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Characterisation of Acrylonitrile Styrene Acrylate Modified Asphalt Cement with Nano Iron Oxide and Nano Silica Particles

Kurthan Korutürk & Mustafa Alas*

Faculty of Civil and Environmental Engineering, Civil Engineering Department, Near East University, Near East Boulevard, Nicosia, 99138 North Cyprus, Turkey

**Corresponding author: mustafa.alas@neu.edu.tr*

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ABSTRACT

The current study investigated the physical properties and rheological characteristics of asphalt cement (AC) modified by nano ironoxide (Fe3O4) and nano silica (SiO2) particles. Seven different blends including base AC, Acrylete-Styrene-Acrylonitrile (ASA)/ Nanosilica (Si) and ASA/ Nanoironoxide (Fe) were the subject of experimental investigations. ASA was used at 5% for all modified blends while the nanomaterials were blended in 3, 5 and 7% concentrations by the weight of AC. Temperature susceptibility, rotational viscosity (RV) and storage stability for the base and polymer nanocomposite modified asphalt cement (PNCAC) were evaluated by the physical testing procedures while the frequency sweep test and the multiple stress creep recovery (MSCR) tests were conducted using a dynamic shear rheometer. The results showed that, the viscoelastic properties of the ASA/Si and ASA/Fe modified binders were improved when subjected to a range of temperatures and loading conditions. ASA/Fe composite modified AC demonstrated superior high temperature performance characteristics while ASA/Si composite modified AC although presented less significant improvement at the high temperature conditions, it was also able to improve the intermediate temperature fatigue resistance of asphalt more than the ASA/Fe composite modified AC. The optimum concentration of additives were found to be 3 and 5% for the ASA/Fe and ASA/Si modified AC respectively. Further addition of nanomaterials beyond the abovementioned concentrations resulted in degradation of the enhancement in physical and rheological properties of PNCAC, which was associated to the occurrence of agglomeration of nanoparticles and phase separation between the polymer-nanomaterial-asphalt matrix.

Keywords: Polymer/nanocomposite; nano ironoxide; nano silica; dynamic shear rheometer; multiple stress creep recovery

INTRODUCTION

Asphalt cement (AC), although used in small quantities in the design of hot mix asphalt (HMA), it has a significant influence on the performance of flexible road pavement structures (Piuzzi et al. 2021). The viscoelastic nature of AC makes it a suitable option for flexible pavement design as it possesses valuable engineering properties to resist dynamic vehicular loading and extreme climatic conditions, which the pavement roads encounter during the service life. However, from the previous reports of field observations and the research conducted in the literature, it has been acknowledged that, the neat asphalt binder used for paving applications may not always ensure sufficient stability, durability and desired performance characteristics (Wang et al. 2021). In the search for enhancing the performance of asphalt cement and mixtures, numerous investigations have been concentrated on asphalt modification with polymers, nanomaterials and polymer nanocomposites due to their versatile properties needed to design superior performing asphalt pavements (Günay & Ahmedzade 2020; Li et al. 2017; Porto et al. 2019).

Polymers have proven to be effective modifiers in enhancing the physical and rheological properties as well as improving the durability of AC, thus reducing the life cycle costs (Porto et al. 2019). Polymer modified asphalt is considered to be cost-effective in extending the service life of the pavements by up to 2-3 years, given that the cost of modification does not surpass the cost of base asphalt by more than 100% (Behnood & Gharehveran 2019). Based on their characteristics, polymers are mainly categorized as plastomeric and elastomeric materials. A popular type of polymer used to modify asphalt is a copolymer of polystyrene and polybutadiene. It is usually called Styrene-Butadiene (SB) or Styrene-Butadiene-Styrene (SBS). Since it adds viscosity and elasticity to the asphalt, this copolymer family is called an elastomer. SB and SBS constitute to a percentage. of more than 75% of the polymers used in asphalt modification (Masad et al. 2020). Another type of polymer type that is used in the asphalt modification is the plastomeric polymers. They are commonly formed from copolymers of polyethylenes and polyesters. Since it adds stiffness and plasticity to the asphalt, this family of copolymers is called plastomers (Airey et al. 2016).

According to (Ameri et al. 2013), elastomeric polymers are capable of improving both high and low service temperature performance characteristics of AC whilst, plastomers are known to be efficient modifiers at improving the high temperature performance for AC. Although the positive effects of polymer modifiers on the viscoelasticity of AC, a number of researchers have reported a major shortcoming for the polymer modified AC, which is the compatibility issue between the polymer and AC (Galooyak et al., 2010). (Al-Mansob et al. 2017) stated that, the incompatibility is correlated with the occurrence of phase separation due to the variations in density, molecular weight and solubility between the polymer and asphalt. Due to the shortcomings of the polymer modified AC, researchers were gravitated towards the investigation of the potential of nanomaterials and nanocomposite materials as modifiers to asphalt cement.

The application of asphalt modification with materials in nanoscale has been incorporated in pavement engineering in the last decade. A number of special properties of nanomaterials such as increased surface area to volume ratio and the quantum effects resulting from the spatial confinement added certain beneficial features to the modified asphalt cement such as; enabling better dispersion and stability in the mix, improving the aging resistance in the short and long term and enhancing the viscoelastic properties of the asphalt cement at extremely high and low temperatures (Bhargava et al. 2016). For nanomaterial modified AC, the enhancement or deterioration of asphalt properties such as ageing resistance, workability, stability and rheological properties depends on the molecular structure and density of the nanomaterials. In addition, particle size, specific surface area and purity of the nanomaterials are the influential factors that determine the new structural formations and changes in the performance characteristics of asphalt cement. Carbon nanotubes, nano silica, nano clay and nano iron are among the common nanomaterials that were previously used in the modification of asphalt cement. As utilised in the current study, nanosilica is considered as one of the most substantial advancements in asphalt modification (Bhargava et al. 2016; Yang & Tighe 2013). A study conducted by (Yao et al. 2013) discovered that 2% to 4% use of nanosilica by the weight of AC can reduce the rut depth up to 50%. Additionally, (Arabani & Faramarzi 2015) achieved remarkable enhancement in fatigue resistance of up to 37% than the neat asphalt. On the other hand, nano ironoxide particles were found to postively affect the performance characteristics of AC. A study conducted by (Kordi & Shafabakhsh 2017) with stone mastic asphalt modified by Fe2O3 nanoparticles demonstrated that, a small amount (0.9% by the weight of AC) of Fe2O3 addition to asphalt cement increased the fatigue life of samples by 15%-35% compared to the neat asphalt. Additionally, their research reported that, the amount of rut depth of asphalt samples containing 0.9% Nano Fe2O3 was about 25–40% lower than the neat asphalt samples. Further, Pirmohammad et al. (2020) investigated the fracture properties of asphalt mixtures modified with Nano Fe2O3 and CNT's. The outcomes of their study showed that, 0.8% and 1.2% of modifier concentrations for Nano Fe2O3 and CNT's respectively resulted in remarkable improvement of fracture properties under two different types of mix loading modes.

From the literature, it was acknowledged that polymer based and nanomaterial modified AC were commonly adopted to improve the viscoelastic behaviour of neat asphalt. However, it was observed that polymer and nanomaterial additives alone do not always meet expectations, both in terms of cost and the desired strength due to major shortcomings such as the phase separation and agglomeration issues. For this reason, the search for different additives has been started and more recently, polymer/nanocomposite modified AC (PNC) is considered to be the latest advancement in the field of pavement engineering that facilitates the design of roads with superior performance and increased durability (Porto et al. 2019). A peculiar feature of polymer nanocomposites is the cost effectiveness compared to polymer modified and nanomaterial modified AC due to lowered modifier contents added in the blend (Bala et al. 2019). It is also acknowledged that a number of nanoparticles including nano silica and nano iron oxide can have positive influence in order to eliminate major drawbacks of polymer and nanomaterial modified AC thus, resulting in longer lasting and better performing pavement roads. Studies showed that, the drawbacks of well-known polymers such as SBS, PP and PE were found to be satisfactorily recovered with the addition of nanosilica (Bala et al. 2017; Bala et al. 2018; Rezaei et al. 2016). Although, there have been no published findings found in the literature regarding the use of nano iron oxide as secondary modifier to a polymer modified AC, it was acknowledged that sole modification of AC by pure nano ironoxide particles were reported to be infective to meet the strict requirements for use in regions where extreme high and low temperatures are anticipated (Zhang et al. 2015).

Considering the gap in the literature regarding the use of nano silica and nano iron oxide as secondary additives in the polymer modified AC, the current study aims to investigate the physical properties and viscoelastic characteristics of ASA/Si and ASA/Fe modified AC by using conventional and dynamic shear rheometer testing procedures.

EXPERIMENTAL DESIGN

MATERIALS AND SAMPLE PREPARATION

The neat asphalt that was used as the base (also referred to as the control sample) was 80/100 penetration grade asphalt. The modification of neat asphalt was performed first by adding an elastomeric polymer named Acrylate-Styrene-Acrylonitrile (ASA) in the first 20 minutes of blending process. ASA was used at 5% concentration by the weight of base asphalt. This content was found optimum in a number of studies conducted by (Ali et al., 2015) and (Mubaraki, 2019). In the first set of samples, the nano iron oxide (Fe3O4) particles and in the second set of samples nano silica (SiO2) particles were gradually added in the blend for further 40 minutes at 3%, 5% and 7% concentrations by the weight of asphalt/polymer composite matrix. The properties of the additives were listed in table 1. As a result of the blending processes three different set of experimental samples and seven different asphalt specimens were produced including the control specimen which was not blended but only stirred after being heated to a desired temperature of 160oC and placed in moulds for preparing to testing. The total duration of the blending process for the modified asphalt samples was 60 minutes and it was performed by a high shear mixer at 5000 rpm and 160oC. To ensure the fine dispersion of ASA and nanoparticles in the blend, the softening point test results were examined every 20 minutes during the blending process. Once the softening point results were stabilised, the additives were considered to be finely dispersed and the above mentioned shear rate, duration and temperature for the blending process were determined.

TABLE 1. Physical properties of the base binder and the modifiers

Properties	Nano-Silica	Nano-Ironoxide
Formula	SiO_2	Fe_3O_4
Molecular Weight	6.3-6.49	N/A
Color and Odor	Black Powder/ odourless	White
Form	Nano powder	Powder
Purity	0.9999	0.99
Average nanoparticle size (nm)	70- 100	30-40
Bulk Density (g/cm^3)	N/A	0.68
Melting Point (°C)	1326	825

CONSISTENCY AND TEMPERATURE SUSCEPTIBILITY

The penetration, softening point and rotational viscosity (RV) tests were conducted according to the American Society for testing and materials (ASTM) D 5 – 06, ASTM D 36 – 95 and ASTM D 4402 – 02 respectively. Each test was performed with at least three repetitions and the mean of the test results were recorded in order to improve reliability. Penetration and softening point tests were used to evaluating the consistency of the base and modified asphalt samples against temperature variations while the RV tests results were used as a measure of the amount of hardening after the modification process. Also, the combination of all the above mentioned test results aided in the computation for the Penetration Index (PI) and the Penetration Viscosity Number (PVN) which were used in assessing the temperature susceptibility of the base and polymer/nanocomposite modified asphalt cement (PNCAC).

STORAGE STABILITY

The storage stability test was utilized to assessing the integrity and homogeneity of the polymer nanocomposite modified asphalt (PNCAC) during storage at high temperatures in the production facility and while transporting the materials to the on-site.

The testing procedure was followed by emptying the PNCAC samples into aluminium foil tubes that had a diameter of 3cm and a height of 16cm. The top of the aluminium tubes were sealed and the samples were rested in an oven at ±163^{o}C for 48 hours in vertical position. After removing the tubes from the oven, the tubes were cooled down at room temperature and they were split into three equal sections which the upper and lower third sections were taken for the softening point investigation. The test results were evaluated based on ASTM-D 5892.

DYAMIC SHEAR RHEOMETER (DSR)

Frequency Sweep Test

The frequency sweep tests were performed at temperatures ranging from 10^{o}C to 82^{o}C with 6^{o}C increments and a range of frequencies between 0.159 Hz and 15.92 Hz, where the former represented the anticipated field temperatures and the latter simulated the speed of traffic up to 90 km/hr (Ramadan & Abo-Qudais, 2017). The test was run in two phases. The high temperature characteristics for the asphalt binders were measured between 46^{o}C - 82^{o}C with a 25mm spindle diameter and 1mm sample thickness. In this range of temperatures, the tests were conducted on fresh and short-term aged samples. To evaluating the viscoelastic properties of AC at intermediate temperatures between 4^{o}C - 46^{o}C, an 8mm diameter plate geometry and 2mm thickness of samples were tested under long-term aged conditions.

The short-term aging and long-term aging conditions were simulated by the rolling thin film oven (RTFO) and pressure aging vessel (PAV) respectively. Additionally, to control and maintain a constant temperature during the testing procedure, DSR was equipped with a temperature control unit and fluid bath system which were used in order to maintain a stable temperature within ±0.1^{o}C as suggested in the SuperPave specification.

MULTIPLE STRESS CREEP RECOVERY

A MSCR test was conducted to simulate a continuous traffic load in order to determine the amount of permanent deformation that occurs during the repeated dynamic loading. DSR was used with a 25mm plate geometry and 1mm sample thickness on RTFO aged samples. The standard protocols followed in the testing were as described in AASHTO TP70. The MSCR test was performed at 64^{o}C with 100 Pa and 3200 Pa stress levels which represented low and heavy traffic conditions respectively. The testing procedure was started by applying a one second load in haversine form at 100 Pa which was followed by a resting period of nine seconds. The 10 second interval was considered as one cycle. After completing 10 consecutive cycles at 100 Pa, the test was further continued for another 10 cycles under 3200 Pa loading to complete a total of 20 cycles.

RESULTS AND DISCUSSION

PHYSICAL PROPERTIES AND TEMPERATURE SUSCEPTIBILITY

Conventional properties of the base AC and the PNCAC were evaluated based on the experimental outcomes from the physical testing procedures including the penetration, rotational viscosity and the softening point tests. The penetration and the softening point test results as presented in Figure 1 illustrated that introducing nanomaterials into the asphalt matrix resulted in hardening of the asphalt binder which was deducted from the significant decrease and considerable increase in the softening point values.

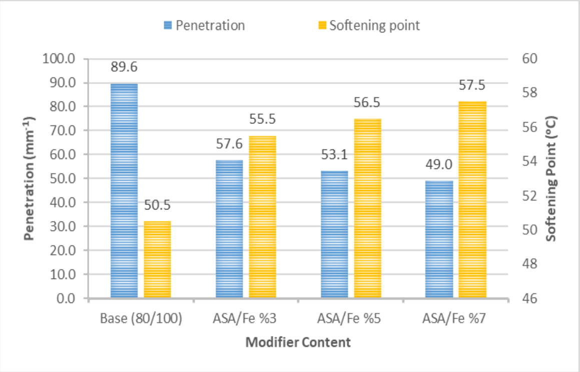


FIGURE 1. Penetration and Softening Point for Base and PNC modified AC

The temperature susceptibility of the AC was further evaluated by computing two parameters, namely the PI and PVN indices. RV, Penetration, and the softening point parameters were used to compute PI and PVN indices by eq.1 and eq.2 - eq.4 respectively. The PI and PVN indices for asphalt binders range from -3 to +7, where lower values indicate a more temperature susceptible AC and higher values indicate the opposite. The analytical outcomes for the PI and PVN were demonstrated in Table 2.

$$\mathrm{PI} = \frac{1952-500\log(\mathrm{Pen25})-20\mathrm{S.P}}{50\log(\mathrm{Pen25})-\mathrm{S.P}-120} \tag{1}$$

$$\mathrm{PVN} = \frac{\mathrm{LogL}-\mathrm{LogX}}{\mathrm{LogL}-\mathrm{LogM}} \times 1.5 \tag{2}$$

Where L is the rotational viscosity value at 135°C for a PVN of 0.00,

$$\mathrm{LogL}=4.25800\text{-}0.79670\mathrm{LogPen} \tag{3}$$

M is the rotational viscosity value at 135°C for a PVN of -1.50,

$$\text{"M=3.46289-0.61094LogPen"} \tag{4}$$

X is the rotational viscosity value at 135°C and is the logarithmic penetration value for AC at 25°C.

TABLE 2. Temperature Susceptibility

	Penetration	Softening point	RV	PI	PVN
Base AC (80/100)	89.6	50.5	474	0.43	1.819
ASA/Si %3	63.3	54.5	648	0.45	1.852
ASA/Si %5	61.1	55.0	888	0.47	1.950
ASA/Si %7	53.4	57.0	1050	0.57	1.972
ASA/Fe %3	57.6	55.5	640	0.43	1.825
ASA/Fe %5	53.1	56.5	732	0.44	1.850
ASA/Fe %7	49.0	57.5	756.	0.46	1.848

ROTATIONAL VISCOSITY

The RV measurements at 135°C and 160°C are used to evaluate the workability of AC during the production and construction stages. Higher viscosities yield to a less workable HMA due to increased stiffness which results in higher energy consumption in the production plant because of increased mixing temperatures and also it requires more effort to compact the asphalt mix in the construction field. The rotational viscosity testing procedures were applied for the base and PNC modified AC in a range of temperatures from 120°C to 180°C by 15°C increments in order to observe the variation of viscosity at elevated temperatures and the results were illustrated in Figure 2.

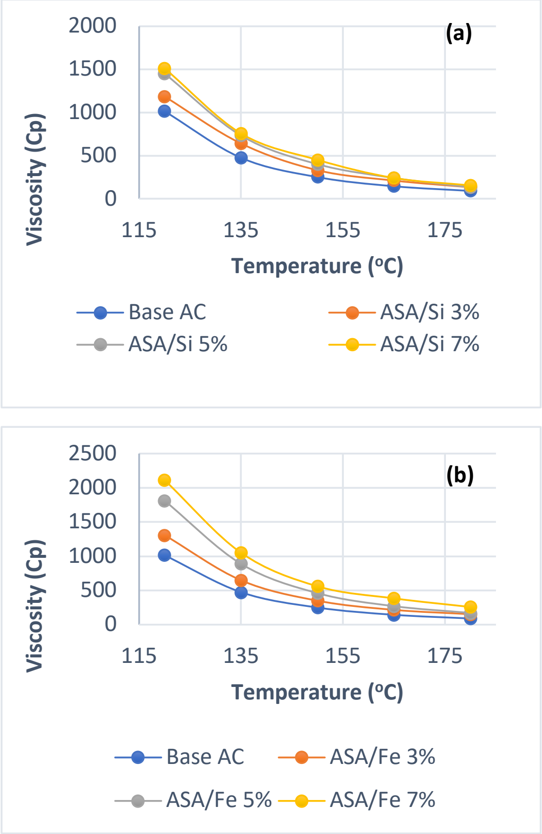
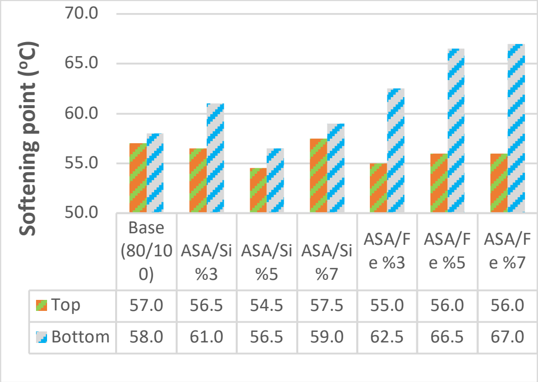


FIGURE 2. RV for Base and a) SiO_2 b) Fe_3O_4 modified AC

It can be observed from Figures 2.a and 2.b that, the viscosity for all samples were reduced as the test temperature increased regardless of the modifier content. This was due to the viscoelastic nature of asphalt. Furthermore, the increase in the modifier content resulted in higher viscosities because of stiffening of the asphalt due to the modification process. The samples that contained the highest percentage of modifier concentrations possessed the highest viscosities at 120oC which were 1618 Poises for the ASA/Si and 2220 Poises for the ASA/Fe composites at 7% modifier content by the weight of asphalt. The viscosities for both of the polymer nanocomposites were reduced significantly compared to the findings of a research conducted by (Ali et al. 2015) which utilised ASA as sole modifier to base AC of 60/70 grade. However, it is difficult to distinguish whether the difference in viscosities was a result of the influence of the additives or the effect of the penetration grade of AC since, different penetration grades were utilized in the two studies. Nevertheless, it can be noted that, with the application of polymer nanocomposites in the current research, the workability of the AC was improved and the mixing and compaction temperatures were reduced owing to achieving lower viscosities.

STORAGE STABILITY

Storage stability at high temperatures is an important property for a modified AC because it is related to the integrity and homogeneity of asphalt mixtures. Although polymer modified AC offers promising enhancement in the physical and rheological properties for asphalt, a common issue with the polymer modified asphalt is the phase separation problem due to dissimilarity between the polymer and asphalt chemical structure such as the solubility and density (Al-Mansob et al. 2017). The phase separation is associated with the accumulation of polymer particles in the top section of the AC at high temperatures, which forms a steady position resulting in a phase separated structure in the asphalt matrix. On this basis the nanoparticles were added in the polymer modified AC and the storage stability test was conducted to evaluate the effect of additives on the storage stability of the resulted PNCAC. Although, other test methods for evaluating the storage stability of the AC such as the RV and DSR techniques were available, the softening point test results, as utilized in numerous high-class investigations were utilized in the present study due to its simplicity. The results of the storage stability tests were illustrated in Figure 3.



	Base (80/100)	ASA/Si %3	ASA/Si %5	ASA/Si %7	ASA/Fe %3	ASA/Fe %5	ASA/Fe %7
▨ Top	57.0	56.5	54.5	57.5	55.0	56.0	56.0
▨ Bottom	58.0	61.0	56.5	59.0	62.5	66.5	67.0

FIGURE 3. Storage Stability test results

The disparity between the softening points of the upper and lower sections of the asphalt samples extracted from the aluminium foil tubes was measured to analyse the storage stability. According to the literature, although a difference of up to 4ºC-4.5ºC, between the top to bottom parts of the conditioned samples was considered within acceptable limits, the common perception among the researchers is that in order to classify AC as storage stable, the difference in softening points should not exceed 2.5 (Alhamali et al. 2015).

From Figure 3 it was observed that, the storage stabilities for the ASA/Si samples were improved as the modifier to asphalt concentration was increased. It was observed that, the softening point of the samples extracted from the top part of the conditioned sample were not higher than 2.5^{o}C for 5% and 7% ASA/Si modified AC, while at 3% ASA/Si concentration, the difference in softening points was 4.5^{o}C. On the contrary, ASA/Fe modified AC samples demonstrated poor storage stability. The top section of the samples were significantly higher than the bottom sections. The differences were 7.5^{o}C, 10.5^{o}C and 11^{o}C for ASA/Fe 3, 5 and 7% compositions respectively.

RHEOLOGICAL CHARACTERISTICS

Master Curves

A master curve is a representation of stiffness (G*) in a range of temperatures and frequencies which is displayed in a single graph. The frequency sweep test was utilized to run the experiments and the experimental outcomes were used to construct a master curve by utilizing time-temperature superposition theorem which was performed by shifting the G* data points at different temperatures on the vertical axis corresponding to the matching frequencies on the horizontal axis. Herein, a reference temperature of 64^{o}C was selected and the data points at the other temperatures were shifted horizontally to and fro by using the convenient shift factors. The results were illustrated in Figure 4 and Figure 5 for ASA/Si and ASA/Fe modified asphalt cement samples respectively.

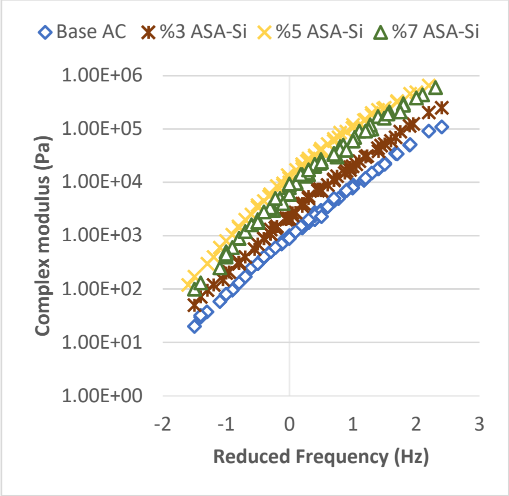


FIGURE 4. Master curves for Base and ASA/Si modified AC

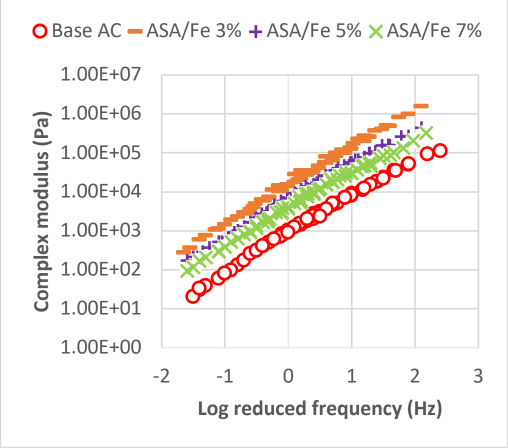
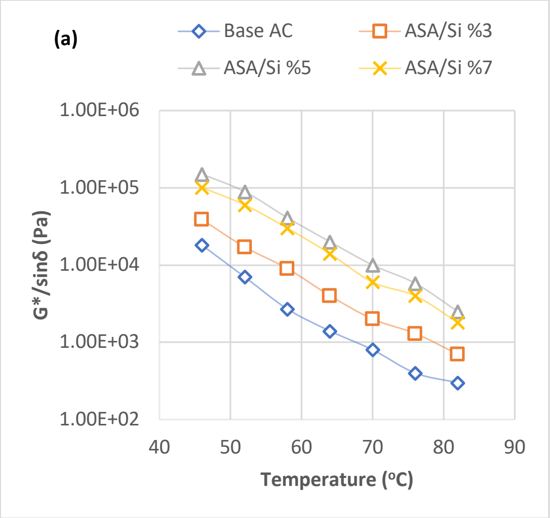


FIGURE 5. Master curves for Base and ASA/Fe modified AC

From Figures 4 and Figure 5, it was observed that, the stiffness of all PNC modified AC's were higher than the base asphalt under unaged conditions, indicating that the modification process has led to enhanced complex modulus. The increase in G* was more remarkable for the ASA/Fe composites than the ASA/Si composites. This showed that, the ASA/Fe composites were more suitable for paving applications in regions where the high temperature conditions are anticipated. It was also noted that, the influence of the additives when used in different concentrations affected the rheological behaviour of the PNCAC. The maximum enhancement in G* was obtained at 5% ASA/Si and 3%ASA/Fe compositions and therefore they were considered to be the optimum compositions for improving the high temperature performance characteristics of AC, as further addition of PNC resulted in lower enhancement in the rheological properties. The reduction in enhancement of rheological properties was attributed to the occurrence of agglomeration of the nanoparticles in the case of ASA/Si composites while formation of the phase separated structures was one of the possible causes for the ASA/Fe composites. From the literature review, it was acknowledged that the nanoparticles tend to agglomerate and form clusters within the composite asphalt matrix which may have led to a reduction in the performance enhancement at usage in higher concentrations. On the other hand, the phase separation phenomenon for the ASA/Fe composite modified AC was a possible cause for reduced G* which was also evidential from the storage stability test results (Yin et al. 2019).

RUTTING AND FATIGUE RESISTANCE PARAMETERS

Rutting resistance is defined as the ratio of the viscous and the elastic portions of an asphalt binder and it is denoted with the formula G*/sin δ. According to Superpave standards, at a loading rate of 1.592 Hz, a minimum of 1 kPa is the allowable requirement for an unaged sample of binder. The rutting parameter was evaluated in a range of temperatures from 46oC- 82oC at a loading rate of 1.592 Hz by using the G* and δ outcomes from the frequency sweep test results. As illustrated in Figure 6a, G*/sinδ was the lowest for base AC. Binders containing ASA/Si composites up to 5% by the weight of AC demonstrated the highest G*/sinδ value, while the addition of ASA/Si composites above 5% concentration led to reduced G*/sinδ. The compatibility problem between the polymer nanocomposite and the asphalt was considered to be the factor leading to the reduction in rutting resistance parameter at 7% ASA/Si concentration. From Figure 6b, it was observed that, among all of the compositions for the PNC modified AC including the ASA/Si composites, the ASA/Fe composite modified AC at 3% modifier content resulted in the highest G*/sinδ. For ASA/Fe composites at 5% and 7% modifier content, although the G*/sinδ was higher compared to the base AC, they were considerably lower compared to the ASA/Fe 7% and also ASA/Si composites at 5 and 7% modifier contents. The different behaviour was associated to the phase separation between the polymer and the nanoparticles. In general, it can be concluded that, the rutting performance of all the PNC modified AC were enhanced significantly compared to the base binder and satisfied the minimum requirement of 1 kPa at 1.592 Hz and at 64^{o}C.



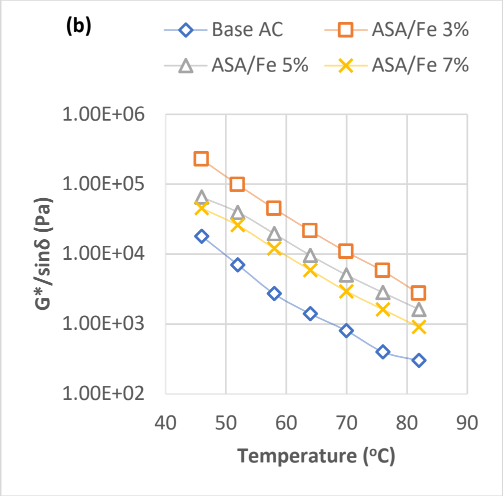


FIGURE 6. Rutting Resistance Parameter for a) ASA/Si b) ASA/Fe Modified AC

The effect of additives on the intermediate temperature fatigue cracking behaviour of AC was analysed by the fatigue resistance parameter which was denoted by the formula G*.sinδ. The minimised G*.sinδ parameter was an indication of better fatigue cracking resistance for the AC. The Superpave method considers 5000 kPa as the maximum limit of fatigue cracking at low and intermediate temperatures. From Figure 7a and Figure 7b it can be observed that, the Superpave maximum criteria were met for ASA/Si composites at a temperature of 18^{o}C while, the ASA/Fe composite modified AC samples were shown to be resistant to fatigue cracking at temperatures around 26^{o}C. Additionally, as illustrated in Figure 7b, the ASA/Fe composites were shown to demonstrate weaker fatigue resistance than the base AC which was associated with the stiffening due to the modification process. On the other hand, ASA/Si composite modified AC at 3% and 5% compositions were found to have lower G*.sinδ compared to the base AC and at 7% composition the results were slightly similar to the G*.sinδ at temperatures up to 15^{o}C. Beyond this trend, G*.sinδ for PNC modified AC were increase continuously. This outcome demonstrated that the ASA/Si composite modified binders were able to improve both the high and low temperature performance characteristics of AC while ASA/Fe composites were only efficient in enhancing the high temperature performance characteristics AC.

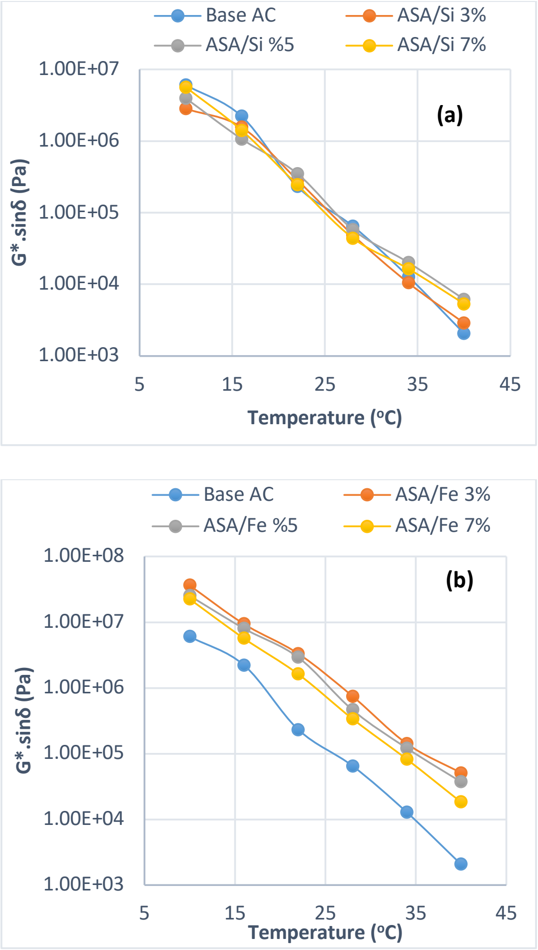


FIGURE 7. Fatigue Resistance Parameter for a)ASA/Si b)ASA/Fe Modified AC

MULTIPLE STRESS CREEP RECOVERY

The MSCR test was utilised to simulate the movement of traffic flow on the highway surface and to evaluate the resistance and recovery of asphalt binder against rutting. The test was conducted in two phases which were the creep phase for one second and the recovery phase for the following nine seconds. The test was conducted at two stress levels; 100Pa and 3200Pa for ten cycles at each stress levels and the accumulated strains for ASA/Si and ASA/Fe modified asphalt samples were recorded and illustrated in Figure 8a and Figure 8b respectively.

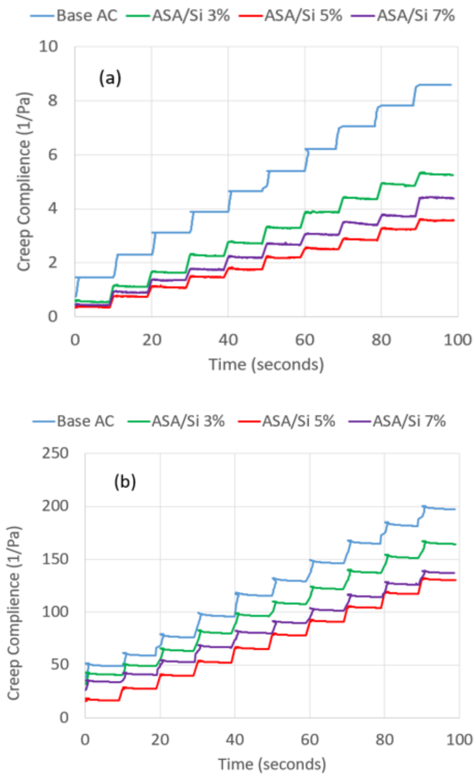
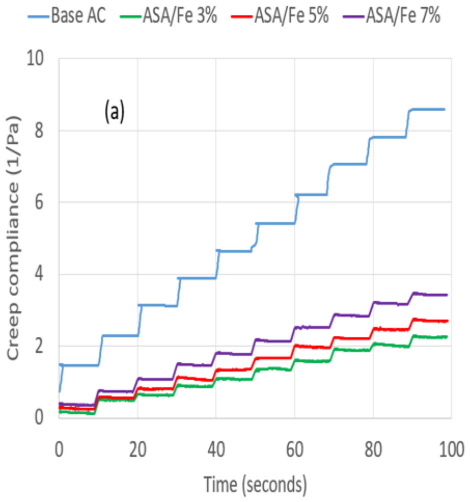


FIGURE 8a. Creep compliance for ASA/Si composites (a) 100 Pa (b) 3200 Pa



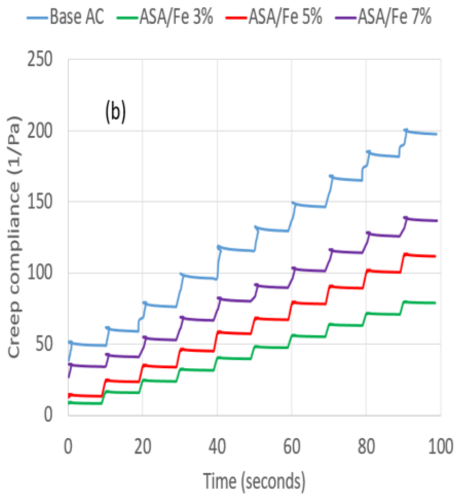
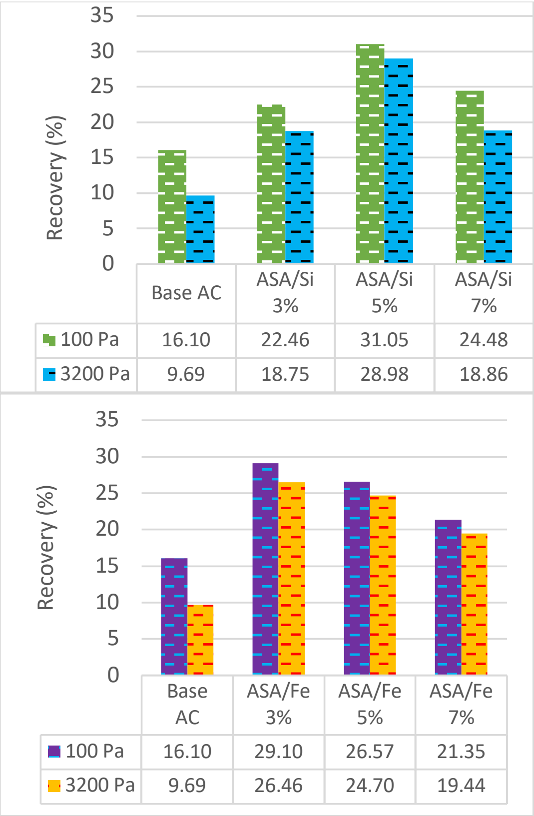


FIGURE 8b. Creep compliance for ASA/Fe composites (a) 100 Pa (b) 3200 Pa

It was observed that, the accumulation of strain was higher for the ASA/Si composites than the ASA/Fe composite modified binders. This was due to ASA/Fe composite modified binders yielded better resistance to permanent deformation (rutting) at high temperatures which was also evidential from the frequency sweep test results. Additionally, it is noteworthy to mention that, the gap of strain accumulation between the base and PNC modified AC for both modifiers reduced at higher stress levels which indicated that the influence of additives were more prominent at low stress levels.

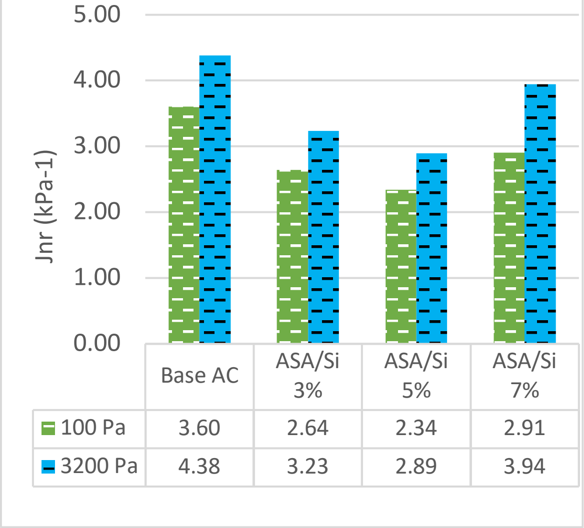
The influence of additives on the performance characteristics was further investigated by analysing the non-recovered strain after the loading phase and the recovery rate during the resting period. Non recoverable strain was denoted as the Jnr and the recovery rate was referred to as the %R. A higher R% and a lesser Jnr were the favourable properties of AC in order to improving the viscoelastic behaviour of AC. From Figure 9a, enhancement in the elastic recovery (%R) for the ASA/Si composite modified AC up to 5% composition was observed, while this increase was less for the 7% ASA/Si modifier content. At 100 Pa stress level, the percentage increase for the ASA/Si at 3, 5 and 7% contents compared to the base asphalt were 39.5%, 92.8% and 52.0% respectively, while at the 3200 Pa stress level the percentage increase were 93.4%, 199.0% and 95.0%. The percentage increase in terms of %R was 81.1%, 65.0% and 32.6% at 100 Pa and it was 173.1%, 154.9% and 100.61% at 3200 Pa for the ASA/Fe composite modified binders. Apart from the %R, the computed Jnr results illustrated in Figure 9b demonstrated that, the non-recoverable creep was reduced after the modification process. The reduction of the Jnr for the ASA/Si composites compared to the base AC were 22.0%, 43.5% and 17.7% at the 100 Pa stress level and it was 31.9%, 41.30% and 12.9% for ASA/Si 3, 5 and 7% respectively. For the ASA/Fe composites 3, 5 and 7% addition of additives resulted in reduction of Jnr by 48.8%, 37.8% and 20.27% at 100 Pa stress level and by 51.3%, 41.3% and 26.1% at 3200 Pa stress level respectively.



	Base AC	ASA/Si 3%	ASA/Si 5%	ASA/Si 7%
🟩 100 Pa	16.10	22.46	31.05	24.48
🟦 3200 Pa	9.69	18.75	28.98	18.86

	Base AC	ASA/Fe 3%	ASA/Fe 5%	ASA/Fe 7%
🟪 100 Pa	16.10	29.10	26.57	21.35
🟨 3200 Pa	9.69	26.46	24.70	19.44

FIGURE 9a. Elastic recovery for base, ASA/Si and ASA/Fe composites



	Base AC	ASA/Si 3%	ASA/Si 5%	ASA/Si 7%
🟩 100 Pa	3.60	2.64	2.34	2.91
🟦 3200 Pa	4.38	3.23	2.89	3.94

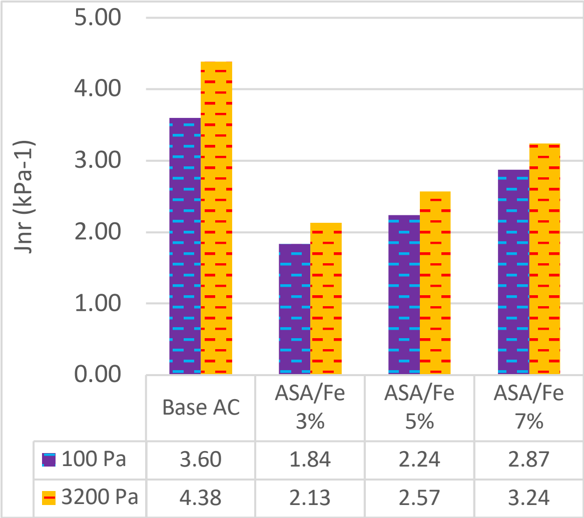


FIGURE 9b. Non-recoverable creep compliance for base, ASA/Si and ASA/Fe composites

CONCLUSION

The significant remarks of the experimental investigations are summarised as followed;

1. Based on the frequency sweep and the MSCR test results it was observed that, 3% addition of nano ironoxide in the blend resulted in the G* at high temperatures which indicated enhanced rutting resistance. However, the increase in the rutting resistance was at the cost of reduced fatigue resistance due to increased brittleness.
2. For the ASA/Si composite modified AC, the improvement in viscoelastic characteristics was consistent up to 5% addition of nano silica in the blend. Beyond this composition, a reduced enhancement in performance characteristics was observed. Although the enhancement in rutting resistance for the ASA/Si composites was lower than the ASA/Fe composites, ASA/Si composites were also able to improve the low temperature performance characteristics of AC.
3. The performance enhancement at 7% ASA/Si composition was slightly less than that of 5% ASA/Si modified AC which was a sign of agglomeration. A similar trend of results was observed for the asphalt cement modified with Fe_3O_4 nano particles where beyond 3% ASA/Fe composition the improvement of viscoelastic properties of AC was reversed. A possible justification for such trend in the results was the occurance of phase separation between the nano particles and the polymer matrix which was also evident in the storage stability test results.

Overall, it can be concluded that promising improvement in the high and intermediate temperature performance of the PNCAC was achieved.

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DECLARATION OF COMPETING INTEREST

None

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