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# Improving aging resistance and fatigue performance of asphalt binders using inorganic nanoparticles

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## HIGHLIGHTS

- Nano $TiO_2$ and nano $CaCO_3$ particles improved aging resistance of asphalt binder.
- Nano $SiO_2$ modified binders outperformed other binders in terms of fatigue behavior.
- Linear amplitude sweep test results were consistent with the time sweep test.
- Larger specific surface area of nano $SiO_2$ contributed to modification of the binder.

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## ABSTRACT

The use of inorganic nanoparticles in asphalt binders and mixtures to improve performance related properties have gained a popularity in recent years. Extremely large interface area with asphalt binder matrix is the most important feature of these nanoparticles which translates in a higher degree of changes in the microstructure of asphalt binder. In this research, different inorganic nanoparticles of $SiO_2$, $TiO_2$ and $CaCO_3$ were mixed with asphalt binder and the chemical and microstructural properties of the nanocomposites were evaluated using X-ray diffraction and scanning electron microscopy, respectively. The effect of oxidative aging has been investigated using Fourier Transform Infrared spectroscopy as well as the determination of viscoelastic properties of the modified binders. Furthermore, strain-controlled Time Sweep and Linear Amplitude Sweep tests were used to evaluate the performance of nanocomposites against fatigue cracking. The results indicated improved aging resistance by addition of $TiO_2$ and $CaCO_3$ nanoparticles to asphalt binder. Moreover, nano silica enhanced fatigue properties in the whole range of strain levels. However, different variables such as additive content and strain level affected the fatigue behavior of $TiO_2$ and $CaCO_3$ modified binders.



## 1. Introduction

One of the most common distresses in asphalt pavements is fatigue cracking. Fatigue cracking starts with the creation of micro cracks in the asphalt mixture, and after more repetitions of loading these micro cracks nucleate, and finally, this trend leads to the structural rupture of the asphalt mixture. Fatigue cracking generally happens in later stages of the pavement life under repeated traffic loading in intermediate temperatures and is intensified by excessive brittleness and lack of flexibility [1–3]. Fatigue in the asphalt mixture is a function of chemical and rheological properties of the asphalt binder, aging phenomena, type, characteristics and gradation of aggregates, volumetric properties of asphalt mixtures, traffic loads as well as the environmental condition [4–7]. Cracking typically occurs either within the asphalt binder or at the asphalt binder–aggregate interface [8]. Given the above-mentioned points, the role of an asphalt binder in fatigue resistance of asphalt mixtures is undeniable.

Asphalt binder is a substance whose behavior generally depends on the time of loading, temperature, and the level of strain or stress. In shorter loading times or higher frequencies, the asphalt binder behaves more elastically similar to its behavior at lower temperatures. In such conditions, there is not enough time to reconstruct the molecular structure [9,10]. In higher temperatures or extended loading times (or low rates of loading), asphalt binder behaves more viscous; in other words, in higher temperatures, the molecular motion increases, and this gives rise to the viscous

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behavior of asphalt binder [9,10]. The behavior of asphalt binder in intermediate temperatures in which fatigue failure is more probable is a combination of viscous and elastic behavior. According to the current method of Performance Grading (PG) of asphalt binder [11], it is suggested that the modulus of asphalt binder in intermediate temperature has to be reduced, and more elastic behavior is favored in order to improve the resistance of the asphalt binder and asphalt mixture against fatigue cracking. Modifiers are usually used to improve the performance of asphalt binder against fatigue cracking.

The modification of asphalt binder with polymeric materials and rubber powder is among the most common methods to improve the performance of the asphalt binder against most types of distresses. Another method for the improvement of asphalt binder characteristics that has recently gained attention is the modification of asphalt binder with nanoparticles. Nanomaterials have morphological features that possess special characteristics stemming from their nanoscale dimensions [12]. Due to the very tiny size and huge surface area, the properties of nanomaterials are much different from the normal-sized materials [13]. The addition of these nano-sized particles to another material may overcome the limitations of monolithic, and asphalt binder is no exception. Adding nanomaterials changes the rheological properties of the asphalt binder and might also lead to changes in the intermolecular forces within the asphalt binder structures [14]. This is due to the fact that as dimensions reach the nanometer level, interactions at phase interfaces become largely increased. There are several nano-materials that have the possibility to be utilized for modification of asphalt binders, such as nano-clay, nano-silica, nano-hydrated lime, nano-sized plastic powders, polymerized powders, nanofibers, and carbon nanotubes [15–18]. Different levels and various combinations of such nanoparticles in the asphalt binder are used in order to enhance the resistance of the asphalt binder and asphalt mixtures against permanent deformation [18–23], low-temperature cracking [24–27], general improvement of asphalt binder's rheological characteristics [13,24–26,28,29] as well as aging resistance [30,31].

Although studies that have investigated the effect of nanoparticles on high-temperature characteristics of asphalt binders, storage stability of polymer modified binders, moisture susceptibility of the asphalt mixtures, and aging resistance of asphalt binders are very extensive, less attention has been paid to the evaluation of their effect on the fatigue performance of asphalt binder and mixture. This may be due to the fact that there is more complexity associated with the fatigue phenomenon as well as due to the imperfectness of the common methods of evaluating the fatigue behavior of asphalt binder.

Different mechanisms have been provided that show the high potential of nanoparticles in improving fatigue properties of the asphalt binder and mixture. Nano-modification impacts the dynamics of wetting and diffusion stages of the healing process, which results in restoration of mechanical properties [32]. Moreover, interfacial regions between matrix (asphalt binder) and nanoparticles that can break during a fracture may also exhibit weak intermolecular bond reformation, thus actively participating in the wetting phase of the healing phenomenon [32,33].

It has been suggested that the current method of Superpave performance grading for asphalt binders lacks the capability of proper assessment of fatigue properties [10,34]. Therefore, new testing methods, such as time sweep or linear amplitude sweep tests, have been implemented for evaluating the fatigue performance of asphalt binders [10]. For instance, Kavoosi and Barghbani (2016) in a research aimed at examining the effect of nano-clay, hydrated lime, and nano-hydrated lime, on the fatigue performance of asphalt binder, used the linear amplitude sweep test for short-term aged asphalt binder samples at 22 °C [35]. Results showed that among these materials, modification of asphalt binder with nano-clay leads to an extended fatigue life [35]. Also, nano-hydrated lime showed the less negative effect on fatigue behavior of asphalt binder compared to hydrated lime [35].

Santagata et al. in a study to investigate the fatigue properties of bituminous binders reinforced with carbon nanotubes (CNTs), used repeated oscillatory loading in the controlled-stress mode with a dynamic shear rheometer. Their results showed that CNT can significantly improve fatigue properties of asphalt binders if it is adequately dispersed in the asphalt binder [36]. In another study, the same authors showed that CNT prolongs the lifespan of the neat bituminous mastic by means of several interdependent mechanisms that can affect either the fatigue cracking resistance or the self-healing capability [32]. Azarhoosh et al. used the surface free energy concept and showed that the addition of nano $TiO_2$ to asphalt binder increases the total surface energy of the asphalt binder, which results in an increased adhesion to aggregate and prolongs the fatigue life of the asphalt mixture [37]. The same results were observed for nano-zinc oxide particles [38].

## 2. Research objectives

Due to the size and high specific area, nanoparticles show high-performance potential and their application is extending in order to improve the performance characteristics of the asphalt binder. One of the points that has rarely been the subject of previous researches is the performance of the asphalt binders modified with metal oxide nanoparticles against the accumulation of micro cracks and fatigue, and its relation to oxidative aging.

Another noteworthy point is the inappropriateness of the current method of performance grading at an intermediate temperature range or the evaluation of resistance against fatigue for asphalt binders. In this study, the fatigue performance of neat and modified asphalt binders with 2% and 4% (by wt) of nanoparticles of silicon dioxide ($SiO_2$), titanium dioxide ($TiO_2$), and calcium carbonate ($CaCO_3$) are evaluated. The characteristics of the resulting nano composites are determined using X-ray Diffraction (XRD) and Scanning Electron Microscope (SEM). The viscoelastic characteristics and fatigue performance of neat asphalt binder and nanoparticle modified samples are examined using the frequency sweep test, linear amplitude sweep test, and time sweep test. Also, in order to understand the modification mechanism of asphalt binder and evaluate the performance of these materials on aging that is one of the effective phenomena on fatigue failure, functional groups associated with oxidizing, such as carbonyl and sulfoxide, are analyzed through Fourier Transform Infrared Spectroscopy (FTIR).

## 3. Materials and sample preparation

### 3.1. Asphalt binder and nanoparticles

In this research, a 60/70 penetration graded bitumen was used as the base asphalt binder (neat asphalt binder) and its physical properties are presented in Table 1. Nano silicon dioxide ($SiO_2$),

**Table 1**
Physical properties of neat asphalt binder.

| TEST | Standard | Unit | Result |
|---|---|---|---|
| Penetration | ASTM D5 | 0.1 mm | 70 |
| Softening Point | ASTM D36 | °C | 50.4 |
| PI | – | – | −0.268 |
| Viscosity@135 °C | ASTM D4402 | cP | 315 |
| Ductility | ASTM D113 | cm | 126 |
| Flash Point | ASTM D92 | °C | 275 |