118 is demonstrated [19]. The selected gradation in this research was located in the middle of the limits.

Table 3 FHWA SMA TWG Recommended Gradation

Sieve Size, mm	Percent Passing
19.0	100
12.5	85-95
9.5	75 max
4.75	20-28
2.36	16-24
0.600	12-16
0.300	12-15
0.075	8-10
0.020	<3

Calcium carbonate (CaCO₃) was used as a mineral filler and it was passed through the sieve No. 200 and had the specific gravity of 2.635.

2.2. Preparation of SBS/nano-SiO₂ composite and modified asphalt

SBS and nano-SiO₂ were mixed for forming SBS/nano-SiO₂ composites in a two-roll mill at 50 °C for 30 min. The modified bitumens were prepared by a high shear mixer. Bitumen was heated in the mixer until becoming a fluid at 160 °C. Then, SBS/nano-SiO₂ compound was added to the bitumen and the mixture was blended at 3000 rpm for 1 h.

2.3. Physical properties of modified bitumens

The physical properties of the modified bitumens (such as softening point, penetration and ductility) were measured based on ASTM D36, ASTM D5 and ASTM D113, respectively.

2.4. High-temperature storage stability test

The high temperature storage stability of the modified asphalts was tested in the following way: some of the mixed modified asphalt was poured into an aluminum foil tube (32 mm in diameter and 160 mm in height). This tube was then sealed and placed vertically in an oven at 163 °C for 48 h. After that, it was taken out, cooled down to the room temperature and cut into three equal sections. The specimens obtained from the bottom and top sections were applied for evaluating the storage stability of the SBS/nano-SiO₂ modified asphalts, which was done by measuring their softening points. If the difference of the softening points between the bottom and top sections was less than 2.5 °C, the modified asphalt was considered stable under high temperature storage condition. Otherwise, it was labeled unstable.

2.5. SMA sample preparing and mix design

The typical asphalt mixture design method (Marshall (ASTM D1559) program) was used in this study. The optimum asphalt content was selected as having maximum stability, maximum unit weight and the median allowed limits for percent voids of air (VA limits for wearing coarse is 4.0–7.0%). At these three values, the content of the average asphalt cement (AC) was selected and checked in order to satisfy the VA, VMA, stability and flow specification limits. The obtained optimum bitumen content for the control mixtures was 6.3% which was used for preparing all other modified mixes in order to maintain consistency throughout the study. In the SMA mixture design, the Marshall method of mix design is usually used for confirming the satisfactory voids in SMA mixtures. Laboratory specimens are prepared using fifty blows of the Marshall hammer per side. SMA mixtures have been more easily compacted on the roadway to the desired density compared with the effort required for the conventional HMA mixtures [20].

All mix design methods apply density and voids for determining basic HMA physical characteristics. Two different measures of density are typically chosen: bulk density and theoretical maximum density. Then, the following volumetric parameters of the HMA are calculated using these densities: voids in the total mix (VTM), voids in the mineral aggregate (VMA) and voids filled with asphalt (VFA).

2.6. Marshall strength test

The Marshall test is an empirical test in which cylindrical compacted specimens, 100 mm in diameter by approximately 63.5 mm in height, are immersed into the water at 60 °C for 30–40 min. Then, they are loaded to failure using the curved steel loading plates along a diameter at a constant compression rate of 51 mm/min. The Marshall stability value (in KN) is the maximum force recorded during the compression while the flow (in mm) is the deformation recorded at maximum force [21].

2.7. Indirect tensile strength and tensile strength ratio

The indirect tensile strength was determined using the following equation:

$$ITS = 2P_{\max}/(dh) \quad (1)$$

where P_{\max} represents the breaking load (N) of the specimens under diametral compression and d and h are average values of the diameter (mm) and height (m) of the Marshall specimens, respectively.

Resistance to stripping which is directly correlated to the susceptibility of the mixes to the action of water was quantified using diametral compressive strength tests according to the ASTM D 4867 [22]. Resistance to moisture and the effect of SBS and nano-SiO₂ on the moisture-induced damage of asphalt concrete mixtures were also evaluated. For each group of mixtures, three unconditioned (dry) and three conditioned (wet) specimens were tested. Wet specimens were vacuum-saturated with distilled water so that 50–80% of their air voids was filled with water; then, they were wrapped tightly with the plastic film. The specimens were placed into a leak-proof plastic bag containing approximately 3 ml of distilled water. Then, the wet specimens were subjected to one freeze–thaw cycle. One freeze–thaw cycle consisting of freezing for 16 h at -18 °C was followed by soaking in a 60 °C water bath for 24 h. At the end of the cycle, the bag and the wrapping were removed and placed in a water bath for 1 h at 25 °C before being subjected to failure. The indirect tensile strength of dry specimens were directly determined. Dry specimens were only placed in a water bath for 1 h at 25 °C before being subjected to failure. The indirect tensile strength ratio (TSR) was determined using the following equation:

$$TSR = 100 * P_{\text{cond}}/P_{\text{uncond}} \quad (2)$$

where P_{cond} and P_{uncond} are the indirect tensile strength of the wet specimens and dry specimens, respectively. The TSR value must be higher than 0.70 after one freeze–thaw cycle according to the ASTM D4867.

2.8. Indirect tensile stiffness modulus test

The most popular form of stress–strain measurement is the stiffness modulus of asphalt mixtures measured in the indirect tensile mode which is considered a very important performance characteristic for the pavement. It is a measure of the load-spreading ability of the bituminous layers which controls the level of traffic-induced tensile strains at the underside of the road base; such strains are responsible for the fatigue cracking along with the compressive strains induced in the subgrade which can lead to permanent deformation.

The indirect tensile stiffness modulus (ITSM) test defined by BS DD 213 [23] is a non-destructive test which has been identified as a potential means of measuring this property. The ITSM S_m in MPa can be defined as follows:

$$S_m = F(R + 0.27) / (LH) \quad (3)$$

where F is the peak value of the applied vertical load (N), H is the mean amplitude of the horizontal deformation obtained from five applications of the load pulse (mm), L is the mean thickness of the test specimen (mm) and R is the Poisson's ratio (assumed to be 0.35). The test is normally performed at 20 °C.

3. RESULTS AND DISCUSSION

3.1. Physical properties of modified bitumens

Table 4 demonstrates the effect of the nano-SiO₂ and SBS content on the properties of the modified bitumen's. It is shown in this table that penetration and softening point decreases and increases, respectively, by the increase of the percentage of nano-SiO₂ in the modified binders. The decrease and increase in penetration and softening point demonstrate the increased hardness and stiffness of the modified binders.

Table (4) Basic properties of SBS-RGP modified asphalt binders

Property	5%SBS +0.0% nano	5%SBS +0.5% nano	5%SBS +1.0% nano	5%SBS +1.5% nano	5%SBS +2.0% nano
Penetration, (d-mm)	55	54	51	48	46
Ductility (cm)	100+	100+	100+	100+	100+
Storage Stability					
Top ring and ball, (°C)	54.4	54.7	56.7	58.9	59.0
Bottom ring and ball, (°C)	51.5	53.5	55.6	58.0	58.3
Difference	2.9	1.2	1.1	0.9	0.7

The maximum difference of 1.2 in top and bottom softening points are shown in Table 4 for SBS-nano-SiO₂ modified binders implying the improvement of the storage stability of SBS-nano-SiO₂ modified binders.

3.2. Marshall stability and flow test

Table 5 reveals the Marshall stabilities and flows for each of the mixtures. These values were obtained as the average of three samples. As can be seen, Marshall stability increases with the nano-SiO₂ content. It appears that adding nano-SiO₂ induced the increase of the binders' stiffness. Therefore, the stability of the mixtures containing nano-SiO₂, results is at the values higher than those of the control mixtures. Also, it was determined that through applying only SBS and SBS plus nano-SiO₂ together, the Marshall stability values increased by 9% and 83%, respectively.

Table (5) Marshall Test Results of Control and SBS-nano-SiO₂ Asphalt Mixtures

Sample No.	Mixture type	BSG	VTM(%)	VMA(%)	VFB(%)	Stability (kN)	Flow (mm)
1	60/70 binder	2345	4.33	14.92	70.11	6.42	3.4
2	5%SBS +0.0% nano	2333	4.93	17.38	71.65	7.00	3.13
3	5%SBS +0.5% nano	2328	5.85	18.56	68.50	8.86	3.33
4	5%SBS +1.0% nano	2325	5.17	17.83	70.97	10.13	2.90
5	5%SBS +1.5% nano	2302	5.81	18.75	69.01	11.20	3.35
6	5%SBS +2.0% nano	2299	5.70	18.24	68.75	11.74	2.85

3.3. ITS and TSR

Fig.1 shows the indirect tensile strength of the mixtures. According to this figure, it can be concluded that adding SBS and nano-SiO₂ together to the mixtures can improve the adhesion and cohesion of binder; therefore, it provides more reasonable mixtures than the conventional ones.

The tensile strength ratio of the specimens prepared with different nano-SiO₂ contents after one freeze-thaw cycle is shown in Fig. 2. It can be seen that TSR values increase comparatively with the nano-SiO₂ content. The “6” specimens had the highest TSR value as 0.91 and none of the specimens had a TSR lower than 0.7 in this group. Thus, it can be concluded that the nano-SiO₂ is more effective than SBS as far as moisture damage is concerned.

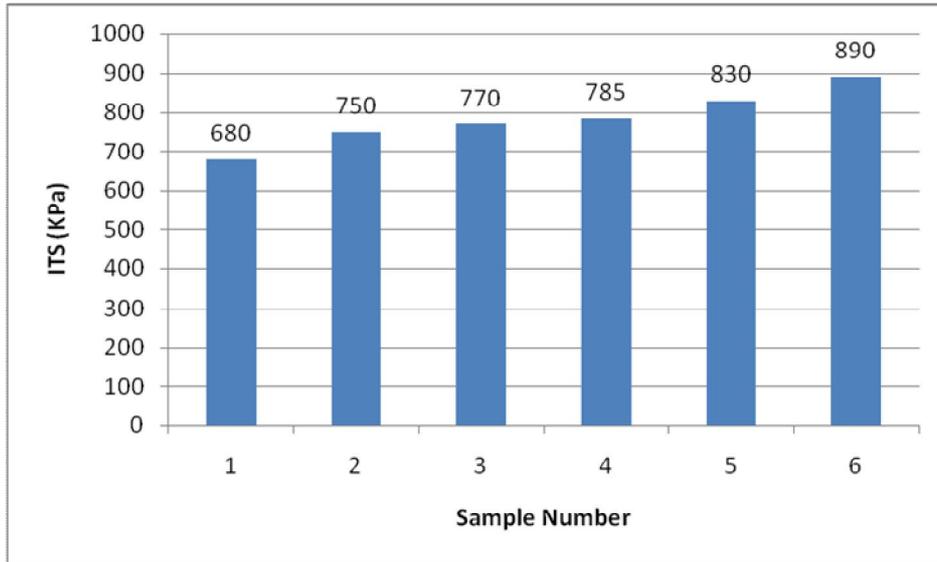


Fig. 1. The indirect tensile strength of the mixtures

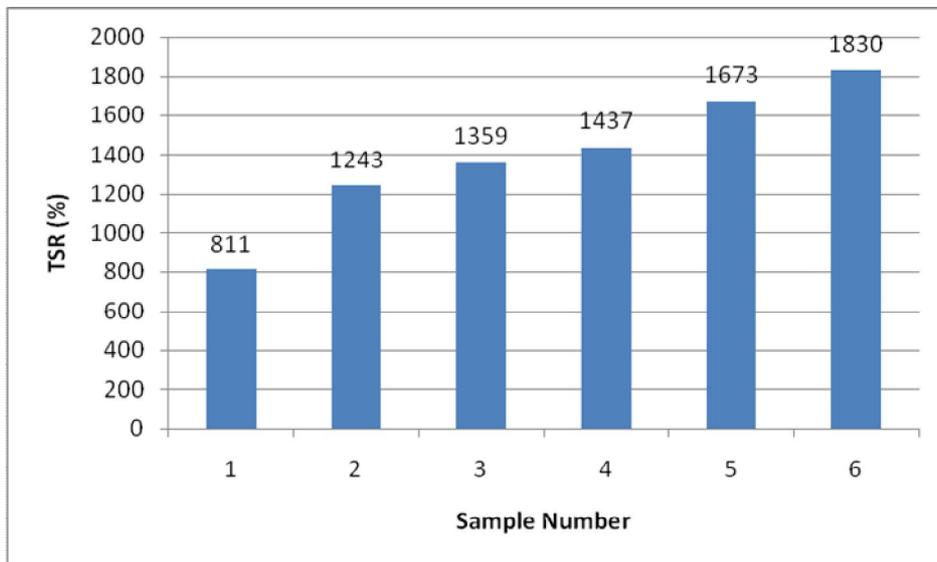


Fig. 2. Tensile strength ratio for prepared specimens

3.4. ITSM

All types of specimens were subjected to the indirect tensile stiffness modulus test (ITSM) at 20 °C. Fig. 3 shows the results of average stiffness modulus from six different types of mixtures. Each of these values was taken from three specimens. The stiffness modulus of the mixtures increased by increasing the nano-SiO₂ content of the conventional mixtures. Moreover, it was determined that the “6” specimens had the highest modulus, 2.3 times

higher than those of the control mixture. The stiffness modulus of the “2” specimens, including only 5% SBS by the weight of total aggregate was approximately 53% higher than that of the control mixture.

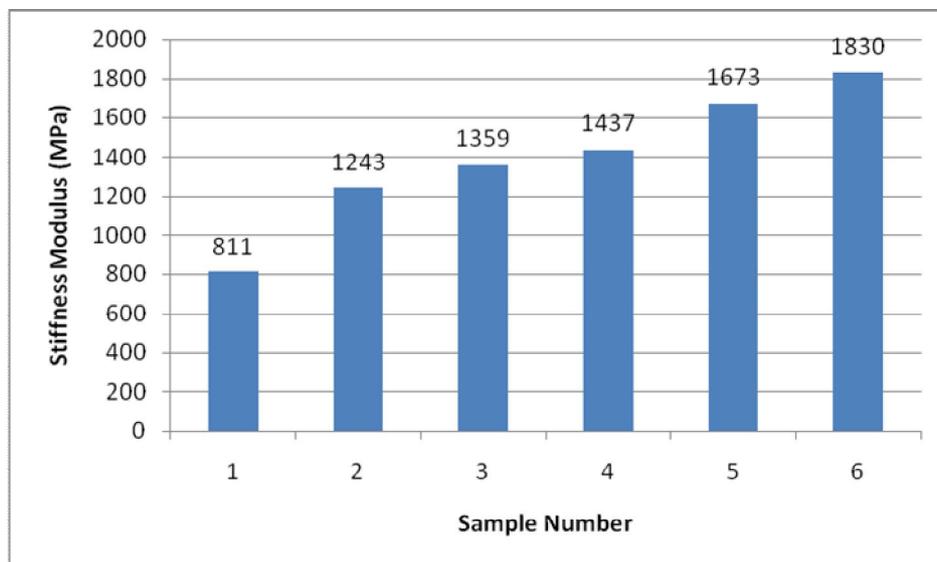


Fig. 3. Stiffness modulus for prepared specimens

4. Conclusion

The main goals of the present research were to characterize the rheological and mechanical properties of SBS/nano-SiO₂ modified asphalt binder and mixtures. Adding modifiers to the pure bitumen improved the viscoelastic behavior of the bitumen and changed its rheological properties. The modifiers included nano-SiO₂ and styrene butadiene styrene (SBS) and had different levels of (decreasing or increasing) influence on the rheological properties of asphalt binder from the same source. After laboratory tests were conducted on the asphalt binder and mixtures with different modifier content and the data were analyzed and the results were compared, the following conclusions were made:

- Nano-polymer-modified bitumen is commonly more viscous than the unmodified binders and tends to show improved adhesive bonding for aggregating the particles.
- Storage stability of SBS/nano-SiO₂ modified binders is higher than that of the control mixtures.
- Stability of the mixtures containing SBS and nano-SiO₂, results in higher values than those of the control mixtures.
- Adding SBS and nano-SiO₂ together to the mixtures improves the adhesion and cohesion of binder; therefore, it provides more reasonable mixtures than the conventional ones.
- Comparatively, TSR values increase with the content of nano-SiO₂.
- The stiffness modulus of the mixtures increased with the increase of the nano-SiO₂ content for the conventional mixtures.
- Among all types of mixtures prepared using SBS and nano-SiO₂, the mixtures with 5% SBS and 2% nano-SiO₂ have the most improved mechanical behavior.

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