Performance Evaluation of Nano-silica Modified Bitumen

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Abstract
In this study, different contents of Nano-silica, 2 wt.%, 4 wt.% and 6 wt.%, have been added to bitumen to modify the physical, mechanical and rheological properties of warm mix asphalt (WMA). WMA is containing 2 wt.% of Sasobit (mixture of long-chain hydrocarbons). Various quality control tests have been carried out to characterize the modified bitumen and WMA. The rheological investigations showed that the complex modulus of base bitumen increases by increasing the percentage of Nano-silica from 2 to 6 wt%. Phase angle and rut factor for the Nano-silica modified bitumen have also decreased significantly. From rheological analysis, 6 wt% Nano-silica has been selected as the optimum content. Results of investigations on the asphalt mixtures demonstrated the fact that by increasing Nano-silica content, the quality of the warm mix asphalt has been improved. By increasing the Nano-silica content, the resilient modulus of WMA has increased; as such, the pavement response under the traffic loading has decreased. By adding Nano-silica to the Sasobit WMA, the depth of cracking has decreased dramatically. So, fatigue life of WMA has been extended in the presence of Nano-silica. Also, the results of wheel tracking test demonstrated that the rutting depth of modified samples have been reduced. The results on WMA showed that 6 wt.% of Nano-silica is the optimum content. This result was in compliance with the rheological investigations.

Keywords: WMA, Sasobit, nano-silica modified bitumen, rheological properties, resilient modulus, fatigue, rut depth.
1. Introduction

WMA has become quite attractive for many researchers during the recent decades. The researchers try to improve the quality of this type of asphalt mixture. Due to its environmental advantages and low fuel consumption, this type of asphalt mixture could be offered as an alternate for the hot mix asphalt (HMA) [Qin et al. 2014]. The most important advantage of WMA is reducing required energy for heating of materials in mixing period, and this issue results in lower cost. Other advantages of WMA compared to the normal HMA are less required asphalt fumes during mixing and lay-down, as well as reduction of emissions of toxic gas from asphalt plants [Zhao and Guo 2012]. For these reasons, usage of WMA has been increased rapidly in recent years. [Qin et al. 2014 and Sampath, 2010].

The National Center for Asphalt Technology published some reports regarding the use of Sasobit, a synthetic wax, as a modifier in warm mix asphalt. In some researches, Sasobit have been used as much as 2.5% base on the base binder, noting that this proportion should not exceed 3 wt%. In some other research projects, proportions of 1% to 1.5% have been recommended. Sasobit decreases the compaction and mix temperatures, and as a result it reduces the required heat. Also, it reduces the CO2 emission significantly, if combined with the recovered bitumen from RAP [Yan et al. 2013, Zhao and Guo 2012].

A nano particle has at least one dimension lower than 100 nanometer (nm) [Golestani et al. 2012, Yang and Tighe 2013, You et al. 2011, Yu et al. 2010]. Regarding to their small size, nanomaterials have usually higher reactivity and higher specific surface area [Yu et al. 2015]. Due to the above-mentioned benefits for nanomaterials, they are suitable for utilization in paving technology [Arabani and Faramarzi 2015; Galooyak et al. 2010; Golestani et al. 2015; Jamal Khattak et al. 2013; Karahancer et al. 2014]. In recent years, Nanoclay material has been used to improve the various properties of base binder. It was found that, Nanoclay could enhance the complex modulus and decrease the failure strain of base binder [Jahromi and Khodaii 2009; Ziari et al. 2014]. Furthermore, it could reduce the moisture susceptibility of final asphalt concrete [Yang and Tighe 2013, Yao et al. 2012a, You et al. 2011].

The carbon nano-fiber has also been utilized for modification of the asphalt binder [Shafabakhsh et al. 2014; Yao et al. 2012b; Ziairi et al. 2013]. From the results of dynamic shear rheometer (DSR) and viscosity tests, it has been concluded that the viscoelastic response and rutting resistance of carbon nano-fibers modified binder have been improved [Khattak et al. 2012, Shafabakhsh et al. 2015, Yao et al. 2012b].

Nano-silica is used in several industries including medicine and engineering. The benefits of Nano-silica are the low production cost and high performance features. Nano-silica has large surface area, strong adsorption, good dispersal ability, high chemical purity, and excellent stability. Due to these beneficial properties, Nano-silica has the potential to be used as an asphalt modifier for modification of asphalt performance [Lazzara and Milioto 2010, Yao et al. 2012b]. The combination of Nano-SiO2 and SBS has been utilized with stone matrix asphalt (SMA), and the results implied that the physical and mechanical properties of asphalt binders and mixtures have been improved [Santagata et al. 2015, Yao et al. 2013]. Lu and co-authors (2013) investigated the effect of Nano-SiO2 modified asphalt mixture. In this research, 5% of Nano-SiO2 was selected as the optimum percent [Lu et al. 2013]. Nano-SiO2 increased the moisture resistance of mixture, significantly. But, it had negative effect on the low temperature cracking of mixtures. 59.5% modification was observed for the samples containing Nano-SiO2 compared to the base bitumen. The effect on Nano SiO2 and Nano TiO2 on the performance of neat asphalt was determined by Chao and co-authors (2009) [Chao and Huaxin, 2009]. Different samples contains 0.5, 1, 3 and 5 wt% of SiO2 were prepared and physical properties, softening points and needle penetration, were determined. By increasing the Nano SiO2, the penetration decreased and softening point and penetration index increased, which indicates the modification in properties on base bitumen. Arabania and colleagues investigated the fatigue properties of modified asphalt mixtures with Nano-SiO2 after short term aging condition. In this study, by varying the nano contents from 2 to 6 wt%, the fatigue properties were assessed by indirect tensile strength device. According to this research, 4wt% nano SiO2 could increase the fatigue life of asphalt mixture to 37%. Therefore the sample prepared...
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with 4% nano SiO$_2$ showed the superior fatigue resistance [Arabania et al. 2012].

The effect of Nano-silica on the properties of Sasobit modified WMA has been investigated in this study. Nano-silica with different contents (including 2, 4 and 6 wt.%) has been added to the Sasobit WMA and various qualification test methods have been conducted on the prepared samples to determine the effect of Nano-silica on the rheological, physical and mechanical properties of prepared samples. The results showed that, 6% Nano-silica improves the rheological properties of base binder, significantly. The results of characterization of Nano-silica modified Sasobit WMA were in compliance with the rheological results.

2. Materials

2.1 Nano-silica

The Nano-silica (Sigma-Aldrich, USA) with the properties presented in Tables 1 and 2 has been used in this study. As shown in Table 2, the surface area of Nano-silica was too high to influence the properties of binder.

2.2 Bitumen and Sasobit

The most common bitumen grade, 60/70 penetration grade, was supplied from an Iranian refinery and used as a base binder in this study. The characteristics of the base bitumen are been presented in Table 3. Flake Sasobit, a synthetic wax with the long-chain hydrocarbons, produced by a Fischer-Tropsch synthesis, was prepared from SasolWax (SasolWax Co., South Africa).

3. Experimental Method

In this study, the hot melting method has been used for mixing the base bitumen with modifiers, Sasobit and Nano-silica. In hot melting method, the bitumen modifiers have been introduced to the neat binder at the elevated temperature (160 ºC for Sasobit and 180 ºC for the Nano-silica). 2 wt% of Sasobit has been utilized for all of the samples, while the Nano-silica was varied from 2 to 6 wt%, based on the base binder. A Silverson homogenizer with shear rate of 3000 rpm and mixing time of about 30 min has been used for preparation of the modified bitumen composites. It has been assumed that the viscosity of binder is too low to peel off the gallery spacing of Nano-silica and prevent the agglomeration at such a high temperature. The physical properties of the modified binder is shown in Table 4.

<table>
<thead>
<tr>
<th>Table 1. Analysis of Nano-silica</th>
</tr>
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<tbody>
<tr>
<td>SiO$_2$</td>
</tr>
<tr>
<td>&gt;99%</td>
</tr>
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</table>

<table>
<thead>
<tr>
<th>Table 2. Properties of Silicon Oxide Nanoparticle</th>
</tr>
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<tbody>
<tr>
<td>Particle Size</td>
</tr>
<tr>
<td>20-30 nm</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Table 3. Characteristics of utilized bitumen</th>
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</thead>
<tbody>
<tr>
<td>Ductility at 25 ºC, cm (ASTM D113)</td>
</tr>
<tr>
<td>+ 100</td>
</tr>
</tbody>
</table>
Table 4. Physical characteristics of modified bitumens

<table>
<thead>
<tr>
<th>Test</th>
<th>Base Binder</th>
<th>Nano-silica content with 2% Sasobit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ductility at 25 °C, cm</td>
<td>+100</td>
<td>92  88  83  81</td>
</tr>
<tr>
<td>Softening Point, °C</td>
<td>51</td>
<td>54  55  57  58</td>
</tr>
<tr>
<td>Penetration at 25 °C, 0.1mm</td>
<td>68</td>
<td>62  60  56  54</td>
</tr>
</tbody>
</table>

Table 5. Asphalt mixture mixing model (passing from sieve, %)

<table>
<thead>
<tr>
<th>Sieve Size</th>
<th>(binder and surface coat)</th>
</tr>
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<tbody>
<tr>
<td>50 mm(2&quot;)</td>
<td>-</td>
</tr>
<tr>
<td>37.5 mm(1/2&quot;)</td>
<td>-</td>
</tr>
<tr>
<td>25 mm(1&quot;)</td>
<td>100</td>
</tr>
<tr>
<td>19 mm(3/4&quot;)</td>
<td>90-100</td>
</tr>
<tr>
<td>12.5 mm(1/2&quot;)</td>
<td>-</td>
</tr>
<tr>
<td>9 mm(3/8&quot;)</td>
<td></td>
</tr>
<tr>
<td>4.75 mm(number 4)</td>
<td>44-74</td>
</tr>
</tbody>
</table>

Table 6. The properties of the utilized aggregates

<table>
<thead>
<tr>
<th>Test</th>
<th>Result</th>
<th>Standard limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Los-Angeles (%)</td>
<td>12</td>
<td>25</td>
</tr>
<tr>
<td>Maximum weight loss with sodium sulphate (%)</td>
<td>2</td>
<td>8</td>
</tr>
<tr>
<td>Maximum water absorption (%)</td>
<td>0.8</td>
<td>2.5</td>
</tr>
<tr>
<td>Maximum content of flat and aggregates (%)</td>
<td>1</td>
<td>15</td>
</tr>
</tbody>
</table>

As depicted in Table 5, the standard mixing model of warm mix asphalt has been used for the preparation of the asphalt concretes. The characteristics of the utilized aggregates are presented in Table 6. The aggregate temperature of 135° C is employed to produce the asphalt concretes containing the Nano-silica modified bitumen.

4. Results and Discussion

4.1 Rheological properties of Samples

The rheological properties of conventional and modified bitumen have been studied at 10 and 50 °C by sweeping the frequency between 0.1 and 700 rad/s in a dynamic shear rheometer. The frequency dependence of rheological parameters for the neat and modified bitumen are illustrated in Figures 1-3.

The variation of complex modulus (G*) with frequency at 10 and 50 °C have been shown in Figure 1. As indicated in this figure, the complex modulus of bitumen has increased by adding Nano-silica to the base bitumen. This modulus, indicating the stiffness of samples at heavy traffic conditions, has increased by increasing the Nano-silica content from 2 to 6 wt%. The complex modulus in Figures 1 and 2 indicates a significant increase in complex modulus of modified samples at low frequencies. In these frequencies, the modified Nano-silica modified bitumen shows more resistance against rutting at heavy loading condition which led to significant increase in complex modulus of modified bitumen. Also G* of bitumen is sensitive to temperature. At 50° C the viscosity of bitumen is too low to allow the Nano-silica to disperse well. Therefore the modification of stiffness of Nano-silica modified bitumen is much higher than the base bitumen, relative to its corresponding stiffness at 10° C. The sample with 6 wt% Nano-silica showed more stiffness, especially at low frequencies and high temperature.
Bitumen shows viscous behavior at high service temperatures and elastic behavior at low service temperatures. Complex modulus is affected by elastic and viscous modulus of bitumen. Therefore it is necessary to determine the phase angle, difference between the applied shear stress and the shear strain. Phase angle is more sensitive to the physical and chemical structure of bitumen. By this parameter, viscous or elastic response of samples for an applied stress is specified. An ideal bitumen shows plateau region in complex modulus and phase angle diagram by varying the frequency of rheometer. The effect of Nano-silica on the phase of base bitumen is illustrated in Figures 3 and 4. By decreasing the applied frequency, phase angle of samples have increased. Nano-silica has shown more significant effect on the base bitumen at 50 °C than 10 °C. As seen in Figures 3 and 4, the sample containing 6 wt% Nano-silica has the lowest phase angle at 10 and 50 °C. So, 6 wt% Nano-silica has been selected as the optimum Nano-silica content.
Variation of rut factor (G*/sinδ) versus frequency for the Nano-silica modified samples at 10 and 50°C is observed in Figures 5 and 6. Nano-silica has increased the rut factor of the base binder at low frequencies. At these frequencies which are indicative of severe traffic conditions, the Nano-silica has increased the stiffness and elasticity of the base binder. Therefore superior performance of Nano-silica modified samples was observed at low frequencies and high service temperature.
The variations of loss modulus and storage modulus at 50°C are observed in Figures 7 and 8. The sample with 6% Nano-silica has shown a significant increase in storage modulus which is in agreement with other results.
4.2 Characterization of WMA

4.2.1 Resilient Modulus Test (According to ASTM D4123)
In order to study the resilience efficiency, the asphalt mixtures were stored at the temperature of 25°C for 24 hours. The load of 1000 N was applied on the mixtures in a repeatable loading period with the frequency of 1 Hz. The loading and resting period was 0.1 and 0.9 s, respectively. The test was performed by measuring the ITS of mixtures following ASTM D4123 standard test method.

Resilient modulus is the ratio of applied stress to the recoverable part of strain, after a certain number of loading cycles. This modulus is an estimation of the pavement response to traffic loading. The effect of Nano-silica addition on the resilient modulus of warm mix asphalts have been illustrated in Figure 9. As shown in this figure, Nano-silica has promoting effect on the resilient modulus of asphalt mixtures. It can be observed that the value of resilient modulus has been enhanced by increasing the contents of Nano-silica from 0 to 6%.
4.2.2 Indirect Tensile Test (According to ASTM D6931)

The three steps of the fatigue mechanisms for the WMA prepared with Nano-silica modified bitumen have been illustrated in Figure 10.

1- Firstly, some tiny cracks appeared on the thin layer of bitumen between the sands. This step has been shown at the initial part of the curves in Figure 10. These steep variations have occurred at the transition stage of the thermo mechanical phenomenon, which is due to the viscoelastic subsidence in the asphalt mixtures.

2- Secondly, these changes were completed and the initial cracks were developed which resulted in larger crack pattern.

3- Finally, at the third step, a big crack was rapidly appeared and the asphalt mixture was broken.

The outcome of this test was the measuring of fatigue response of asphalt mixes (100 mm diameter and 150 mm height) subjected to the repeated loading cycles. These mixtures have been prepared by gyratory compactor at 88 gyrations. The asphalt concrete samples were subjected to loading and relaxation for the large number of cycles using an indirect tension apparatus. In each cycle, the mixtures have been subjected to 25-millisecond loading and 125-millisecond resting time. The whole operation has been carried out with the frequency of 0.66 Hz. The test temperature was 25°C and the test was conducted for up to 10000 cycles or until observing permanent deformation of 10-millimeter.

Figure 10 illustrates an exact evaluation of fatigue resistance of the warm asphalt produced by Sasobit and Nano-silica. There is a direct relevance between the Nano-silica content and fatigue life of asphalt mixtures (the number of the loaded cycles that is necessary for cracking), as shown in Figure 10. It has been demonstrated that the control mixture (prepared with unmodified binder) has the highest cracking depth, while sample prepared with 6% Nano-silica has the lowest depth of cracking. Nano-silica shifts the fatigue life of asphalt concrete to the higher cycles and increases the cracking resistance of mixtures at specified loading cycles. The sample containing 6% Nano-silica showed much higher resistance cracking against; therefore, this percentage has been selected as the optimum content. Nano-silica decreases the air voids of asphalt mixtures; consequently, it increases their fatigue life [Sampath, 2010].
4.2.3 Rutting in Wheel Tracking Analysis (According to AASHTO T324)
The wheel tracking device was used to illustrate the rutting and permanent deformation of asphalt mixtures (260 mm×320 mm×40 mm). The rutting depth has been measured in millimeter and the results have been depicted in Figure 11.
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The results illustrated that the rutting depth has decreased by increasing the Nano-silica content in the asphalt mixtures. The asphalt mixtures containing 4 and 6% Nano-silica had 38 and 48% reduction in rutting depth, respectively. As a result, the asphalt mixtures modified with Nano-silica were more resistant against permanent deformation and rutting. This phenomenon is due to significant decrease in temperature susceptibility of Nano-silica modified binders.

5. Conclusions
In this study, the effects of introducing Sasobit and Nano-silica to the asphalt mixture were investigated. The rheological properties of modified bitumen with Nano-silica were also evaluated. The results showed that the modified bitumen has more stiffness at sever traffic condition and high service temperature. Also, rut factor of Nano-silica modified bitumen was less than base bitumen, which indicates superior performance of modified samples at low frequencies and high temperature. Rheological investigations indicated that 6% Nano-silica could significantly improve the rutting performance of base binder; as such, this concentration was selected as the optimum amount. The results of investigations on the control WMA and Nano-silica-modified WMA demonstrated that the Sasobit and Nano-silica could improve the tensile strength as well as the rutting resistance of WMA. Asphalt concretes with Nano-silica modified binders showed less temperature susceptibility and less rutting and permanent deformation at a specified loading condition. The WMA prepared with Nano-silica modified bitumen had less fatigue cracks. Finally, 6 wt% Nano-silica was selected as the optimum content for the modification of the bitumen.

6. References


