



Effects of the Addition of different sizes and contents of polyethylene terephthalate (PET) on hot mix asphalt.

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Abstract

Several studies focus on recovering plastic waste for use in road construction rather than banning it or using environmentally harmful methods such as incineration or landfills. Moreover, the intensity of road traffic is increasing today.

Using plastic waste as an additive in road projects would overcome the ecological crisis and improve its properties because of its availability, lightness, and flexibility advantages.

This study aims to assess the performance of bituminous concrete with the addition of PET plastic waste by dry process. The optimum bitumen content used in the bituminous mixture was replaced by 3, 5, and 7% content to prepare modified asphalt mixes. The results of the Marshall test, creep-recovery test, rutting test, the Retained Strength Test, and the leaching test of the PET-modified asphalt mix were evaluated and compared to the control asphalt mix (without PET). The results concluded that adding 5% of this coarse and fine polymer to mixtures helps to increase the rutting resistance by 34% and 26%, respectively, and the Marshall quotient by 95% and 81%. It also reduces deformation by creep recovery by 25% and 20% compared to the control asphalt mixtures. It provides excellent behavior in the presence of water, where the water resistance of the control asphalt mix improved after modification by 5% (19% for coarse PET, and 12% for fine PET). All of these can modify numerous properties and improve the mechanical performance of bituminous mixes, PET does not present any particular risk of aggressiveness at ambient temperature.

Keywords: PET plastic waste, the bituminous mixture, Marshall quotient, creep-recovery, rutting, the Retained Strength test, leaching test.

1. Introduction

From 2009 to 2013, the average annual growth rate of the polyethylene terephthalate (PET) bottle industry was 4.3% (Padhan et al., 2013). According to the Earth Day organizing committee, one million bottles are bought every minute in the United States, and the average American citizen buys 167 plastic bottles a year (Foolmaun and Ramjeeawon, 2012). In 2024, the worldwide production of PET is projected to reach 35.3 million tons, indicating a growth of 4.8 million tons compared to the output in 2019 (PET Preform Market Source., 2023). In the same respect, PET plastic accounts

for 3.57% of the total waste generated annually in Algeria, equivalent to 15.60 billion 1.5-liter drinking water bottles. Hence, the consumption of mineral water is growing by more than 22% every year, with new brands always appearing (The National Waste Agency, 2021). Indeed, the various markets of PET packaging are expected to be worth a total of USD 74.2 billion by 2026; the forecast period of 2021-2026 is expected to have an annual growth rate of approximately 4.93%. One way PET waste can be reused is in the surface course of pavement, where it can be added to the asphalt mix or used as a substitute for fine aggregate (Luu and Baker, 2021; Ahmadinia et al., 2012). As a result, using PET waste in flexible pavement construction and road restoration can potentially eliminate a significant quantity of several million tonnes, with positive environmental effects such as reduced pollution and environmental impact (Yan et al., 2019), reduced drainage problems due to drainage systems clogged with PET waste, longer landfill life, preserving of natural resources used in asphalt mixes, like bitumen, and improved road performance (Ben Zair et al., 2021).

Hinisliog et al. (2005) used PET plastic waste with seven different contents by weight of bitumen (2.5-15%). The best increase in Marshall's stability (MS) of 25.44% was achieved with 10% PET. Hassani et al. (2005) used the Marshall and rutting test, and their results concluded that the replacement of 20% of the fine aggregate was equivalent to 5% of the weight of the bituminous mix. The asphalt mix that gave the best stability at the lowest flow and therefore the highest Marshall Quotient (MQ), was the PET size from 2.36mm to 4.75mm; After that, the density of the modified bituminous mix was reduced by 2.8%, which is less than that of the unmodified asphalt mix. It was also found that replacing the aggregates with PET increased the rutting resistance but decreased the stability and stiffness of the mixtures. Ahmadinia et al. (2011) determined the effect of incorporating 2 to 10% PET of the total mix weight on the characteristics of bituminous mixes. The researchers observed that the stability values exhibited an upward trend upon incorporating

PET, culminating in a peak value at a PET content of 6%, and then began to decrease. The only mix with a lower stability value than the control mix was the 10% PET. Moghaddam et al. (2013) used different PET contents (0.2 to 1% by weight of bituminous mix) with a size of 2.36 mm. It has been shown that the incorporation of polyethylene terephthalate particles up to 0.4% increased the MS value by 9.98%, while the further addition of PET (1% PET) reduced this value. On the other hand, adding the optimal amount of 1% PET to the bituminous mix increased the flow of the PET-modified mix by 12.84%. The authors observed a decrease in sample stiffness as PET increased. It also concluded that the modified mixture's permanent deformation was remarkably reduced compared to the control mixture at all stress levels and test temperatures. A range of PET waste contents, from 2.5 to 15 %, were used by Ahmadinia et al. and found that PET, which represents 6% (maximum value) of the total weight of the bitumen and can pass through a 1.18mm sieve during the dry process, increased the MS value by 16%, compared to the control mix. When the optimum PET content of 2% was added to the mix, the polyethylene terephthalate-modified asphalt mix's indirect tensile strength (ITS) increased by 12.74% (Ahmadinia et al., 2012). Ziari et al. (2016) sought to assess the impact of including PET waste on the properties of asphalt mixtures. The rutting performance of asphalt mixtures included various percentages of PET (0, 0.25, 0.5, 0.75, and 1%) and various sizes of PET (10×2.5 , 20×2.5 , and 30×2.5 mm). The results indicated that the rutting resistance of the mixtures increased with the addition of PET content. With the increase in PET dimensions, the permanent deformations of bituminous mixtures will decrease. Jan et al. (2017) confirmed that temperature directly affected the ITS of specimens when they tested them at 5 and 20°C, where they found that adding 2% of the plastic waste (PET) improved the indirect tensile strength at these test temperatures. Furthermore, the ITS decreased steadily as the PET content increased. When an optimal PET content of 2% was added to the mix, the ITS of the PET-modified bituminous mix increased by 13.1%. The study by Choudhary et al. (2018) was

based on three factors: the first was the mixing method, either dry or modified dry; the second was the addition of three different contents of PET plastic, using 2.5, 5 and 7.5% of the bitumen weight; as for the third factor, it was represented by the two sizes of PET used, 1.18 to 2.36 mm and 0.15 to 0.30 mm. Both processes were significantly more stable than the unmodified mix at the optimum 5% content. Overall, the coarse PET size blends (2.36–1.18 mm) produced by the modified dry process performed relatively better than the fine-size mixes (Choudhary et al., 2018).

This study aims to investigate the mechanical performance of bituminous mixtures without and with modification by coarse and fine PET plastic waste at three different contents under constant stress (creep) and dynamic load (rutting) at high temperatures, as well as the effect of water on these asphalt mixtures and the harmfulness of this waste plastic.

2. Materials

This study utilised several materials, including bitumen (40/50), plastic bottle waste, and various aggregates such as 8/15 gravel, 3/8 gravel, 0/3 sand, and limestone filler.

2.1. Bitumen

The bitumen used in our study was supplied by the company's asphalt plant (EPTP) in Bechar, Algeria. Table 1 summarizes its characteristics.

2.2. Plastic waste (PET)

This time, the polyethylene (PE) used is polyethylene terephthalate (PET) obtained by recycling plastic bottle waste. The waste from these bottles takes the form of granular powders; after passing through the shredder, they must be washed and dried.

This study utilized two primary sizes of waste PET (Figure 2): the first size passed through a 2.50 mm sieve and was retained on a 1.25 mm sieve (referred to as 'coarse' waste PET), and the second size passed through a 0.315 mm sieve and was retained on a 0.16 mm sieve (referred to as 'fine' waste PET). Table 2 shows the characteristics of this waste.

Aggregates

The present study used limestone aggregates obtained from the SARL BOUCHETA crushing station in Bechar. The three grading classes sampled are crushed sand 0/3, gravel class 3/8, and 8/15 for formulating a semi-graded asphalt mix 0/14 according to the grading scheme proposed by CTTTP-Algeria. These aggregates were subjected to physic-mechanical characterization and production tests. Table 3 shows the results of the various analyses.

According to the results obtained, considering the high carbonate content and clean fillers, the aggregates behaved well in hardness and adhesion, which played an important role in formulating a good control mix.

2.3. Formulation of control bituminous mixes (without modification)

The formulation study for bituminous mixes, in general, includes two phases:

- Granular composition;
- Bitumen content.

Fig. 2 shows the best aggregate composition compatible with the SETRA-LCPC 0/14 mm asphalt concrete reference for wearing courses.

Based on the various binder content results from the Marshall test, the optimum bitumen dosage of 5.23% gives acceptable results and is adopted as the control asphalt concrete (Bekhedda et al., 2024).

2.4. Formulation of PET-modified bituminous mixtures

In our study, we chose PET plastic waste because of its many advantages and ease of use. Two methods can be used to incorporate plastic waste into the bituminous mix: wet and dry. The first involves mixing the plastic into the bitumen using a mixer and then adding it to the aggregate. The second is to add the plastic directly to the heated aggregate and mix it with the bitumen.

The modification principle in our study is to replace variable percentages of bitumen of 3, 5, and

7% with crushed PET of two sizes. The aggregates were dried in a ventilated oven at 170°C for about three hours; the bitumen was heated at 150°C for one hour. We then mix all the components (sand, aggregate, filler, bitumen, and waste bottle) in a mixer for five minutes at 160°C to produce a visually homogeneous mixture. These test conditions were chosen according to (Bekhedda and Merbouh, 2022).

After this modification, a comparative study was conducted between the characteristics of the control bituminous concrete (without PET) and the bituminous mix obtained from the modification to determine the mechanical performance of these products and to study the influence of crushed PET. A series of tests were carried out under different conditions, the Marshall test, the Retained Strength test, and the leaching test.

We note:

C-HMA: it is control hot mix asphalt;

Coarse-PET HMA: it is hot mix asphalt modified with coarse PET plastic waste;

Fine-PET HMA: it is hot mix asphalt modified with fine PET plastic waste.

2.5.1. Marshall test

The Marshall Test concept was developed in 1948 by Bruce Marshall at the Mississippi State Highway Department, USA (Boucherba., 2019). In the laboratory, at a given temperature and compression energy, this test measures the resistance of a specimen to deformation when a load is gradually applied, and the deformation of this specimen at the time of fracture when the maximum load is applied known as stability and Marshall Flow. Several standards govern this process, such as the European standard. The Marshall characteristics are believed to provide clear indications of pavement performance (White and Magee, 2019).

Four-point bending

Our study used a four-point bending test to study the creep-recovery behavior of bituminous mixes.

This test allows us to better simulate the physical phenomenon of a vehicle axle on two tires on the roadway. On the other hand, this test is very important in evaluating the resistance of bituminous concrete to viscoelastic deformations (the Marshall test is not sufficient). It was also associated with the rutting test carried out on the real roadway structure.

The bituminous mixture was manufactured in a parallelepiped plate of dimensions 40×30×5 cm (Figure 3). Compaction was carried out according to the European standard (EN. 12697-33:2003) using a mild steel roller compactor. This plate was sawn into several prismatic specimens of dimensions 20x20x5 cm (Figure 4) according to the conditions of the creep-recovery tests that will be carried out. The samples were tested at a high temperature (50 °C). The HMA is subjected to a constant stress $\sigma_0 = 17$ kPA for a duration $t_M = 1$ hour from $t = 0$ S, and its deformation evolution is followed (Figure 5). An object is said to creep or to present the creep phenomenon if the deformation of the material under constant stress is a function of time until maximum (or total) deformation ϵ_{max} , i.e., one hour after stabilization. After a duration of t_M , the stress suddenly drops to zero. We continue to follow the residual deformation (ϵ_{Res}) over time. The complete curve consists of two parts; the first part is the creep, while the second part is called the recovery curve.

2.5.2. The rut

The rut in the roadways results from the repeated passage of vehicles on the roadway. Characterize the resistance to permanent deformation of bituminous mixtures under conditions similar to road stresses, according to the European standard EN 12697-22 standard for large-scale devices.

In the laboratory, on a reduced scale, the test consists of passing a wheel over a bituminous mixture plate with 30 x 30 x 5 cm dimensions. The variables of this test are the loading time (number of cycles) and the temperature at which the test was carried out (60 °C). Two pneumatic wheels in sinusoidal alternating motion, with or without skidding effect, each pass over a bituminous coating

plate whose degradation is observed under severe temperature conditions (Figure 6). The rutting test is performed on the control and PET-HMA. After applying 1000 cold cycles, these plates are kept for 16 hours, in our case, at 60°C inside the rutting apparatus, and we start the simulation of 1000, 3000, 10000, and 30000 cycles.

2.5.3. Immersion Compression Test (index of retained strength)

The water sensitivity of asphalt mixes indicates their susceptibility to moisture damage, which is the most common damage observed on our roads (Wang et al., 2019; Qin et al., 2019). The build-up of moisture in asphalt mixes can lead to several types of damage, including stripping, cracking, and the sudden appearance of large potholes (Ahmadinia et al., 2011). Many tests exist to assess materials' resistance to water-induced degradation and are mostly standardized.

This method specifies a test to determine the water resistance of hot mix asphalt and to provide other resistance indices from the stability ratios with and without immersion of the specimens measured by the Marshall test (Ge et al., 2019) according to the standard AASHTO T 165-02. Six specimens were used; Three specimens were stored for 24 hours at ambient temperature (25°C) on a flat surface without immersion. Subsequently, the remaining three samples with identical composition were subjected to immersion in water at a maximum temperature of 60°C for 24 hours. The determination involves calculating the index of retained strength ratio (MS₀) by comparing the compressive strength of the samples that have been immersed (S₂) with the compressive strength of the samples that have been kept dry (S₁).

2.5.4. Leaching test

When asphalt mixes are leached, this phenomenon leads to the release of salt and the formation of pores connected to the outside. The pores form due to the expansion of the soluble salts caused by the accumulation of water. Over time, this porosity leads to an increase in the surface area accessible to the leaching solution.

This test aims to determine the harmfulness of waste polyethylene terephthalate waste to the surrounding soil (environment) and living organisms (plants, animals, and insects) on the road with modified asphalt. Furthermore, it elucidates the nature of leaching when water comes into contact with unmodified and modified asphalt concrete. We analyzed the sulfate and chloride amounts in this water after 0, 3, and 7 days of immersion. The pH was also monitored. The samples used for this test are always of the Marshall type. We take two control samples (unmodified by PET) and place them in a two-liter water bath at ambient temperature. The same procedure was used for the other PET-modified asphalt samples. After 03 and 07 days, a quantity of water is taken from each bottle for chemical analysis at the LTPO laboratory (Figure 7).

3. Results and discussion

3.1. Marshall Parameters

Marshall stability:

The figures below show the Marshall test characteristics of the coarse and fine waste PET-modified asphalt mixes compared to the control hot mix asphalt (C-HMA).

Figure 8 shows the Marshall stability results for the unmodified and modified bituminous mixes.

The stability values of modified hot mix asphalt are always higher than those of unmodified mixes for all PET percentages and sizes. Additionally, Figure 8 demonstrates that employing a coarser PET size increases Marshall's stability values across all PET percentages. Furthermore, the impact of size remains constant across all PET concentrations.

Adding plastic waste to the bituminous mix positively affects Marshall's stability for the different percentages of PET studied, as it improves the adhesion between the materials in the mix. This confirms that PET has fulfilled one of the purposes for which it was introduced into HMA.

For the HMA modified, adding 5% PET resulted in a 48% and 36% improvement in Marshall stability compared to the C-HMA for coarse and fine PET sizes, respectively. This shows that the

incorporation of waste PET increases the Marshall's stability of the mixture up to a certain PET content of 7%, where we have the lowest stability (26% at 7%Coarse-PET HMA and 21% at 7%Fine-PET HMA). Adding more PET reduces the compressive strength of the mix. The increase or decrease in stability can be attributed to the increased viscosity of the bitumen in the presence of PET waste (Modarres and Hamedi., 2014; Ewa et al., 2024).

Since we are dealing with the same granular composition, the improvement in the MS results is due to the to the bitumen-aggregate bonding parameter; according to Boucherba. the mechanical properties and durability of bituminous mixes are ensured by the stability of the granular skeleton (Boucherba, 2019), the cohesive characteristics of the bitumen, and good adhesion between this binder and the aggregates (Jegatheesan et al., 2020; Agha et al., 2023).

In this case, and according to the results obtained, it can be said that PET increased the adhesion energy, which led to an increase in resistance (stability). This increase is due to the change in bitumen's physico-chemical structure and behavior, which becomes more consistent, more adhesive, and less sensitive to temperature. This interpretation is discussed by various researchers, such as (Hinisiog̃ lu et al.,2005; Hassani et al., 2005; Eisa et al., 2019).

Marshall Flow

Figure 9 shows the evolution of the permanent deformation known as flow in the Marshall test.

There was no clear trend in the contents or sizes of PET waste.

Nevertheless, all the flow values respect the specified range of 2 to 4 mm; for bituminous mixes with a coarse PET size, the Flow values are higher than for HMA with a fine PET size at 3 and 5%.

Contrary to the stability values, the deformations decrease by 11 to 24% for HMA with a coarse PET, respecting the minimum threshold of the American specifications, which recommend a minimum flow of 02 mm. The exact effect is seen for fine PET, with the rate of decrease ranging from 8 to 27% as the plastic content increases, and the Marshall's flow rate is increased due to the

lower internal friction compared to other specimens with low PET content. Despite this, both types of polyethylene terephthalate (coarse and fine) positively reduce the deformation rate of HMA.

The difference in the flow values between the two sizes of polyethylene is due to the different effects on the physicochemical structure of the bituminous mixes (Moghaddam et al., 2013., Ewa et al., 2024).

Marshall Quotient

The following parameter that needs to be more discussed by researchers, despite its importance in the behavior of HMA, is the Marshall quotient. According to Hattatoglu, The Marshall Quotient (MQ) represents the ratio between the Marshall stability (MS) and the Marshall flow (MF) (Hattatoglu et al., 2015).

The MQ is frequently employed as a straightforward metric to assess the HMA's ability to withstand permanent deformation during usage. Higher values of the MQ signify a more rigid mixture that exhibits greater resistance to rutting (Ameri et al., 2020); the results of MQ are represented in Figure 10.

Except for the 5% coarse PET blend, slightly above the specified upper limit, all PET-modified HMAs were within the acceptable MQ range of 2 to 5 kN/mm.

A peak is observed at 5% coarse PET content; at 7 % of polyethylene terephthalate, the values of the Marshall quotient are slightly lower, regardless of the size of the PET.

The addition of coarse PET to the mixes increases the MQ by 65, 95 and 43% for 3, 5, and 7%, respectively, while fine PET increases this resistance with rates of 76, 81 and 32% for the three contents respectively; The results concluded that adding polyethylene terephthalate to the mix increases the Marshall quotient by making it stiffer and more resistant to severe deformation caused by heavy loads.

Marshall's properties show that modified-HMAs are, in most cases, stiffer than C-HMAs due to

their relatively higher MQ, MS, and MF. The increased stiffness of modified HMAs is due to the semi-crystalline nature of polyethylene terephthalate (Ahmadinia et al., 2011; Moghaddam et al., 2013). The latter means that part of the polymer is amorphous while the other is crystalline. PET's amorphous part is liquid above its glass transition temperature of around 70°C. On the other hand, the melting point of PET is around 240°C higher than the temperature used in the blend (160°C), so the crystalline portion of PET remains in solid and hard form. The softened/melted portion may improve the bond between aggregate and bitumen, while the hard crystalline portion imparts rigidity.

Air voids

The percentage of air voids is the volumetric factor required to check the susceptibility of bituminous mixes to rutting, cracking, and even aging. Excessive voids cause cracking because the bitumen does not sufficiently cover the aggregate, while low voids can cause permanent deformation (rutting).

The results of the air void percentages are shown in Figure 11; as the PET content of the Coarse-PET HMA increases, the air void content also increases, reaching 4.94% at 7%. This indicates that the addition of coarse PET to the blend reduces processability and leads to an increase in void content.

Nevertheless, all the HMA modified with coarser PET are within the acceptable air void range of 3 to 5% (Jegatheesan et al., 2020; Ferreira et al., 2022).

A fine PET size generally gives air void values below the specified range except at 5%, where the percentage reached 3.25%, corresponding to a comparatively lower bulk density obtained with this size.

Coarse polyethylene terephthalate particles generally exhibit higher bulk density than finer particles, regardless of PET content. The increased surface area that needs to be coated with

bitumen can elucidate the phenomenon. When PET particles are smaller, the bitumen is required to cover a larger surface area. Consequently, this can lead to less workability during the mixing process, resulting in lower bulk.

Modifying the HMA with a coarser PET size (2.50-1.25mm) generally obtained better performance regarding the volumetric and Marshall characteristics.

3.2.Creep recovery test

Figure 12 shows the creep-recovery curves of the control and PET-HMA by 3, 5, and 7% and two dimensions of PET waste at high temperature (50 °C).

On creep:

The measured deformations increase over time, reaching a value $\epsilon_{\text{Max}} = 4.98\%$ to the control HMA, more than 133% and 125% compared to the deformation values of the modified bituminous mixes at 5%Coarse-PET HMA and 5%Fine-PET HMA, respectively. The C-HMA exhibits increased deformability due to the temperature sensitivity of bitumen. More generally, these materials are thermos-sensitive, i.e., their viscoelastic behavior changes according to the temperature (Mashaan et al., 2021; Karimi et al., 2017). On the other hand, initial deformations have become more important. According to the maximum deformation, the rate of this deformation is 29% for the C-HMA and reaches up to 35% at 7% Fine-PET HMA. So, almost half of the deformations are reached in the first phase (elastic behavior), which confirms the results of the study's results by several researchers (Haddadi et al., 2008). Then, during the first minutes of loading (between $t = 15$ s and $t = 600$ s), the bitumen at high temperatures softens and promotes inter-granular sliding, the bond between the bitumen and the aggregates weakens, and consequently, which changes the rate of deformation (Merbouh., 2012; Ameri et al., 2020). Finally, in the third phase of creep (viscous flow), the slope of the linear line becomes more significant from $t = 600$ s to $t = 3600$ s.

Figure 12 shows a remarkable decrease in the measured strains of PET-HMA compared to C-HMA,

especially at 5% of coarse and fine PET. The adhesion between the mixtures containing lower waste PET contents (3% and 5%) and the bitumen may cause a reduction in creep. An increase in creep resistance is also observed when the PET content is higher (7%) because less rigid particles replace the aggregates. Therefore, the reduction in bitumen content around the aggregate particles could be another reason for the reduction in stiffness (Modarres and Hamedi., 2014; Chen et al., 2020). The deformation evolves slowly over time ($600 \text{ s} < t < 3600 \text{ s}$) for the PET-HMA compared to the C-HMA. The deformability of the modified mix is influenced by the presence of bitumen and the PET additive; this is reflected by the decrease in the slope of the regression line in the viscous flow phase (Ghabchi et al., 2021). After one hour, the maximum deformation of the bituminous mixtures of 3% Coarse-PET HMA, 5%Coarse-PET HMA, and 5%Fine-PET HMA decreased by 17%, 25%, and 20%, respectively, following the modification. Which subsequently produces a good resistance to permanent deformation (Bekhedda and Merbouh, 2022). The incorporation of PET during mixing enhances the material's stiffness while simultaneously reducing its thermal susceptibility to high service temperatures, resulting in reduced sensitivity to creep deformations (Ahmadinia et al., 2011; Ahmadinia et al. 2012; Moghaddam et al., 2013).

On recovery:

The reversible deformation of the C-HMA is not too significant ($\epsilon_{ER} = 0.41\%$), the adhesion between the bitumen and the aggregates decreases, and the bonding forces weaken; therefore, the residual deformations are increasingly important at high temperatures ($\epsilon_{Res} = 4.57\%$). In this case, the C-HMA has a visco-plastic behavior and its deformability increases. The accumulation of these permanent deformations can create the risk of rutting the roadway with the loading/unloading cycles. A clear improvement in the phase of reversible deformations is also recorded for the PET-HMA. This recovery was improved by 29% at 5%Coarse-PET HMA, and therefore, the modification by PET reduces the permanent deformations at the end of the test. The PET helps the

bituminous mixture to recover its total deformation; hence, a slight slope ($3720 \text{ s} < t < 7200 \text{ s}$) in the recovery curve for the PET-HMA is noted (Ameri et al., 2020).

The following histogram (Figure 13) clearly shows the increase in deformations measured as a function of temperature at the different phases (initial, total, and residual). The PET-modified mixtures give better results for all PET percentages and dimensions, confirming this modification technique's advantage.

3.3. Rutting test

Rutting resistance is an important property that determines the performance of bituminous mixtures under the effect of temperature-stress coupling (Ma et al., 2018). From the results of the Marshall test (Marshall Quotient) and the results of the creep-recovery test, it is always deduced that the use of 5% of the PET content gives the best performance of the bituminous mixture (The optimum content) regardless of the dimension of this waste with a slight superiority or coarse over the fine, where it becomes more flexible and prevents deformations and dispersion in the mixtures due to the application of temperature. Several researchers have also confirmed this percentage (Ahmadinia et al., 2011; Ahmadinia et al. 2012).

Figure 14 shows the curves of the evolution of the percentage of rutting as a function of the number of stress cycles at high temperatures:-

Rutting resistance is quantified by the rut depth measured during the test; the greater the depth, the lower the resistance, and vice versa. The curve of C-HMA is higher than 4.22%, meaning that it is more deformable and less resistant to rutting. The origin of the ruts of the mixtures is often attributed to the binder (Esfandabad et al., 2020; Mashaan et al., 2021; Chakravarty et al., 2023). Bitumen is very sensitive to temperatures, especially at a high temperature such as that of the test ($60 \text{ }^\circ\text{C}$), from where it softens and presents an elastoplastic behavior (irreversible deformation in the initial state) (Merbouh., 2012; Li et al., 2021).

It is noted that all the PET-HMA have deformation rates lower than the C-HMA deformation. After analyzing of the different parameters of the plates of the seven samples, the instability of the C-HMA can initially be linked to a higher bitumen content than that of the PET-HMA, which 5% of the total bitumen mass.

It is also concluded that there is a significant decrease in total deformation of the order of 18%, 34%, and 26% at 3%Coarse-PET HMA, 5%Coarse-PET HMA, and 5%Fine-PET HMA, respectively, compared to C-HMA which implies a clear improvement in rutting resistance and a higher pavement life, which confirms the results of the study by several researchers (Ameri., 2020; Zhang et al., 2021; Hao and Xiaosen, 2024). On the other hand, at high PET contents (7%), this decrease in permanent deformation is very small in the order of 6% and 9% at 7% Fine-PET HMA and 7% Coarse-PET HMA, respectively. When PET merges into the mixture, it plays the role of binder; it increases the adhesion and bonding between the bitumen and the aggregates. In the molten state, it fills the maximum of voids in the granular skeleton of the asphalt. On the one hand, a good part of the PET in the modified asphalt mixture, the particle size class (0.315-0.16mm), does not play the role of a filling element and does not participate well in the transmission of forces. On the other hand, it is quite different for fine PET particles. It should be noted that the great stability of the modified asphalt lies in the importance of the contacts between the large PET particles (2.50-1.25mm) and the different fractions of the aggregates. Which subsequently produces good resistance to permanent deformation (Modarres and Hamedi., 2014; Mashaan et al., 2021). The addition of PET decreases the deformations measured for high temperatures, improving rigidity by decreasing the thermal susceptibility or blocking the effect inside the material by increasing the consistency.

3.4.Index of Retained Strength Test

Figure 15 shows the compression results for dry and wet samples (an average of three cylindrical

samples).

The results show that the S2 values (compressive strength of immersed specimens) of the HMA modified with waste PET are higher than the control HMA for both sizes and all three PET contents. A peak is observed at 5% PET content, with 71 and 53% increases for Coarse-PET HMA and Fine-PET HMA, respectively. This indicates that the modified HMA can withstand higher compressive strengths before cracking occurs. The results also show significant differences in conditioned S2 values when adding two different PET sizes. Regarding the effect of water on mechanical properties, all materials, with or without PET, exceeded the current Algerian specifications ($MS_0 > 80\%$), which means a clear improvement in resistance to moisture sensitivity. These good results were obtained for all the modified HMA with PET waste, which performs better than the control HMA.

The results of the (MS_0) ratio are shown in Figure 16.

Using a coarser PET yields superior outcomes compared to the control mixture. However, all PET-modified mixtures meet the minimal requirement of 80% MS_0 . Additionally, an asphalt mixture modified with coarse PET waste is given the best resistance to MS_0 water sensitivity compared to fine PET at different waste contents.

The water resistance of the control HMA improved after modification with 5% plastic waste, 19% coarse PET, and 12% fine PET. On the other hand, the HMA modified with 7% PET showed the least improvement (only 3% compared to the control hot mix asphalt).

These results show that the modifiers play a more structural role in the asphalt mix due to their thermal behavior at 60°C (considered a high service temperature), and their size determines their influence. The improved resistance can be explained by the thermal behavior of polyethylene terephthalate, which gives better resistance with a longer curing time (Taherkhani and Arshadi, 2019; Jegatheesan et al., 2020; Ghabchi et al., 2021).

The addition of waste PET improves the adhesion between the bitumen and the aggregates, which leads to a reduction in the damage caused by moisture in the HMA and horizontal deformation (Shahin., 2015; Singh et al., 2017; Yan et al., 2020), and increases the stiffness value of the tensile modulus, thus solving the problem of the asphalt's sensitivity to water, which is in agreement with (Azam et al., 2019; Li., 2021; Ferreira et al.,2022).

3.5.The leaching test

Figure 17 shows the results of the chemical analyses of the water taken during the 0, 3, and 7-day tests for chlorides (CL^-), sulfates (SO_4^{-2}), pH, and the chemical analysis of the water faucet from Bechar City.

Figure 17 shows that the control and modified bituminous concrete can cause chloride (CL^-) diffusion when in contact with water. In each case, the HMA modified leached more chloride than the control mix after 3 or 7 days of immersion. Secondly, the chloride content of the water leached from the hot mix asphalt modified with fine PET showed much higher results than the Coarse-PET HMA with a maximum value equal to 303 mg/l at 7%, more than 151% higher than the C-HMA, but still acceptable according to the minimum threshold of the Algerian specifications (≤ 500 mg/l) (Journal Officiel de la République Algérienne, 2011).

When the control and modified bituminous mixes come into contact with water, the sulfate (SO_4^{-2}) content remains constant compared to the original water initially (after 3 days of immersion). However, over time (after 7 days of immersion) and after increasing the PET content for both sizes, sulfate leaches out in remarkable quantities, with a maximum value of 218 mg/l for 7% Fine-PET HMA more than 267% higher than for asphalt concrete without the addition of PET, but still below the Algerian specification (≤ 400 mg/l) (Journal Officiel de la République Algérienne, 2011); As a result, polyethylene terephthalate is less polluting for the soil, surface, and groundwater.

At the test's beginning, the original water's pH was 7.62. On the other hand, the pH values in the

water containing bituminous samples are higher than in this water. An increase in the pH of the solutions containing the PET-modified mix can already be observed, with a range of 8 to 9.35, while the pH of the solution containing the control mix coating remains constant. In parallel, the conductivity increases slightly in the case of asphalt modified with coarse PET and more so in the case of fine PET, especially at a content of 7%. However, they still comply with Algerian specifications ($6.5 \leq \text{pH} \leq 9.5$) (Journal Officiel de la République Algérienne, 2011). The increase in conductivity is explained by the release of ions and the increase in ion concentration in the solution. Leaching tests show that the asphalt mix leaches toxic substances when it is exposed to water but remains mildly aggressive to nature, which is acceptable according to the Algerian standard (Journal Officiel de la République Algérienne, 2011) and the World Health Organization (WHO) standard (World Health Organization, 2017). Therefore, PET does not present any risk of aggressiveness at ambient temperature.

4. Conclusion

In this experimental study, an attempt was made to evaluate the effects of adding PET waste to bituminous mixes of two different sizes (coarse and fine) with contents varying from 3, 5, and 7% of the total weight of bitumen; these modified and unmodified mixes were subjected to various tests, which gave different results. The conclusions are as follows:

- The process of introducing plastic waste directly into the mixer offers us an easy, fast, and effective technical solution for modifying asphalt concrete;
- Based on the Marshall test results, it was concluded that incorporating PET plastic waste in the three percentages and both sizes by the dry process to obtain HMA modification improved its mechanical properties, with a slight advantage of coarse over fine. Significantly increasing its stability and resistance to permanent deformation; this improvement is best seen at 5% PET content;

- At high temperatures, creep deformation evolves slowly for the modified asphalt mixture. The decrease in deformations is 25% and 20% at 5%Coarse-PET HMA and 5%Fine-PET HMA, respectively. PET decreases the residual deformations at the end of the test;
- The resistance to permanent deformation was improved by around 34% and 26% at 5%Coarse-PET HMA and 5%Fine-PET HMA, respectively, thus reducing the risk of rutting, which led to an increase in the pavement's life;
- The addition of PET decreased all strains measured at high temperatures, leading to a marked improvement in creep and rutting resistance and thermal susceptibility;
- The evaluation of water sensitivity by the immersion compression test shows a significant increase of 3 to 19% of the asphalt modified with waste PET compared to the C-HMA. Therefore, the use of waste PET plastic improves the resistance to moisture sensitivity of bituminous mixes;
- The 5% waste PET modified asphalt mix performed very well in water, regardless of size (95% for 5%Coarse-PET and 92% for 5%Fine-PET), while other combinations of 3 and 7% in Coarse-PET HMA and Fine-PET HMA met specifications but with lower values.
- The results of placing samples of HMA modified in water have showed that PET shown that PET directly affects leaching. It has been shown that the size and content of PET particles have very good linear relationships (strong correlation) with CL^- and SO_4^{-2} leaching concentrations and pH but are still within specifications.

The results conclude that ground that ground polyethene terephthalate can be used as an additive in bituminous concrete. This addition made by the dry process, can improve the mechanical properties of bituminous mixes. Consequently, this study not only explores the effective use of non-degradable plastic waste but also provides an opportunity to improve roads and make them more sustainable.

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List of tables

Table 1. Bitumen Grade 40/50 Characteristics.

Designation	Standard reference	Results	Range of
Needle penetration in 1/10 mm at 25°C	EN 1426	47	40-50
Softening point (°C)	EN 1427	56	52-57
Row 3 Specific gravity at 25°C	EN 15326	1.03	1.03-1.10
Ductility at 25°C (cm)	EN 13589	79	≥60
Resistance to hardening at 163°C		-	-
Retained penetration (%)	EN 12607-1	57	≥53
Increase in Softening point (°C)	(RTFOT method)	6	≤8
Change of mass (absolute value) (%)		0.3	≤0.5
Flash point (°C)	EN 22592	245	≥240
Solubility (%) (mm/mm)	EN 12592	99.50	≥99

Table 2. PET plastic waste characteristics.

Characteristics	PET
High melting point (°C)	235
Fusion time (min)	13
Glass transition temperature (°C)	70
Specific gravity (g/cm ³)	1.35

Table 3. Aggregate characteristics.

Characteristics	Aggregate			Requirement	Reference standard
	0/3	3/8	8/15		
Cleanliness (%)	/	0.98	0.45	<2	NF P 18-591
Flatness (%)	/	18	14	<25	NF P 18-561
Bulk density (g/cm ³)	1.57	1.37	1.36	/	NF P 18-554
Reel density (g/cm ³)	2.60	2.61	2.62	/	NF P 18-555
Los Angeles (%)	/		20	≤25	NF P 18-573
Micro-Deval (%)	/		18	≤20	NF P 18-572
Sand equivalent (%)	52	/	/	≥35	NF P 18-598
Carbonates (CaCO ₃)	75	82	85	/	NF EN 1744-1
Sulfates (SO ₄ ²⁻)		No trace		/	NF EN 1744-1
Modulus of sand fineness	2.92	/	/	2.40 ≤ Mf ≤ 3.10	NF P 18-540
Methylene blue value Vbs (%)	0.75	/	/	≤2	NF P94-068

List of figures



(a)



(b)

Fig. 1. PET sizes used: (a) fine size (0.315-0.16mm); (b) coarse size (2.50-1.25mm).

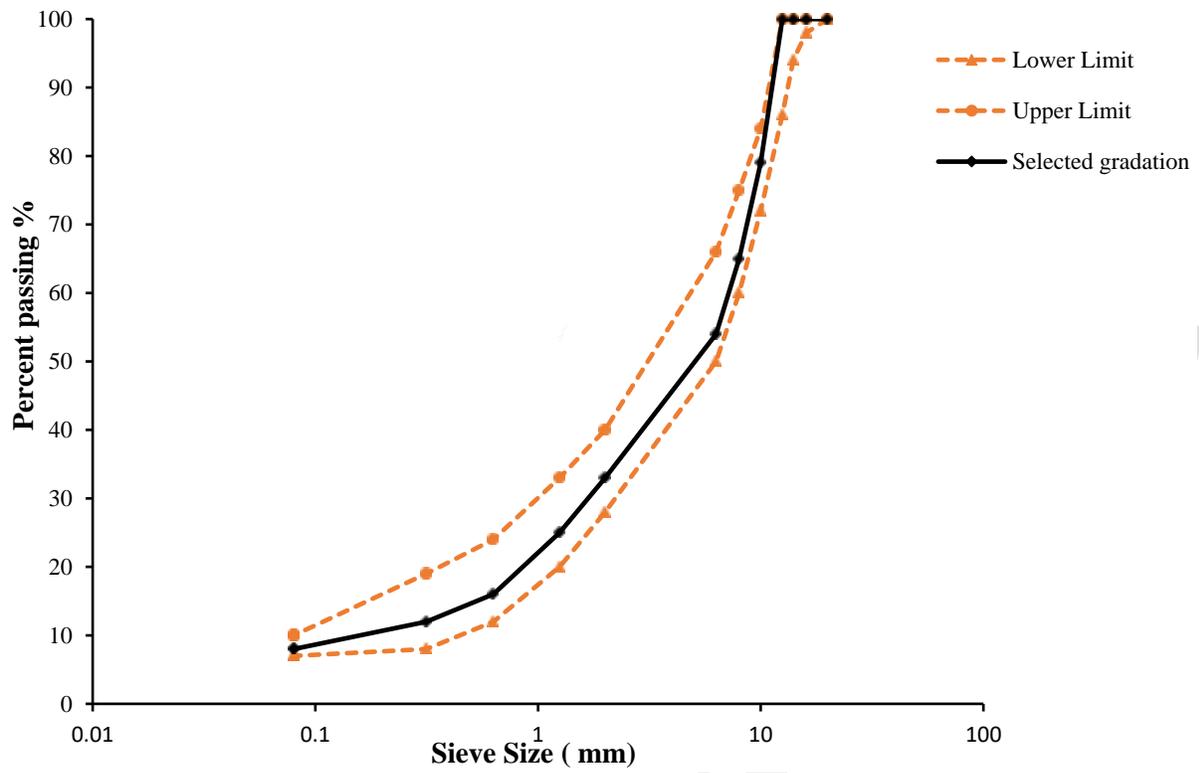


Fig. 2. Grading curves for the spindle and bituminous mixture (0/14).



Fig. 3 Bituminous mixtures plates.



Fig. 4. Prismatic specimens in control and modified bituminous concrete.



Fig. 5. Four-point creep-recovery test devices.



Fig. 6. Rutting test device.



Fig. 7. The leaching test device.

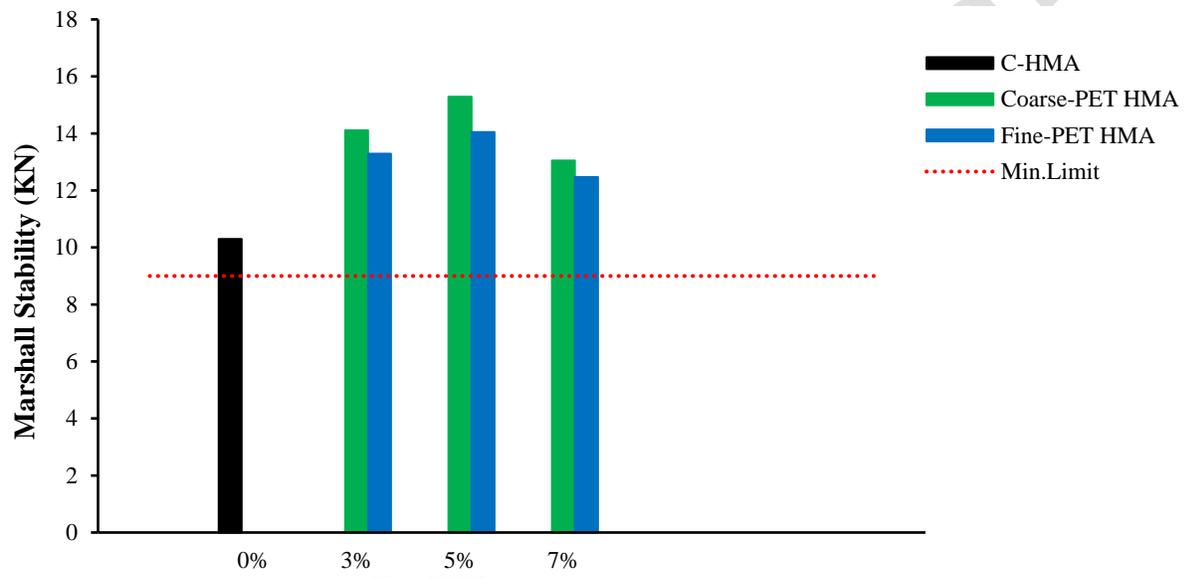


Fig. 8. Marshall stability (MS) results.

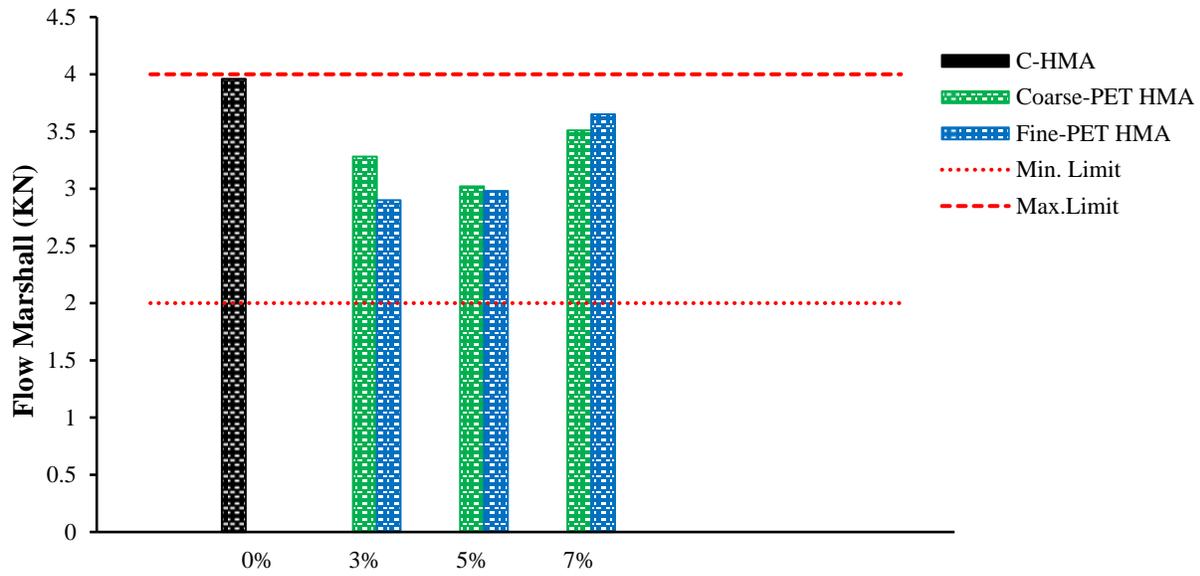


Fig. 9. Marshall Flow (MF) results.

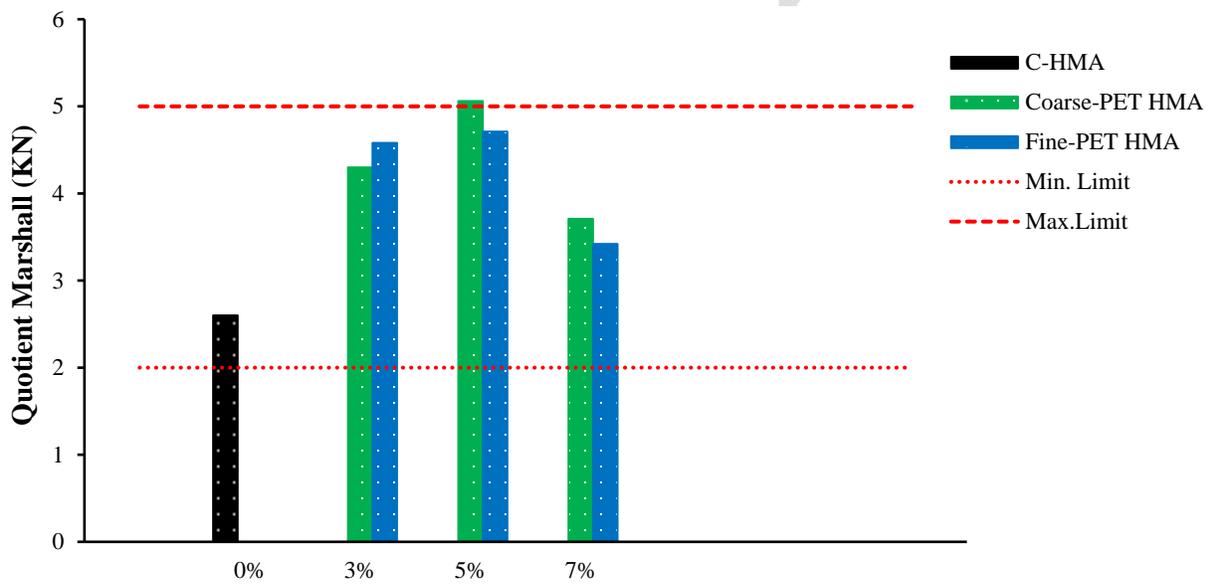


Fig. 10. Marshall Quotient (MQ) results.

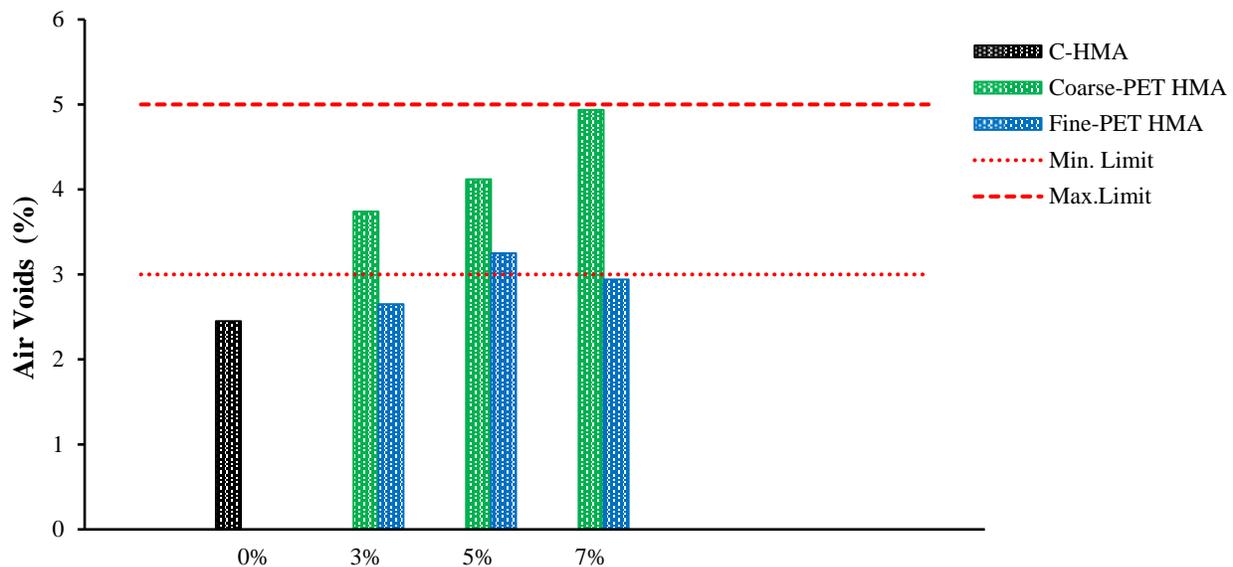


Fig. 11. Air voids results.

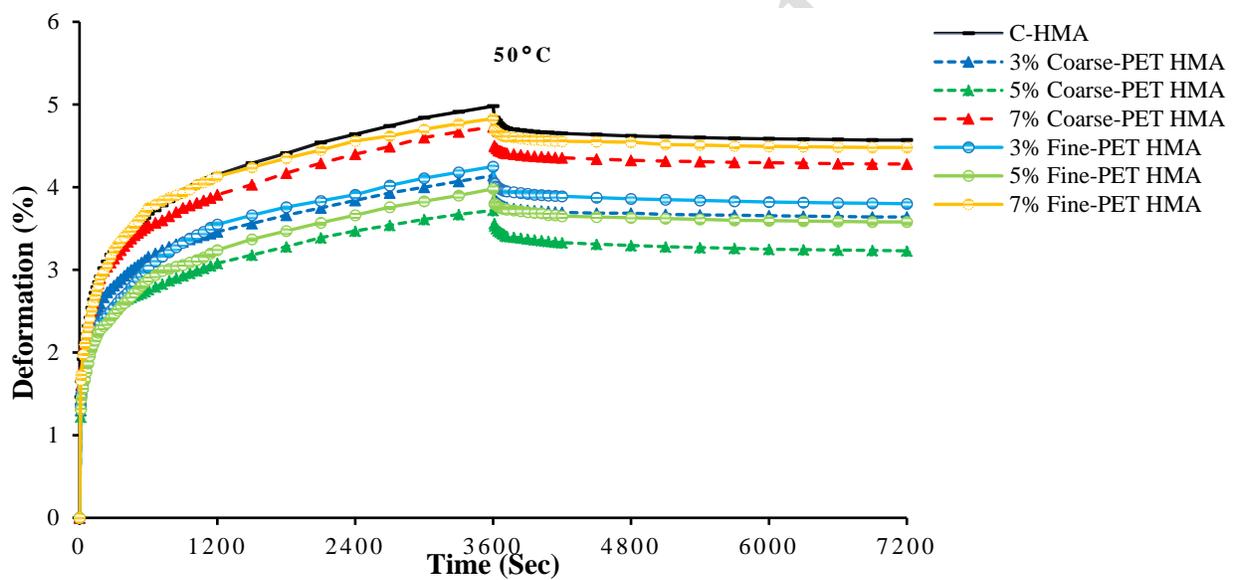


Fig. 12. Effect of PET content on creep recovery of different mixtures at 50°C.

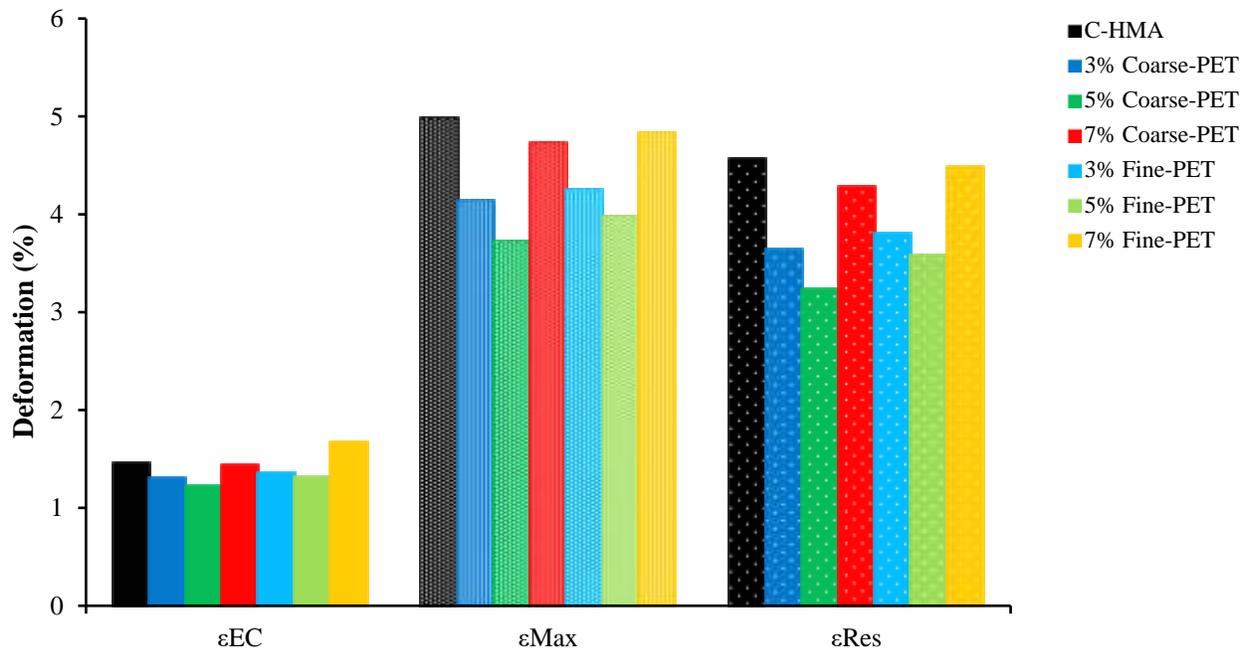


Fig. 13. Comparison of measured deformations between PET-HMA and C-HMA at high temperatures.

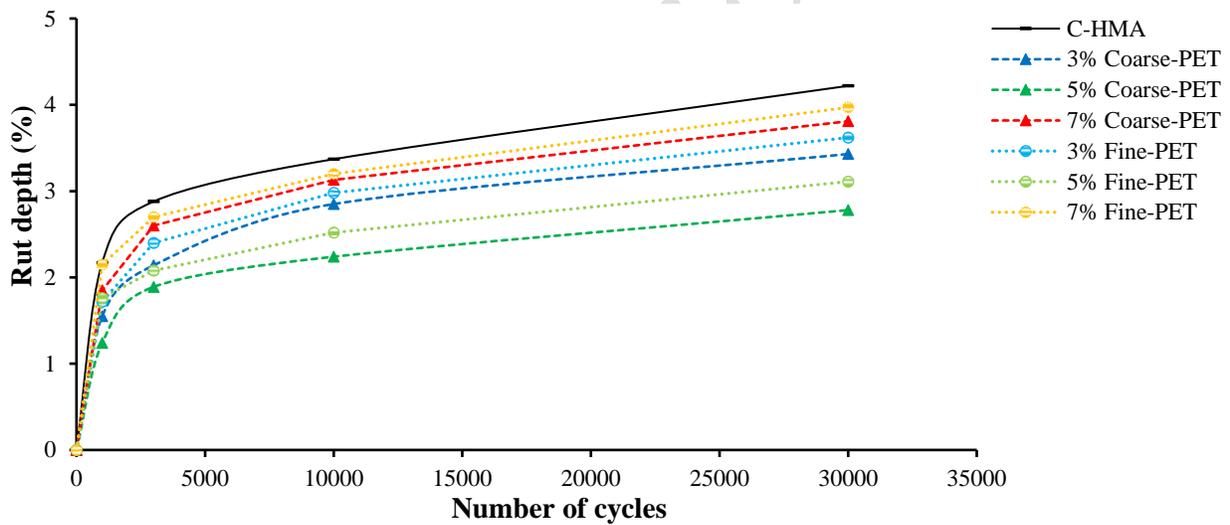


Fig. 14. The rutting depth of different asphalt mixes depends on the number of cycles.

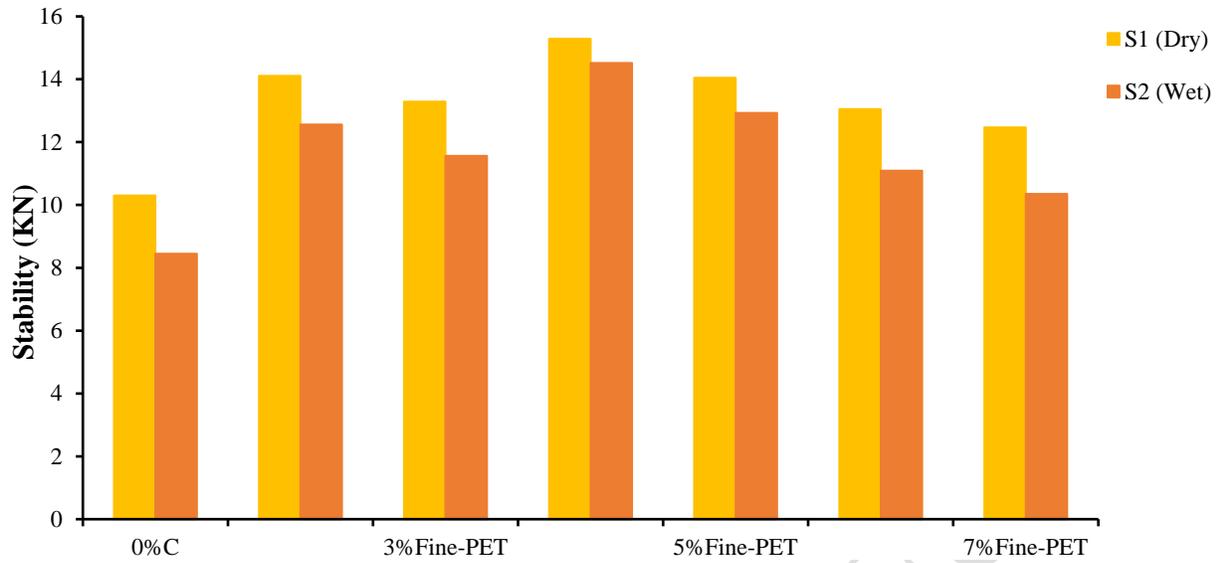


Fig. 15. Results of compressive strength test of dry and immersed specimens.

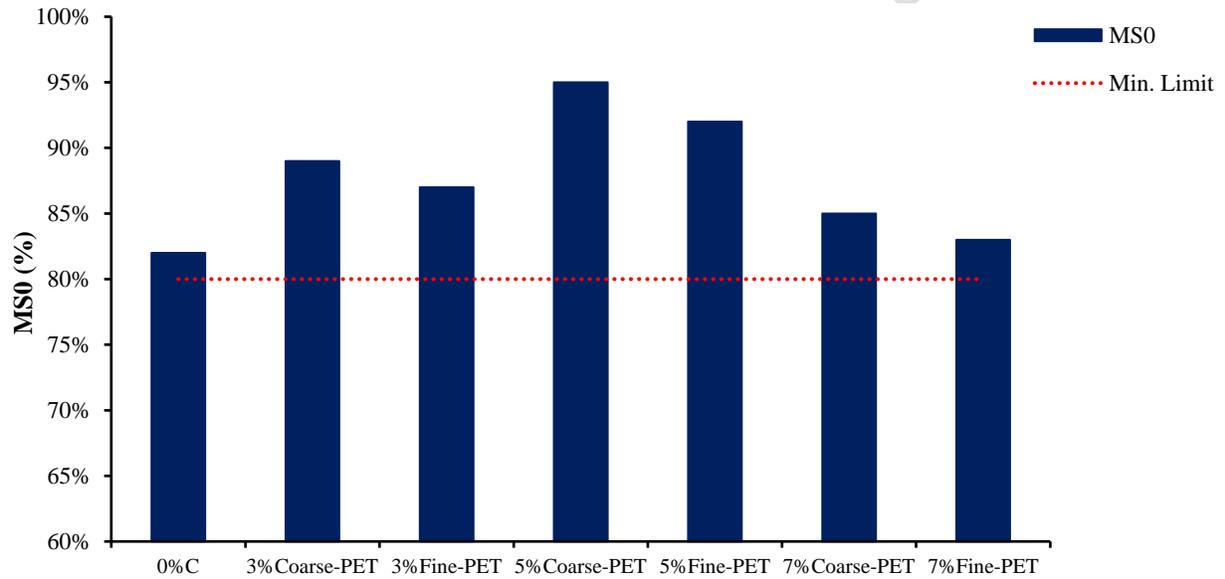


Fig. 16. Results of MS0 test.

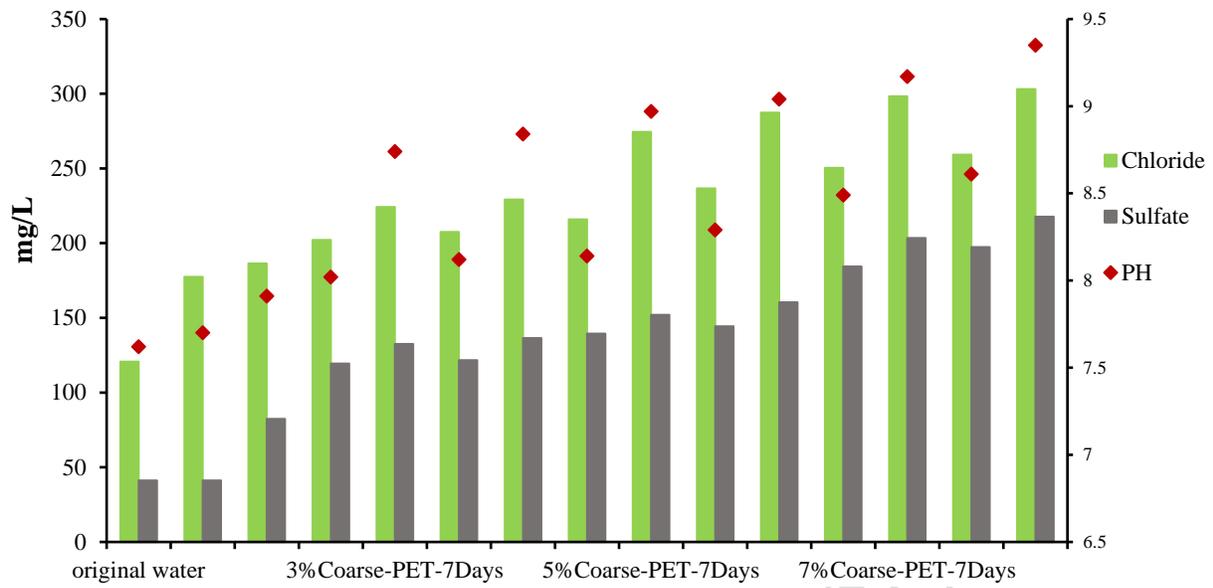


Fig. 17. Results of 0, 3, and 7-day leach tests on tests on C-HMA, Coarse-PET HMA and Fine-PET HMA samples.

Accepted / Not