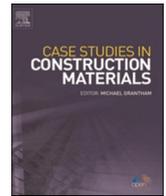




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# Performance of PET and nano-silica modified stone mastic asphalt mixtures

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## ARTICLE INFO

### Keywords:

Nano-silica  
Waste PET plastic  
Hybrid additive  
SMA  
Marshall stability  
Rut depth

## ABSTRACT

Stone mastic asphalt (SMA) has become widely used in the field of pavement engineering. Using specific additives in SMA is necessary due to climate change and load condition variations. Thus, it is essential to utilise a modern mixture with a high capacity to survive the high traffic loads. The current study aims to use a combination of waste polyethene terephthalate (PET) and Nano-silica (NS) as a newly developed hybrid additive in SMA modification to improve rutting and fatigue performance. Waste PET and NS were blended with the C320 bitumen binder by using the wet-mix process, producing SMA mixtures. The Nano-silica was added to the C320 binder (2%, 4%, 6% and 8% by weight of the binder). The engineering properties were assessed through the Marshall stability, wheel tracking test, indirect tensile strength ratio test, resilient modulus test, and drain-off tests. The results indicated that adding 4–8% of Nano-silica with 6% waste PET plastic improves the rutting performance, stiffness modulus, tensile strength ratio, and fatigue performance of the modified SMA mixtures in comparison to non-modified mixtures. The significant performance of rut depth and stiffness modulus, tensile strength ratio and drain-offs were recorded as 1.2 mm, 6100 MPa, 96.4%, and 0.04%, respectively. Results revealed that mixtures modified with the hybrid additive of 6%PET6%NS have superior performance concerning better stiffness and rutting resistance. The results showed that the increase in Nano-silica content with PET plastic increased the fatigue life to the highest extent, at up to 220,490, for the hybrid additive of 6PET6NS mixtures. Based on the results of rutting and fatigue performance, the hybrid additive of 6PET6NS mixtures reflects the optimum and ideal content. The results demonstrated that the inclusion of NS with PET successfully decreased the drain-off rates, increased the rutting resistance, and improved the fatigue life.

## 1. Introduction

The application of waste materials in geotechnical and pavement projects is attracting increased research interest [1–3]. Waste materials had been used in different types of hot mix asphalt including Stone mastic asphalt (SMA), which is showing a significant engineering properties. SMA, first developed in Germany, has proven to be the optimal asphalt pavement, allowing roads to withstand heavy vehicle loading and deterioration. The application of SMA demonstrates the ability to resist rutting and fatigue. SMA has been widely recognised in Europe, Australia, and New Zealand [4].

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<https://doi.org/10.1016/j.cscm.2022.e01044>

Received 31 January 2022; Received in revised form 24 March 2022; Accepted 29 March 2022

Available online 30 March 2022

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SMA is identified by a mixture of gap-graded aggregate, minimising the fine and medium-sized aggregate, resulting in a stable and structurally tough mixture [4,5]. The aggregates stress transformation affect the strength of the mix [3]. The strength and stability of the SMA are due to the stone portion of the coarse aggregate skeleton, which increases the internal friction rate and the resistance of the mixture to shear, enabling it to resist rutting and wearing because of repetitive studded tyre contact [6]. Several benefits are associated with the use of SMA, such as an enhanced asphalt mixture level of resistance to deformation, reduced reflection cracking, better adhesion, good resistance to skidding, stability at high temperature, better durability, and so on [5–7]. More research studies are of obvious interest due to these benefits of using SMA.

Road services and road safety could be impacted by the increasing demand for using road and pavement and increasing weather temperatures due to climate change. As such, the industry and research are working to develop the ideal polymer that could improve the road asphalt mixtures. Various kinds of polymers have been used to modify SMA mixtures. Among these polymers are Styrene-Butadiene-Styrene (SBS), Styrene-Butadiene-Rubber (SBR), Crumb Rubber Modifier (CRM), Ethyl Vinyl Acetate (EVA), and Styrene Polyvinyl chloride (PVC) [5–9]. Adding polymer can further stiffen the bitumen and enhance its resistance to drain down. Furthermore, the polymer can enhance the adhesion property of the bitumen to the aggregate, especially in wet conditions. One of the main advantages of applying polymer technology to bituminous mixtures is to improve the adhesion between the aggregates and binders. A study conducted by Khattak and Baladi [8] on the influence of polymer as a modifier revealed that the modification could increase the resistance of bitumen to loading while making it less susceptible to temperature fluctuations. Numerous studies confirm that the application of crumb rubber, SBS, and waste plastic modified binders to pavement mixtures can improve its resistance against fatigue cracking [5,9–14]. Mashaan et al.'s [15] study investigated the impact of waste PET plastic on the engineering and performance properties of 14 mm dense-graded asphalt, widely used in course surfacing in Western Australia. The study emphasised that the 6% and 8% waste PET are the idealistic contents projected to modify and improve the strength, stiffness, durability, elasticity properties, and rutting resistance of asphalt mixtures. Additionally, the rutting and fatigue properties of SMA mixture using waste high-density polyethylene (HDPE) have been enhanced, as reported by [16]. The study outcome shows that SMA mixtures modified with 4% HDPE have the best fatigue resistance at a fatigue life of 157,090 cycles. However, the 8% HDPE has the better rutting resistance at a rut depth of 1.05 mm.

Besides using polymers as modifiers/additives in asphalt mixtures, the impressive additive of nanomaterials has been used in most hot mix asphalt projects. Nanomaterials display special characteristics compared to normal material and have shown novel properties and excellent performance due to their small nature. Therefore, nanomaterial has a vital feature that can be applied in asphalt pavement as an additive [17]. According to the literature studies [15–19], adding nanoparticles into asphalt improves their physical, engineering, and rheological properties. Studies by [17–30] showed that nanomaterial could fundamentally improve the adhesion and cohesion of asphalt binder/mixture and establish a bridging impact between the asphalt and nanoparticle, avoiding the growth of cracks. Thus, in turn, the life of fatigue may be extended, and the rutting failure could significantly decline [22–26]. Nano-silica's ageing, rutting, and fatigue properties were scientifically investigated [24,25,28,29]. The published results indicated that nano-silica could remarkably improve the bitumen rheological-mechanical characteristics.

According to the literature [24,25,28,29], nano-silica is an excellent additive in pavement modification. There was a new approach to using polymers with nanomaterials as a hybrid polymer to overcome some disadvantages of using a single polymer. Different studies illustrate the different types of polymers and nanomaterials used in asphalt modification and the major findings [25,31–34]. The application of combining 2–4% of Nano-silica with 5% of SBS polymer resulted in improving the asphalt mixture's performance concerning better rutting and fatigue resistance [25,31,32]. The combination uses 1% Nano-silica with waste 5% polyvinyl chloride (PVC) and emphasises that the modified SMA mixture would have the best performance concerning high stability, good moisture failure resistance, and high rutting resistance [33]. Obaid [34] characterised and evaluated the asphalt mixture modified with nano-silica/waste polypropylene polymer (PP). The addition of 5% Nano-silica with 3% PP positively impacts moisture sensitivity, fatigue life, and rutting resistance [34].

In summary, most of the literature of previous studies focuses on the application of nano-clay/polymer in asphalt modification. Additionally, previous studies [25,30–32] mostly highlighted the rheological and physical properties of the bitumen binder modified by Nano-silica. The application of Nano-silica with SBS polymer is well investigated, as discussed in the literature review. Nevertheless, no investigation on the combination of Nano-silica and waste PET plastic modified C320 bitumen has been conducted. As such, it is necessary to investigate and evaluate the engineering, mechanical, and deformation performance properties of combining Nano-silica and waste PET plastic polymer as a 'sustainable hybrid additive'. The study aims to investigate, evaluate, and determine the ideal Nano-silica and waste PET contents to produce a sustainable hybrid additive, which can be effective in the sector of asphalt modification engineering.

## 2. Experimental design

### 2.1. Materials

#### 2.1.1. Bitumen

In this study, C320 bitumen was used. The physical properties of C320 bitumen, including viscosity at 135 °C, penetration at 25 °C and flashpoint, were 0.5 Pa.s, 45 mm and 250 °C, respectively.

#### 2.1.2. Waste plastic

Domestic waste plastic bottles (PET) were assembled, cleaned, ground to a size of 0.45 mm, and used as a bitumen modifier.

### 2.1.3. Nano-silica

The Nano-silica used in this study had a particle diameter of 15 nm, a service area of  $600 \text{ m}^2/\text{g}$ , a bulk density  $< 0.056 \text{ g}/\text{cm}^3$ , a true density of  $2.4 \text{ g}/\text{cm}^3$ , and was coated with 2 wt% Silan- KH220, as shown in Fig. 1, Tables 1 and 2, showed the properties of Nano-silica. The different materials used in this study can be seen in Fig. 2.

### 2.1.4. Aggregate

Granite aggregate, which is the nearly popular natural aggregate in Western Australia, was used. Typical 10 mm SMA for course surfacing was applied. The particle size distribution of the aggregate is presented in Fig. 3.

## 2.2. Preparation of Nano-silica/recycled polymer mixtures and tests

During sample preparation, a total of 6% of PET plastic with Nano-silica concentrations of 0%, 2%, 4%, 6%, and 8% by the weight of the binder were used to prepare the hybrid additive. Before adding the Nano-silica particles, PET plastic is to be first dissolved in a 500 g weight of the base binder. When PET has blended totally in the base binder, the added nano-silica particles are uniform with the PET modified bitumen and blended at a high shearing rate of 4000 rpm. Throughout the blending time, the temperature was kept for 2 h at  $160 \pm 5 \text{ }^\circ\text{C}$ . The SMA samples were prepared following AS/NZS 2891.2.1:2014 [35]. The PET/NS-modified-SMA mixtures were sampled at the optimum binder content following the Marshall method and the wet-mix process. The sample (control binder) used the SMA mixture with 0% PET and 0% NS. In addition, as recommended by the Main Roads Western Australia (MRWA) standard, 1.5% hydrated lime by weight of dry aggregate was used. In the testing stage, the Marshall test [36], wheel-tracking tests [37], IDT resilient modulus test [35], indirect tensile strength test [39], tensile strength ratio [40], and drain-off test [41] were employed to sympathise the effect of PET/NS on the engineering and mechanical properties of modified SMA mixtures. The nanomaterial was supplied by the US Research Nanomaterials Inc, Houston, United States of America.

## 3. Results and discussion

### 3.1. Marshall test

The Marshall test was conducted to investigate and assess the ability of the SMA mixture to withstand substantial loads of traffic and subsequent failure and rutting deformation. Figs. 4–6 display the Marshall stability, Marshall flow, and Marshall quotient results. The Marshall quotient can be defined as the rutting resistance indicator and can be calculated as the ratio of the Marshall stability to the Marshall flow [15]. As shown in Figs. 4 and 6, the increases in different Nano-silica content along with 6%PET has shown an obvious increment in the Marshall stability and Marshall quotient. However, Marshall flow exhibits decreasing results. Studies have indicated that Marshall stability values are an essential indicator of asphalt resistance to shear stress, displacement, and rutting deformation [5,15]. Adding 8%NS and 6%PET could produce the ideal stability and strength properties of the SMA mixture with a high

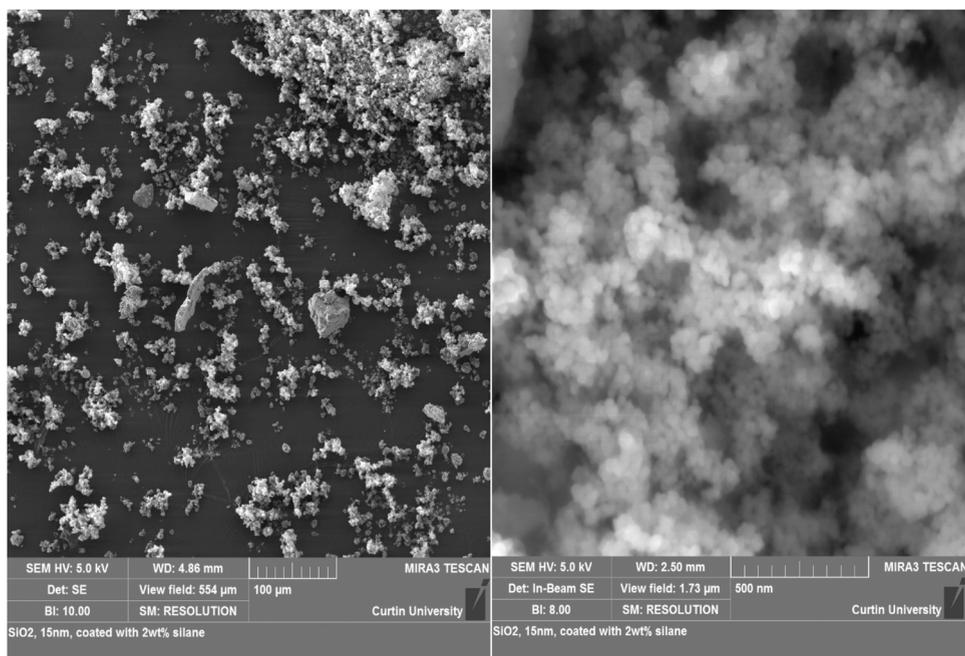


Fig. 1. SEM of Nano-silica used in the study at 100  $\mu\text{m}$  and 500 nm magnification.

**Table 1**  
Elements of NS 15 nm.

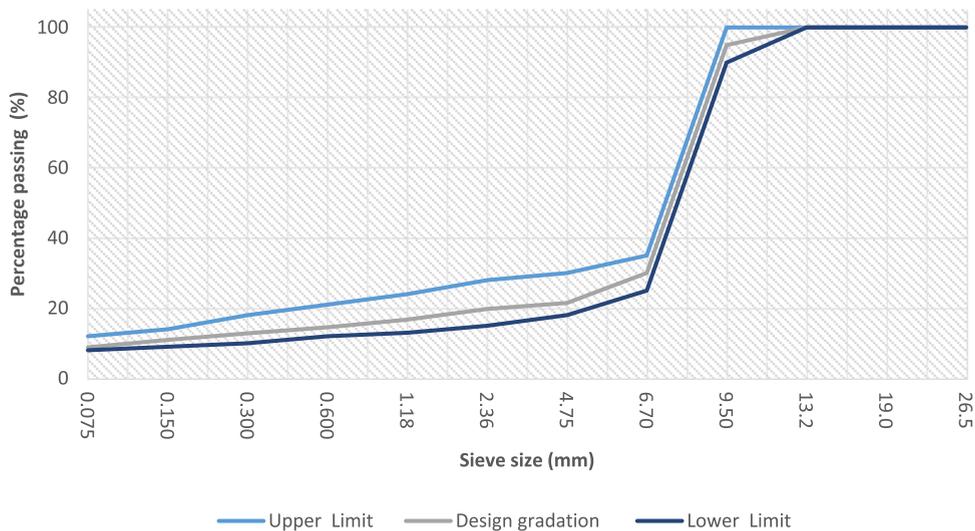
SiO <sub>2</sub> - Silan (wt%)	Mg	Ca	S	Fe
97.3–2.0	75 ppm	< 2 20 ppm	< 126 ppm	< 56 ppm

**Table 2**  
Properties of NS 15 nm.

Service area	Size	Colour	Bulk density	True density
600 m <sup>2</sup> /g	15 nm	white	< 0.056 g/cm <sup>3</sup>	2.4 g/cm <sup>3</sup>



**Fig. 2.** Materials used in the study (nano-silica, waste plastic, bitumen binder, and granite aggregate, from left to right).



**Fig. 3.** Particle size distribution of aggregate.

Marshall stability and Marshall quotient of 20.1 KN and 8.7 KN/mm, respectively. The findings of the current study show a more significant and comparable impact of waste PET with nano-silica in comparison to other previous studies [32,33].

### 3.2. Wheel tracking rutting test

Rutting is mainly the most important distress that can result in the deformation of road pavement, particularly in high-temperature climates. Unsuitable mixture designs, such as higher bitumen content, a higher percentage of coarse aggregate, and an unreasonable

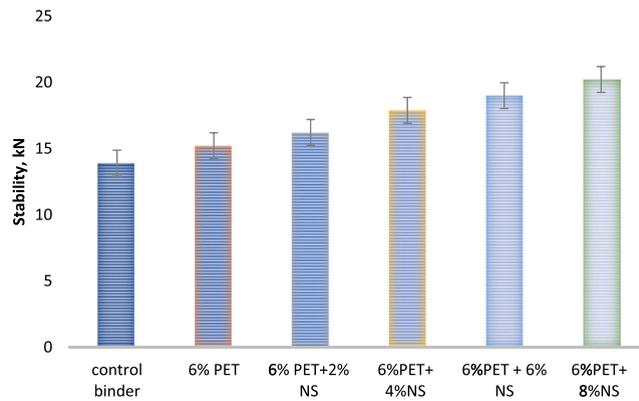


Fig. 4. Marshall stability.

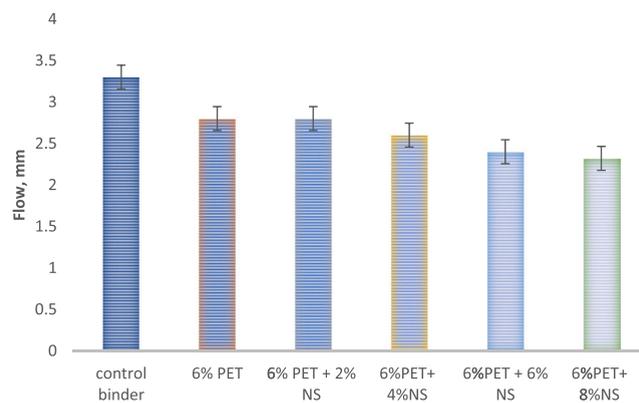


Fig. 5. Marshall flow.

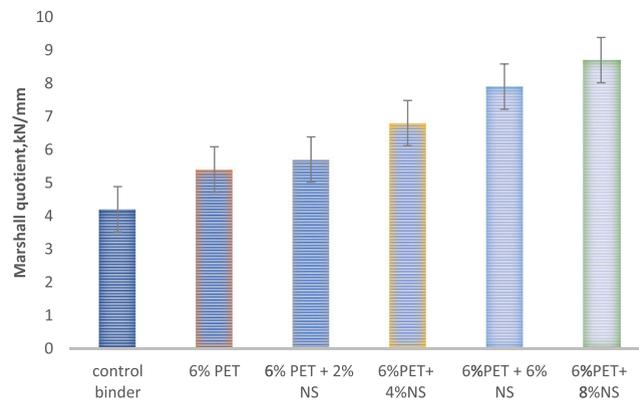


Fig. 6. Marshall Quotient results.

filler amount, could potentially lead to rutting defamtion. Thus, investigating the effect of the modified mixtures to explain the rutting resistance is essential.

The rutting test was conducted per AG: PT/T231 [37]. Fig. 7 displays the rutting depth antonymous to the different modified mixtures, and the data of the rutting test used the average of the three readings for each mixture type and content. Adding PET and NS results in a considerable decrease in the rut depth from 1.2 mm and 2.4 mm for mixtures modified with 6% PET/6% NS and 6% PET/8% NS, respectively. The results indicate the positive effect of NS and PET plastic on rutting resistance. The reason behind this improvement is the growing adhesion force between bitumen binder and aggregate, forbidding the aggregate from slithering due to the influence of compressive loads. As such, the rutting deformation decreased [34]. This result shows that using more Nano-silica in the mixture reduces the rutting depth as compared to non-modified SMA (control binder) and 6% PET-modified SMA. However, the result

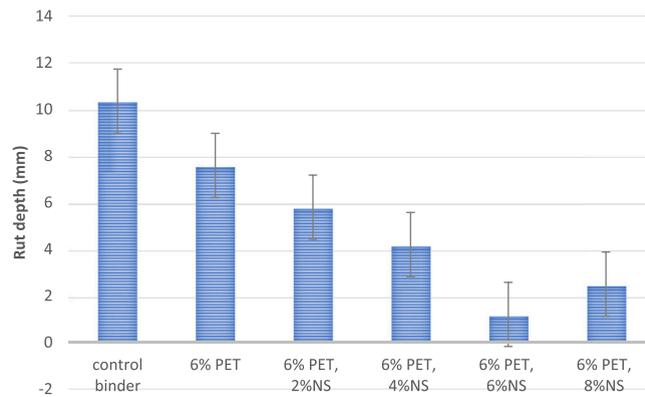


Fig. 7. Results of rutting resistance.

of 6%PET is of concern compared to the non-modified SMA. The findings of the current study demonstrate a more significant and comparable impact of waste PET with nano-silica in comparison to other previous studies [32–34]. The results indicate that adding NS and PET leads to SMA mixture stiffness and enhanced strength. This demonstrates a rise in the capability of the modified stone mastic asphalt to withstand and resist accumulative deformation under heavy traffic loads. Therefore, the rutting resistance of the SMA mixtures is enhanced by utilising PET and NS.

### 3.3. Resilient modulus

Resilient modulus is considered one of the most important tests for the mechanistic design method for asphalt pavement. This test is used to measure the asphalt consequence concerning the dynamic stress and related strains as per [38]. Therefore, the major objective of conducting this test was to determine if adding Nano-silica would bring meaningful change in the stiffness properties of SMA modified mixtures.

Fig. 8 shows the stiffness modulus results using different contents of nano-silica and waste PET. The hybrid binder modified SMA mixture has a comparatively higher stiffness resilient modulus than the control binder (non-modified). The stiffness modulus values are noted to increase from 5010 MPa of 6%PET samples to 5345, 5788, 5897, and 6100 MPa for the hybrid additive of 6PET/2NS, 6PET/4NS, 6PET/6NS and 6PET/8NS, respectively. This implies that the modified mixture would have higher load spreadability, high load-bearing capacity, and more resistance to pavement deformation than non-modified mixtures. This increase in the stiffness-resilient modulus is attributed to the role of nanomaterial in improving the binder performance and the mixture resistance to high temperature and high traffic load. Another reason behind the increase in the stiffness modulus is the increase in viscosity, improving the adhesion between the aggregate and bitumen and preventing the mixture from sliding easily [25].

### 3.4. Indirect tensile strength test

Permanent deformation, cracking, moisture susceptibility, and stripping can be evaluated by conducting the indirect tensile strength test (ITS) [39]. The results of ITS and the tensile strength ratio (TSR) are illustrated in Fig. 9. The non-modified-SMA mixture has a lower tensile strength value than the high values of the plastic and nano modified SMA. The results show that the more Nano-silica content added to the SMA mixture, the further resistance to stripping and less moisture susceptibility will be established.

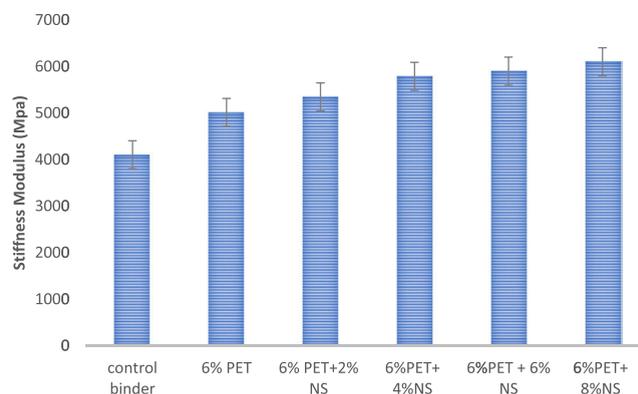


Fig. 8. Stiffness modulus (resilient modulus) results.

The results displayed that the TSR% of non-modified SMA and highest sample modified with 6PET4NS were 84% and 96%, respectively. Nano-silica possesses very small fragment sizes and a bigger surface area so that it can absorb more binder and increase the structurally modified binder [33]. The TSR values of all modified SMA samples increased. This indicates that PET and Nano-silica worked significantly to improve the tensile strength resistance by developing strong bonding forces between bitumen binder and aggregate. This implies that mixing the PET and nano-silica reduced the displacement of the bitumen-coated aggregate through the development of adhesive and cohesive bonds. As such, they developed better resistance to stripping and moisture damage [34].

Fig. 9 also shows that the tensile strength ITS of the conditioned samples is less than that of un-conditioned samples, similar to previous findings [25,33]. The reason behind this reduction of ITS conditioned samples possible would be ascribed to less cohesion in the mixtures, resulting from the long coverage to moisture and wet conditions. In addition, the small difference between the conditioned and unconditioned samples shows that adding the combined additive of PET and NS to the SMA mixture would not effortlessly allow the displacement of bitumen coating the aggregate surface due to the improvement of adhesion bonds between aggregate and bitumen [25]. Fig. 3 shows that all PET/NS modified SMA samples have satisfactory resistance against moisture damage and show the ability to increase the bond strength between the aggregate and bitumen binder. Fig. 9 shows that the mixture modified with 4% NS displays the maximum increase in ITS, about 50% higher than the control un-modified SMA mixture. On the other hand, mixtures with further Nano of 6% and 8% result in a slight reduction of the ITS values of about 1.4–1.2, respectively. The reason for this could be associated with stiffness reduction while adding high percentages of nanoparticles. However, all PET and NS modified mixtures exhibited a fundamental improvement of the modified mixtures concerning better resistance to rutting and fatigue cracking.

### 3.5. Binder drain-off test

SMA is identified as a mixture of gap-graded aggregate, which minimises the fine and medium-sized aggregate. Some weaknesses are distinguishable in the SMA structure, such as drain down, which ensues from the absence of the mid-sized aggregate in the gap-graded mixture and has high asphalt binder content instead. As such, it is important to test the drain down of the mixtures. Austroads AG: PT/T235 [41] drain-off test was followed for this purpose. Fig. 10 shows the results of the drain-off test. As can be seen, there was a tremendous improvement to the varied SMA mixtures in comparison to the non-modified mixture. The drain-off value of the non-modified SMA mixture produced a drain-off approximation of 0.22%, which is within the standard boundary. The addition of the 6%PET resulted in a decrease of drain off of about 0.9%. At the 6%PET with the 8%NS substance, the drain off effectively lessened to 0.04%. As shown in Fig. 10, adding NS and PET as a hybrid additive in the SMA mixture lowers the drain-off and improves resistance to the deformation of the SMA modified mixture. A limit of 0.3% binder drain-off was recommended as the maximum value [25,36]. The results of these findings were contrary to [25,33], where the drain off of PP plastic showed a decrease in adjustment with the adequate limits. Further, a study by [4] using EVA plastic modified SMA revealed drain-off results within satisfactory limits.

### 3.6. Fatigue

The fatigue strength and fatigue life of the PET-Nano-silica-modified asphalt were measured using the four-point bending fatigue test per the Austroads AGPT/233 method. The failure based on the standard was if the flexural stiffness dropped to 50% of the initial flexural stiffness or at 1 million cycles (whichever occurs first). The samples were inserted into the universal testing machine (UTM) for 50 preloading cycles. The value was measured at this stage. The results showed that the increase in Nano-silica content with PET plastic increased the fatigue to the highest level of fatigue life up to 220,490 of hybrid additive of 6PET6NS mixtures. Then, they declined slightly to 215,070 of 6PET6NS mixtures. All the modified mixtures were higher than the non-modified mixture, which had the lowest fatigue life of only 67,680, as shown in Fig. 11. The use of Nano-silica with different contents of 2%, 4%, 6% and 8% had a better fatigue life than samples using only 6% PET. This explains that the Nano-silica enhanced the polymer-bitumen phase. This indicates that Nano-silica particles have a significant effect on the fatigue life of the mixtures. This can be attributed to the surface energy of Nano-silica, which inhibits the aggregation of small PET particles in the mixture, increasing elasticity and the adhesion of the binder to aggregates [23,25].

Fig. 12 shows the initial flexural strength of PET-Nano-Silica-modified asphalt. The flexural strength of all modified asphalt mixtures samples was high in comparison to the non-modified asphalt mixtures, which had 5441 MPa. Then, the flexural strength increased slightly by adding 6% PET plastic to 5997 MPa. However, by introducing the Nano-silica to the 6% PET plastic binder, the flexural strength improved and showed an obvious increase of about 6234 MPa, 6641 MPa, and 6818 MPa at 2%, 4%, and 6% NS.

Fig. 13 shows the initial modulus of the elasticity of the PET-Nano-silica-modified asphalt. The increase in the initial modulus of elasticity of the treated PET-bitumen plastic samples and treated PET-NS-bitumen samples were higher than that of the untreated samples. The highest amount of hybrid additive of 6% PET and 8%NS resulted in the highest initial modulus of elasticity at about 7137 MPa. The figure shows fluctuation increases in the initial modulus of elasticity. This would result in improvements to the fatigue resistance of the modified mixture, demonstrating a more significant effect of using waste PET with nano-silica in comparison to other previous studies [25,31,34].

Fig. 14 shows the phase angle of the PET-Nanosilica-modified asphalt using the fatigue tests. The phase angle of the hybrid additive of 6%PET and 6%NS shows a better elasticity of about 26.7 than the single additive of 6%PET at about 28.2. The combination of Nano-silica to the waste PET plastic created more stable and strong coupling and bonding between the particles of the plastic and nano, enhancing the bonding of polymer-nano phase dispersion in the bitumen particles. The mixtures modified with plastic-nano polymer would have high resistance to deformation [25,31,34]. Improvements in the phase angle would improve the fatigue life and, as such, the resistance to cracking. These findings are in line with previous studies that used a combination of polymers and nanomaterials for

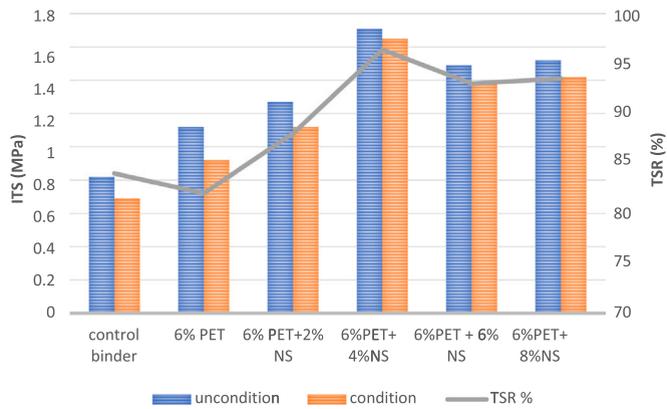


Fig. 9. Indirect tensile strength and TSR % results.

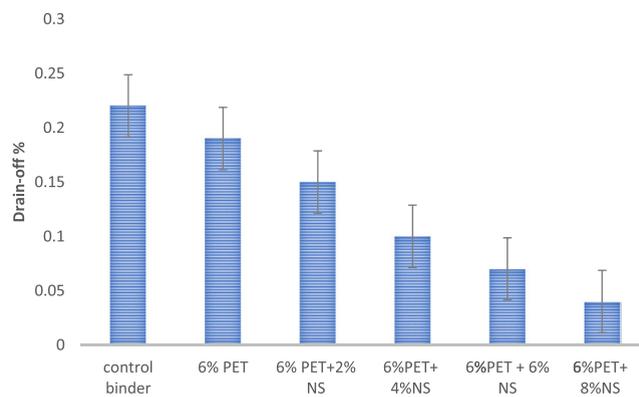


Fig. 10. Binder drain-off results.

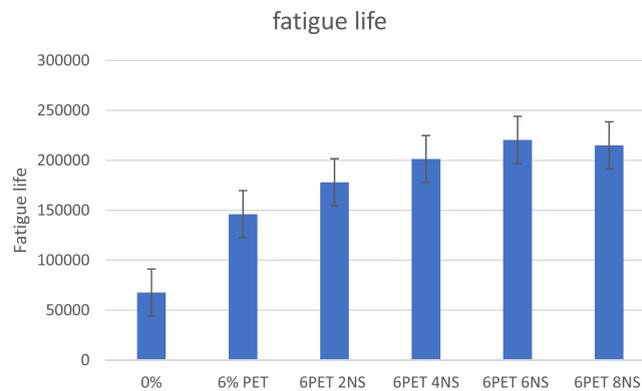


Fig. 11. Fatigue life of different binders.

asphalt modification [25,31,34].

Fig. 15 shows the cumulative dissipated energy of PET-Nano-silica-modified asphalt. The untreated samples and 6%PET treated samples had a high value of dissipated energy of about 1.952 kPa and 1.981 kPa, respectively. By adding 2%, 4%, and 6% Nano-silica to the plastic and producing the hybrid polymer, the dissipated energy decreased to 1.88, 1.79 and 1.716. However, the dissipated energy then increased to 1.85 by adding 8% Nano-silica. This would improve the fatigue resistance of the modified mixture, and these findings further demonstrate a more remarkable effect of waste PET with nano-silica in comparison to other previous studies [25,31, 34].

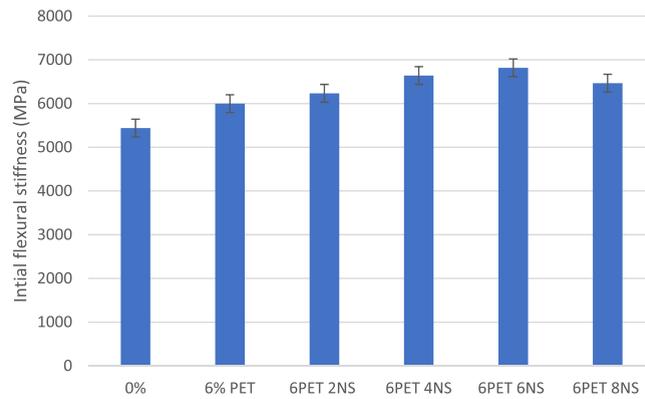


Fig. 12. Initial flexural strength results.

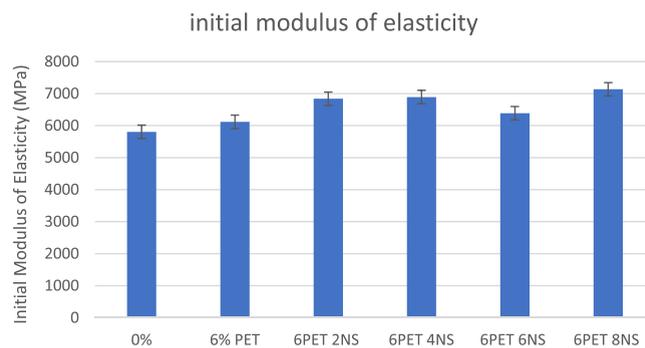


Fig. 13. initial modulus of elasticity result.

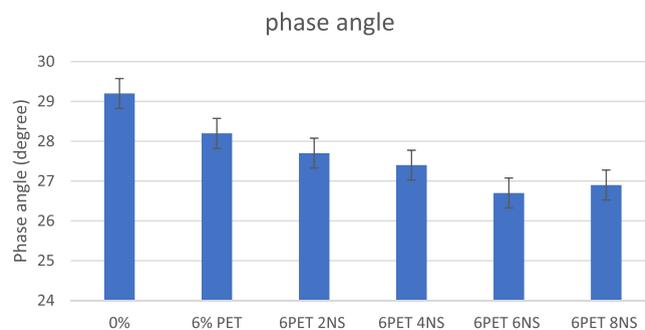


Fig. 14. Phase angle results.

#### 4. Conclusion and recommendations

In this study, the influence of nano-silica (NS) and waste PET on the rutting resistance and permanent deformation of asphalt mixtures were investigated and evaluated. The mechanical and engineering properties were examined and assessed through the Marshall stability, Marshall flow, wheel tracking test, indirect tensile strength ratio test, indirect tensile stiffness modulus test, and drain-off tests. The below conclusions can be drawn:

1. The study shows an innovative approach to recycling plastic in asphalt modification and the asphalt industry. The 6% PET mixtures had a comparative ability to the mixtures with the addition of 2% Nano-silica concerning stability and rutting resistance performance.
2. The increases in different Nano-silica substances alongside with 6%PET demonstrated an obvious increase in the Marshall stability and Marshall quotient. However, Marshall flow shows declined results. The higher Marshall stability values indicate that the hybrid additive of NS/PET samples has become stiffer. Therefore, modified SMA mixtures are more resistant to deformation.

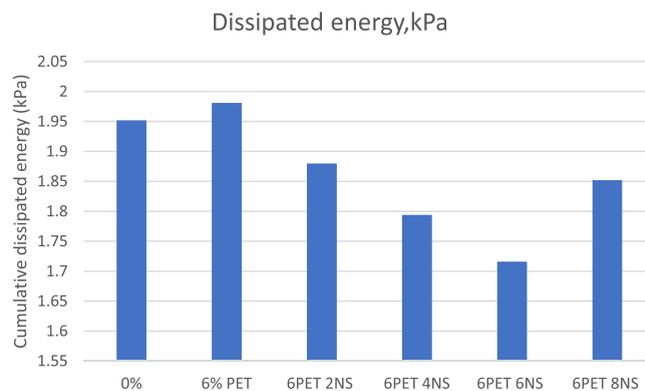


Fig. 15. Cumulative dissipated energy results.

- NS results in a sustainable decrease in rut depth to 1.2 mm and 2.4 mm for mixtures modified with 6% PET/6% NS and 6% PET/8% NS, respectively. The results imply the positive effect of NS and PET plastic on rutting resistance.
- The resilient modulus values were increased up to 6100 MPa for the hybrid additive of 6PET/8NS. In addition, the results displayed that the TSR % of SMA modified with 6PET4NS has the highest value of 96%. This implies that the modified mixtures are more resistant to rutting deformation in comparison to non-modified mixtures.
- The drain-off of modified SMA mixtures confirmed a decrease in values compared to non-modified mixtures. The addition of the only 6%PET resulted in a decrease of drain off of about 0.9%. At 6%PET with 8%NS, the drain off effectively diminished to 0.04%. The results demonstrated that the inclusion of NS with PET successfully decreased the drain-off rates.
- In addition, it is recommended to do more tests at different stresses and temperatures to consider the rutting resistance and fatigue performance. After all, it is recommended to test the fatigue life and fatigue deformation of the different percentages of the hybrid additive using the SMA mixture.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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