Visual Abstract

Recycling of plastic waste polyethylene terephthalate (PET) as a modifier in asphalt mixture: study of Creep-Recovery at Low, Medium, and Hot Temperatures

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ABSTRACT. One environmental problem in most countries is the huge quantities of plastic waste, which has different types of polymers such as polyethylene terephthalate (PET), polypropylene (PP)., These plastics become a problem because of their non-biodegradability, toxicity, etc. The recovery of plastic waste in pavements has become an ecological technique that contributes to the preservation of natural resources.
In this study, PET waste was used as an additive for hot mix asphalt at two sizes (2.50 to 1.25 mm, and 0.315 to 0.160 mm) and three contents (3%, 5%, and 7% by weight of binder), PET modified mixes creep-recovery behavior was tested by four points bending test and compared with control mix (without PET waste) at three different temperatures (0°, 25° and 50°C), were added directly to the mixture as the method of dry process.
The results showed that the addition of PET waste yields better results that which could increase the asphalt bituminous resistance against permanent deformation and a clear rigidity improvement in the various temperature because it's more thermally stable and can limit the degradation risks of the bituminous mixtures compared to the control mixture. Further, can be prepared an eco-friendly road construction with less material cost.

KEYWORDS. Plastic waste; Polyethylene terephthalate (PET); Hot mix asphalt; Creep-recovery; Temperature; Permanent deformation.

OPEN ACCESS

Citation: Bekhedda, A., Merbouh, M., Recycling of plastic waste polyethylene terephthalate (PET) as a modifier in asphalt mixture: study of Creep-Recovery at Low, Medium, and Hot Temperatures, Frattura ed Integrità Strutturale, 60 (2022) 438-450.

Received: 25.10.2021
Accepted: 17.03.2022
Online first: 20.03.2022
Published: 01.04.2022

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INTRODUCTION

Due the population growth rate and the rise in living standards, an increase of the solid waste amount generated worldwide. In Algeria alone, plastics represent 15.31% of the total weight of about 2.1 million tons per year of the generated solid waste (PET which represents approximately 3.57%) [1].

With the plastics' use increasing in various industries, waste polymers discharges will inevitably create several environmental problems in the coming years. The attempts of a variety of organizations and a number of researchers to find a useful application of some of this waste in the construction of roads [2, 3, 4].

Every day, new roads are built in different countries using million tons of raw materials and natural stocks. This enormous consumption causes a daily loss of these materials. At the same time, most developed countries and developing countries are faced with serious problems arising from waste disposal.

Much research has been carried out to improve the blends design, optimize the use of materials and improve the efficiency of construction methods. The bituminous coating constituting material properties have a significant influence on road surface life. An essential property of unmodified or modified bitumen is its ability to adhere properly to an aggregate surface.

Today, one of the main projects using plastic waste has been developed by the Thiagarajar College of Engineering, Madurai, at New Delhi, India, where the use of modified asphalt with a plastic waste mixture by dry process (polyethylene, PE, polypropylene, PP, and polystyrene, PS, has been applied rural roads. The mixture has improved the normal conventional blends' performance by avoiding cracking and rutting [5]. Normally, plastic waste is used independently because of the different properties that the multitude of polymers in the market may possess on one hand, and to better control the resulting mixture on the other hand.

Similar projects with low-density polyethylene (LDPE) showed an increase in indirect tensile strength and modulus of elasticity [6]. As well as resistance to plastic deformation and fatigue [5]. The PP virgin fibers blended with natural aggregates were also used before the addition of the bitumen (dry process) whereby the results showed an increase in the Marshall mixture's stability and the fatigue resistance [7]. However, in a study with PP from plastic waste, it was concluded that its incorporation improves the plastic deformation's resistance, but it had no influence on its resistance to fatigue [8].

The use of PET waste as an aggregate replacement in asphalt concrete was first investigated by Hassani et al. [9] in which they replaced the 2.36 – 4.75 mm natural aggregates with 3 mm diameter PET. Using Marshall's stability and flow, MQ, the compacted mixtures' specific gravity was evaluated as the properties of the mechanical mixture. This study proved that the replacement of 20% of natural aggregates' volume with PET granules made the plastiphalt proper for practical use.

Ahmadinia et al. [10] (2011) used 1.18 mm waste PET as an additive in SMA mixtures in the dry process. The re-using of waste PET had a positive effect on the Marshall and volumetric SMA mixture properties in an environmentally and economical way.

Ahmadinia et al. [11] were investigating the PET modified SMA mixtures performance on the Wheel tracking, moisture susceptibility, resilient modulus and drain down tests. The results showed that the 4% and 6% of asphalt content weight are the appropriate ranges to satisfy the standard requirements for PET modified mixture's performance. Another study evaluates the PET modified asphalt mixtures fatigue properties which proved that 2.36 mm maximum size were assessed. The use of a high PET amount in mixtures showed a higher resistance against fatigue cracking, and the rate fatigue life increased less than the rate of increase of the intensity of the dynamic loading [12].

The application of PET modification under dynamic loading at various temperature and stress decrease the permanent strain. In today's world, polymer-modified asphalt is commonly used in road building [13, 14]. As a result, before commercializing polymer modifiers, the cost benefits of various methods analysis are essential for making projects more practical and cost-effective [14, 15].

The objective of this study is to verify if the addition of waste plastic PET at both sizes and three contents in the bituminous mixture can be a viable alternative to improve the asphalt performance at different service temperatures and an environment-friendly green pavement can be prepared with less material coast.

MATERIALS

This research used various materials including aggregate, bitumen and waste PET plastic.

crushed sand (S: 0/3), aggregates of class (G1:3/8 and G2:8/15) and filler come from the quarries of Boucheta (Djebal Bechar), these materials are calcareous CaCO3. After several identification tests, for experiments on the mixtures, these materials have good intrinsic qualities like the value of Los Angeles is equal to 20%, shall not be more than 25 %, the same for Micro-Deval is equal 17%, shall not be more than 20%, so the aggregates are hard and resist wear. The results are listed in Tab. 1.

A 40/50 penetration grade bitumen was obtained from EPTP Bechar which was used to prepare asphalt mixture. Tab. 2, present the characteristics of bitumen used in the study.

The mix curve was selected between the upper and lower limit with a maximum aggregate size of 14 mm, as presented in Fig. 1.

Characteristics	Reference standard	Requirement	Sand (0/3)	Gravel (3/8)	Gravel (8/15)
Bulk density (g/cm^3)	-	-	1.68	1.50	1.55
Reel density (g/cm^3)	-	-	2.52	2.61	2.61
Flatness (%)	NF EN 933-9	≤20%	/	14	9
Los Angeles (%)	NF P 18-573	≤25%	/	20	20
Micro-Deval (%)	NF P 18-572	≤20%	/	17	17
Sand equivalent (%)	NF P 18-597	≥ 50%	56	/	/
Methylene blue value Vbs (%)	NF P 18-561	≤2	1.5	/	/

Table 1: Characteristics of aggregates.

Description	Reference standard	Requirement	Value
Needle penetration in 1/10 mm at 25°C.	NF T66-004	40-50	49
Softening point (°C)	NF T66-008	46-54	51
Relative density at 25°C	NF T66-007	1.00-1.10	1.03

Table 2: Characteristics of pure bitumen.

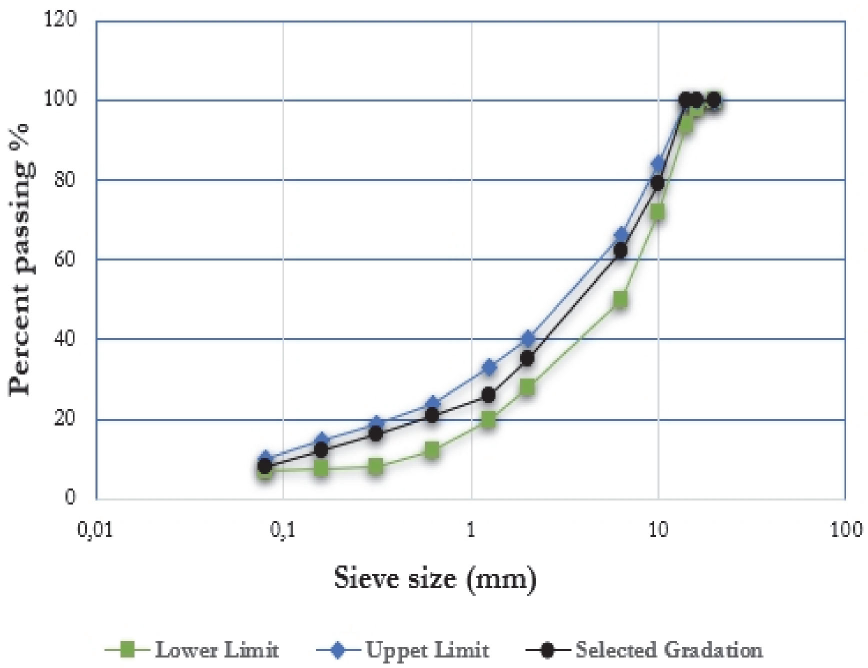
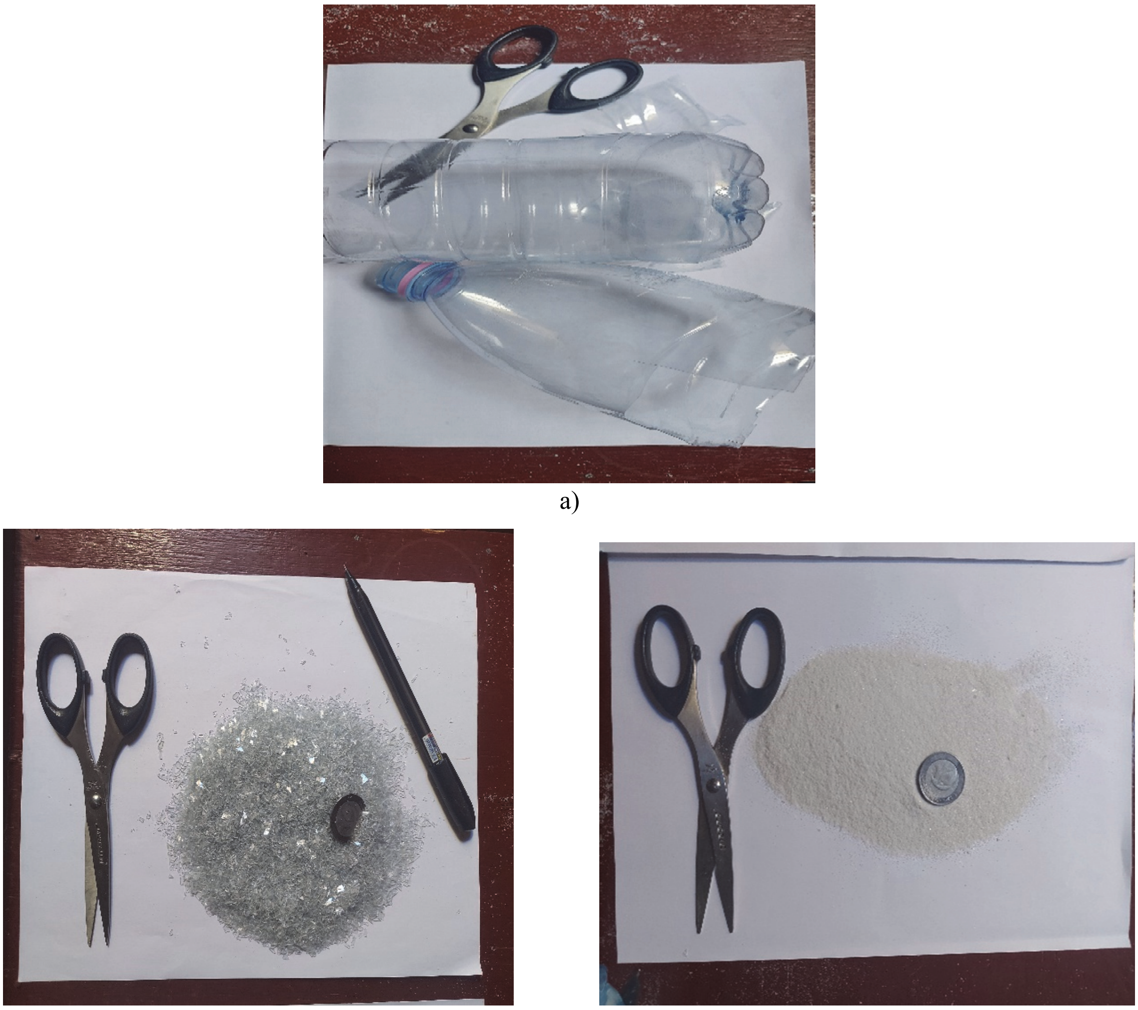


Figure 1: Aggregate gradation curves for hot mix asphalt.

The plastic waste polyethylene terephthalate 'PET' obtained from PET bottles has a high melting point of about 240-250°C with a fusion time is approximately 13 minutes and specific gravity was found to be 1.35 gr/cm^3. To use of waste PET bottles as an additive in asphalt mixture, they must be washed first, dried, and cut in flaky. Two main sizes of waste PET were used in this study: passing the 2.50 mm sieve and retained on 1.25 mm sieve (It is symbolized by 'coarse' waste PET size), and passing the 0.315 mm sieve and retained on 0.16 mm sieve (It is symbolized by 'fine' waste PET size), and added in different percentage (3%, 5% and 7% by weight of bitumen) to the mixture.



a)

b) c)

Figure 2: Waste PET sizes used; a) PET bottles, b) Size 1 (2.50–1.25 mm), c) Size 2 (0.315–0.16 mm)

SAMPLE PREPARATIONS

To prepare modified asphalt mixtures, there are mainly two methods: wet and dry processes, the first one depends on mixing plastic with bitumen to get a PMB (Plastic modified bitumen), after that adding it to the mixture. But due to the high melting point, it had been impossible to mix the waste PET particles with bitumen during a wet process to achieve a homogenous mixture.

The second one (dry process), according to the additive's type and nature of this material is mixed with aggregates before adding bitumen or added after mixing the bitumen and aggregates as a part of solid materials. In this study, the dry process was novelty special use, heated aggregates were mixed and coated with bitumen and waste PET at the same time to obtain the waste PET modified asphalt mixture. The aggregate was heated up to 175°C for 3 hours, the bitumen was heated up to 150°C for 1 hour; Finally, the aggregate, sand, filler, bitumen, and waste PET were mixed at 160°C for 5 minutes. Waste PET particles were blended with two sizes and three percentages by weight of bitumen for each mixture. These test conditions were selected based on the [16-17]. According to the Marshall test, the optimum bitumen content for the unmodified mix was equal to 5.23% [18].

Plate compactor size 400*300 mm has been prepared by a static compactor with a self-propelled mechanical cylinder with forwarding/reverse (Fig. 3), the compaction was determined in accordance to EN 12697-33. According to the creep recovery test conditions to be achieved, these slabs have been cut into several prismatic specimens 200*50*50 mm (Fig. 4).

Figure 3: Plate compactor.

Figure 4: Prismatic test piece (200×50×50 mm).

Creep-Recovery test

There are several types of devices used to perform the creep recovery test. Several different test methods have been developed which have included uni-axial tensile/compression, 2-point bending test, 3-point bending test, 4-point bending test, indirect tensile test [19].
The used device in this study is shown in Fig. 5 (Four bending deformations), which was developed in the FIMAS laboratory - UTMB Bechar-Algeria.

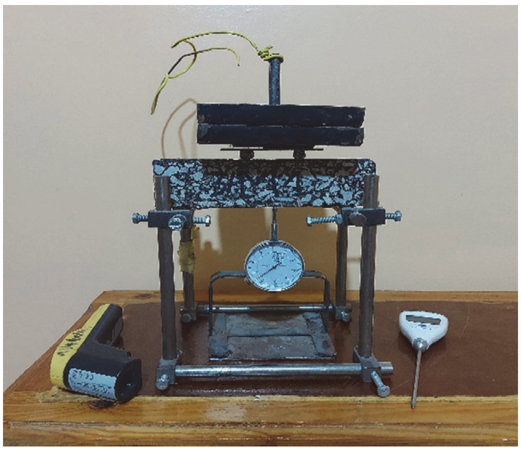


Figure 5: A device of Creep-recovery test.

The conducting procedure of a creep-recovery test is as follows:
-The sample is kept for 4 hours and placed in the device at the test temperature. Mechanical comparator (sensitivity ± 0.002) placed in the middle-lower face of the prism test piece allows measuring all deformation;

-A load of 4 kg shall be applied to the test piece for 1 hour from t = 0h, we record deformation evolution with constant stress (it's creep phenomenon), the applied stress is located in the linear elastic domain [20]. After a time, t_m = 1h, the applied stress σ_0 is removed, the recovery deformation is measured; A long time after removing the load, the deformation tends to a constant value, it is called permanent deformation.

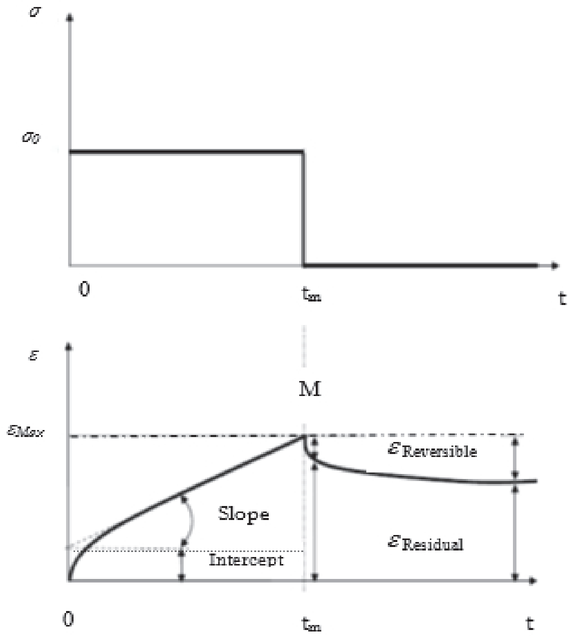


Figure 6: Creep-recovery experience.

Creep-recovery behavior

According to the graph above:
The creep part: evolves in 3 phases: elastic, visco-elastic behavior and viscous flow behavior respectively. The first phase represents the instantaneous deformation ε_{EC} due to the application of the load on the specimen, the second one is a parabolic curve with a significant slope in the primary minutes. The third phase is due to a flow dominated by the viscosity of the material. The slope of the curve is constant, but weaker than that of the previous phase until reaching the maximum strain ε_{Max}.
The recovery part: the prismatic test piece attempts to recover its initial form after the sudden load removal. This part evolves in 3 phases; the material takes an elastic behavior because of a reversible instantaneous deformation ε_{ER} recorded immediately after unloading. Then it continues to recover a part of its viscoelastic deformation (Irreversible deformation ε_{Rev}) so that the material presents a viscoelastic behavior. In third phase, the material can't longer recover its permanent or residual deformation recorded ε_{Res}. The appearance was due to the uncompleted recovery of prismatic deformation.
It has been noted:

- M1: Modified asphalt mixture (With 'coarse' waste PET size);
- M2: Modified asphalt mixture (With 'fine' waste PET size);
- O: Unmodified asphalt mixture (Without waste PET).

Results and discussion

Temperature Effect

Fig. 7, shows the effect of temperature on the deformation evolution of a modified asphalt mixture as a function of time on low (0°C), medium (25°C), and high temperature (50°C) at different waste PET contents and waste PET sizes, and comparing it with an unmodified asphalt mixture.

The deformation values are very small and almost equal to 10% at the low and medium temperature compared to that at high temperature. It's noticed that the effect rises in parallel with the deformation rise and thus the thermal gap becomes more influential, making it extremely difficult to compare these different temperatures on one curve, that's why we used logarithmic curves.
The measured deformations (initial, maximum and residual) are represented in Tab. 3.

Temperature	0°C			25°C			50°C		
waste PET content (%)	ε_{EC} (%)	ε_{Max} (%)	ε_{Res} (%)	ε_{EC} (%)	ε_{Max} (%)	ε_{Res} (%)	ε_{EC} (%)	ε_{Max} (%)	ε_{Res} (%)
0%O	0.230	0.254	0.150	0.08	0.416	0.312	1.36	4.76	4.37
3%M1	0.152	0.183	0.077	0.062	0.284	0.188	1.20	3.94	3.44
3%M2	0.177	0.193	0.088	0.064	0.304	0.200	1.26	4.07	3.56
5%M1	0.142	0.165	0.061	0.046	0.252	0.139	1.12	3.66	3.18
5%M2	0.151	0.171	0.071	0.038	0.271	0.152	1.21	3.81	3.36
7%M1	0.172	0.197	0.086	0.07	0.378	0.278	1.28	4.36	3.88
7%M2	0.188	0.210	0.105	0.064	0.360	0.252	1.27	4.47	3.98

Table 3: Measured deformations of Modified and unmodified asphalt mixture at 0°C, 25°C and 50°C.

Measured deformations evolve in time and in accordance with temperature rising; with significant deformations at high temperatures. At low temperature, the deformations are more rigid and have low deformations.
Fig. 7.a represents the unmodified asphalt mixture, the ordinary asphalt mixture's initial deformation at 0°C is equal to 0.23%, which is three times more than the initial deformation at an average temperature of the same mixture. At the low temperature, the bituminous mixes have an elastic behavior and become more rigid so that these deformations are very important. Because of this rigidity, the parabolic curve is not as significant as the behavior of the bituminous mixes at high temperature (50°C) because the bitumen becomes more rigid.
The maximum deformation's increase rate is about 6% and 72% of rang time between t = 60 sec and t = 3600 sec at T = 0°C and T = 25°C respectively. At a medium temperature, the asphalt mixture loses some of its rigidity, but remains resistant to creep.
In the recovery part, the asphalt mixture has an elastic behavior and it recovers a significant deformation part of 41% at 0°C temperature. At a medium temperature, the residual deformation is about 0.312% (75% of maximum deformation) which is more important and it has a viscoelastic behavior. At a high temperature, deformation increases, which reaches a value ε_{Max} = 4.76% more than 18 times and 11 times for the values of temperatures T = 0°C and T = 25°C respectively. The consistency and asphalt's cohesion decrease with the rise of temperature, otherwise the plastic deformation increases, which leads to a worse anti-high temperature of asphalt mixture properties [21, 22].
At high temperatures, the asphalt mixture becomes softer due to the bitumen viscoelastic behavior because of their sensitivity. The asphalt's temperature sensibility was directly related to the asphalt anti-rutting pavement ability at high temperatures [6, 23, 24].
50% of the creep deformation is achieved in the initial phase between t = 15 sec and t = 300 sec, the same results were obtained by Mazouz et al. [25], Haddadi et al. [26]. Thermal susceptibility refers to the change in mechanical properties of a material with a temperature change.
In the recovery part, reversible deformation is equal to ε_{ER} = 8%, which is not important because the bituminous mixes will be very sensitive due to the viscosity of bitumen at high temperatures, and that the permanent residual deformations are more and more important (ε_{Res} = 4.37%).
In Fig. 7.b and 7.c (Modified asphalt mixture), generally, have been noticed concerning the behavior and variation of the deformations measured as a function of time at different temperatures.
A decrease in the instantaneous elastic deformation values of the modified asphalt mixture for all temperatures and both sizes of plastic waste PET compared with unmodified asphalt mixture which we record up to 48% for T = 25°C at 5%M2. Compared with the unmodified asphalt mixture, the maximum deformation value of the modified asphalt mixture is reduced by 39% for T = 25°C at 5%M1.

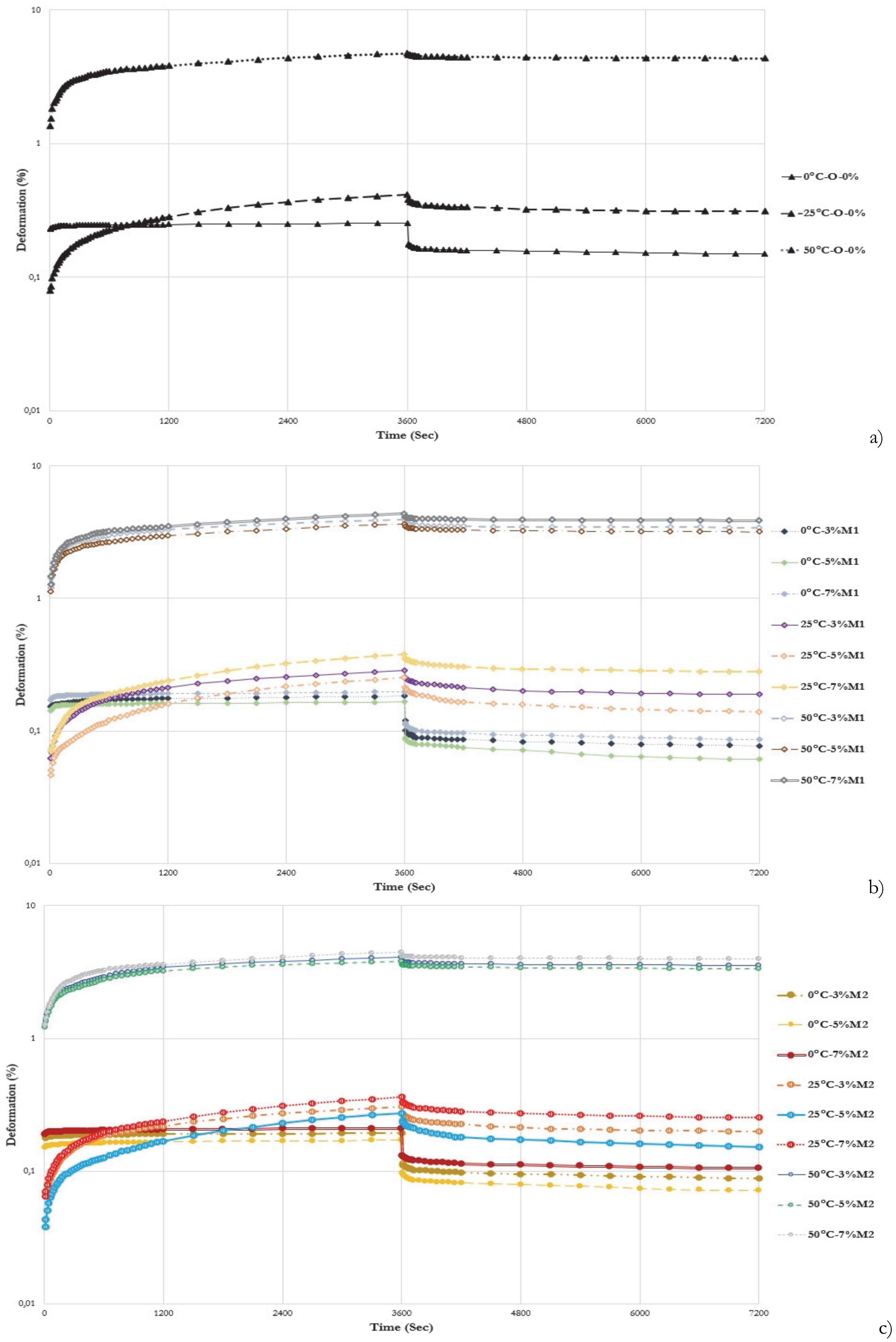
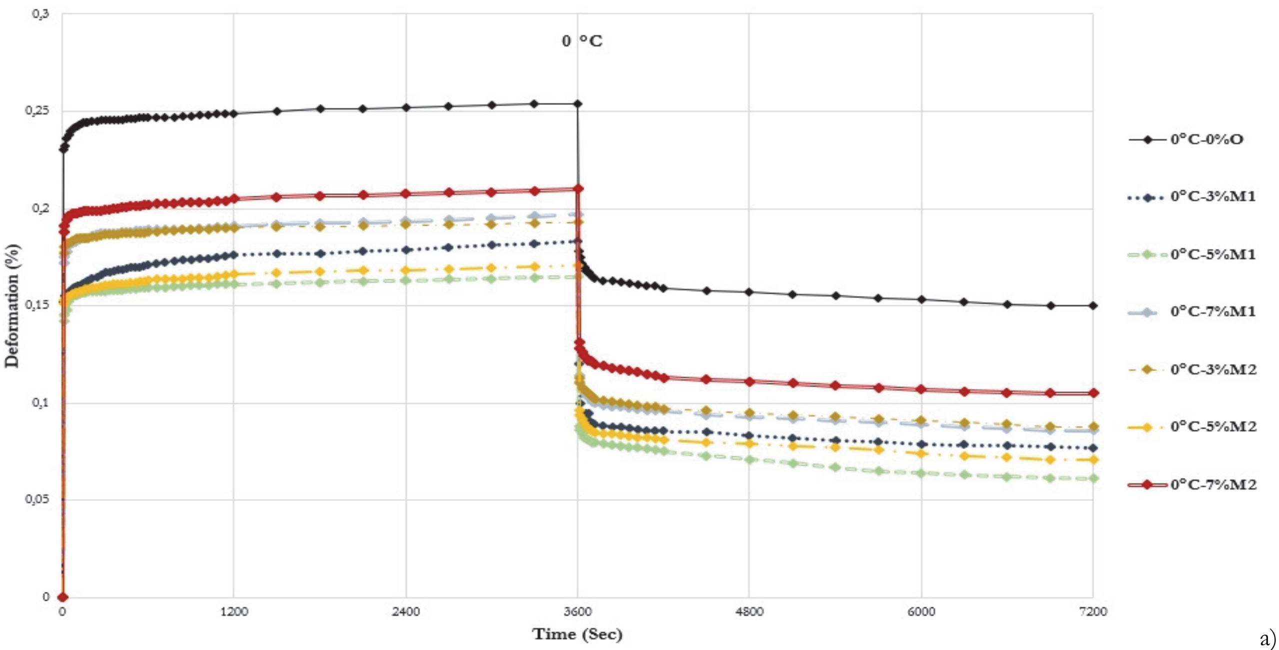


Figure 7: Effect of temperature on different hot mix asphalt; a) Ordinary Mix-Without waste PET, b) Modified Mix “M1”, by 3, 5 and 7% of waste PET, c) Modified Mix “M2”, by 3, 5 and 7% of waste PET.

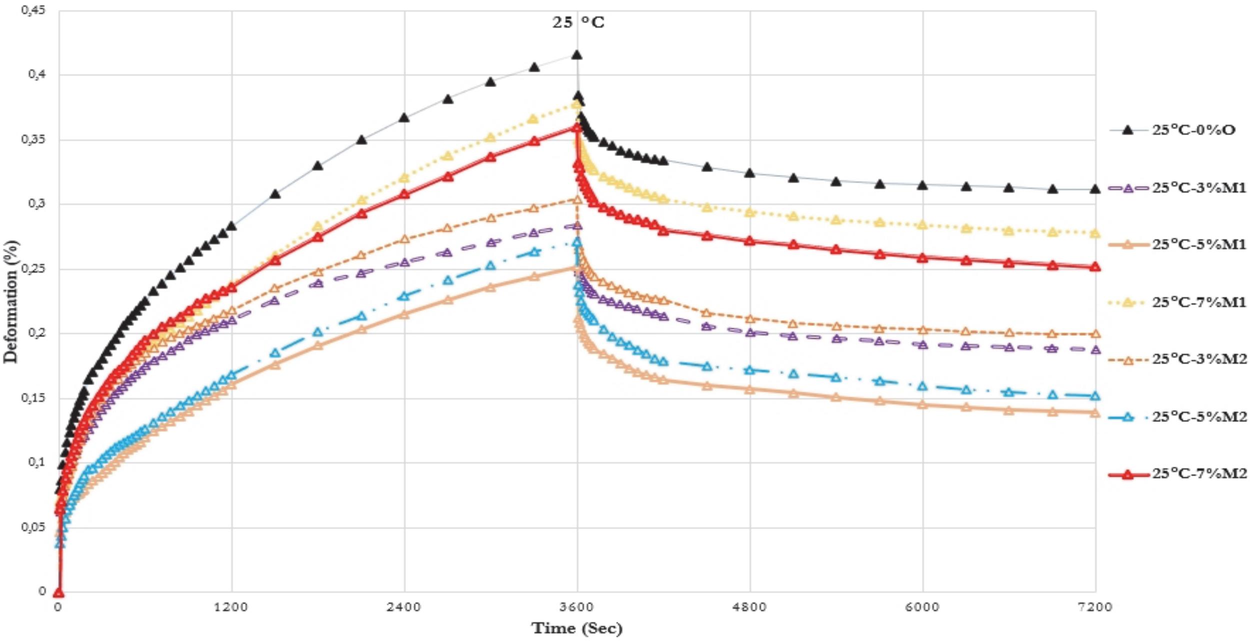
The deformations increase with the rise of the temperature, compared to the unmodified asphalt mixture, we noticed significant deformations at a high temperature, it decreased from a value of 6% up to 23% (which we record at 5%M1).
On the other hand, in the low temperatures, the asphalt mixtures are more rigid and have low deformations [27], (35% lower in the 5%M1 with 'coarse' waste PET size), in which case the modified bituminous mixes are influenced by bitumen and PET plastic waste.
In the recovery part, the rate of reversible deformation according to maximum deformation shows that at medium and low temperatures, the bituminous mix can recover some of its total deformations (which reached a rate of 63% to 5%M1 at 0°C) by comparing to high temperatures.

Modification Effect

The influences of plastic waste (PET) addition by different percentages and with two sizes margins, in the modified and unmodified asphalt, are shown in Fig. 8:
The curve analysis in Fig. 8, shows that there is a reduction in creep recovery for all modified asphalt mixture regardless of the test temperature, waste PET contents, and also their sizes.
The PET (plastic waste) in bituminous mixes is used to reduce temperature sensitivity and to increase the service temperature range. The loading duration and period affect the viscous or elastic behavior [28].
In the creep part, the maximum creep deflection is lower for a PET waste content of 5% for both sizes for all three curves. This reduction appears well in the temperatures T = 0°C and T = 25°C with values between 33% up to 39% compared to the unmodified bituminous mix. The optimum additive contents improve the asphalt mix mechanical properties. The adhesion between the coarse waste PET and bitumen in the mixtures containing lower percentages of PET (at 3% and 5%) is possibly causing this creep deflection reduction. The improvement in the viscoelasticity of the mixture implies that the adhesion improves, the compressive mechanical properties are increased and the creep resistance is improved [6, 23].
On the other hand, by increasing the amount of plastic waste in the mixture, the particle phase becomes larger (7%) and the stability of the mixture under loading is reduced, regardless of the temperature and sizes, it has recorded a weak percentage in modified asphalt mixture's maximum deformation reducing which reaches 6% and 8% for 5%M2 and 5%M1 respectively at T = 50°C, which can be attributed to a reduction in binder workability in presence of waste PET's addition and aggregate full surface's coverage by stiffer binders.
At higher waste PET contents, the aggregate will be replaced by these particles which have less stiffness. In consequent, the reduction of bitumen content around the aggregate particles might be another reason of stiffness reduction [28].



a)



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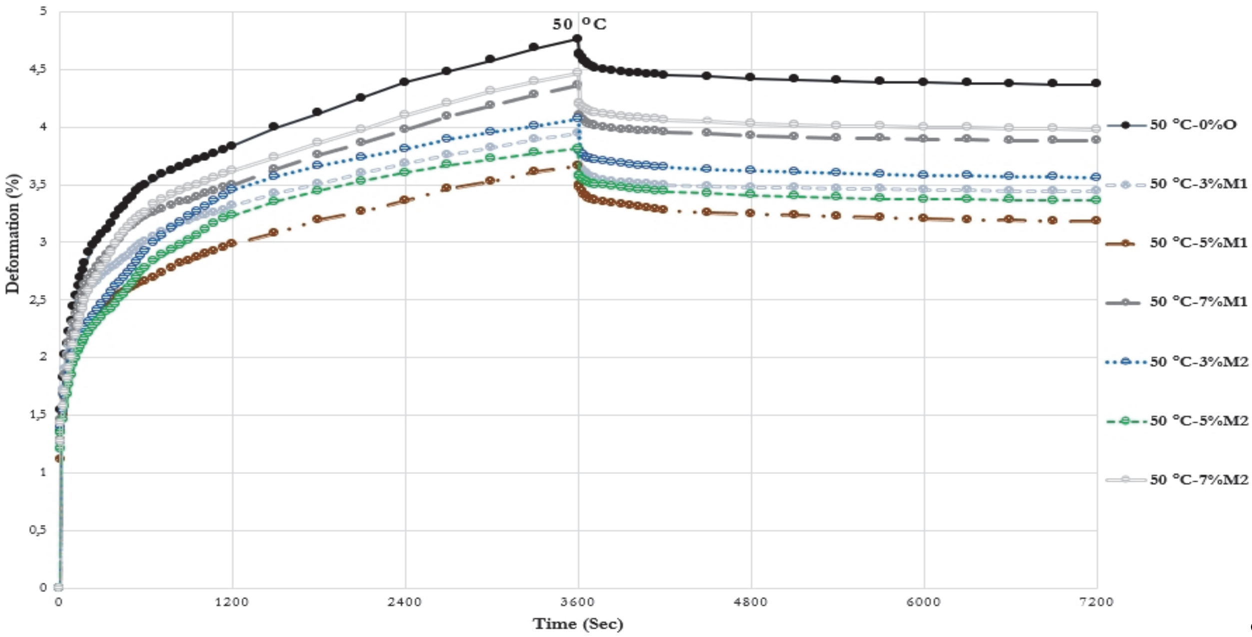

c)

Figure 8: Effect of plastic waste PET on different hot mix asphalt; a) At 0°C, b) At 25°C, c) At 50°C

For low temperatures, it is noticed that the viscoelasticity part submitted by the modified asphalt mixture is not significant as the unmodified asphalt mixture (a weak parabolic curve) for all percentages. The best results have been recorded at a low temperature. At low temperature, where much of deformation is elastic, the PET waste addition reduces pavement cracking with the asphalt mixture contraction properties, so that the lowest maximum deformation rate is recorded for 7%M2.
At medium temperature, it can be seen that the waste PET addition leads to an improvement in the bonding forces between bitumen and aggregates. It has recorded a noticeable deformation decrease estimated at 32% and 39% for 3%M1 and 5%M2 respectively. Waste PET has viscoelastic property, which means that the material's reactivity is temperature-dependent [28].
At a high temperature, it recorded lower rates of maximum deformation, despite this, the addition of waste PET to asphalt mixture enhances the material's rigidity and restricts the permanent deformation under heavy loading conditions especially in the upper pavement layer. When the polymer is applied to an asphalt mixture, they usually result in a higher degree of

stiffness and improve the temperature's susceptibility of the mixture, and, as a result increase the resistance of the mixture against rutting.

In the recovery part, in the same way we observe that the addition of waste PET clearly prevents the permanent residual deformations measured for all temperatures, with a remarkable reduction at the temperature T = 0°C which reaches 59% to 5%M1 compared to the unmodified asphalt mixture. This reduction is reduced according to the increase in temperature which reaches 27% at 5%M1, while we record the weakest percentage 9% for 7%M2 at T = 50°C.

For all temperatures and all contents, the increased stiffness of PET waste's modified mixes can be explained by its natural state which is a semi-crystalline, and its glass transition temperature which is about 70°C [10, 29, 11]. the PET crystalline portions still exist as solid and rigid because PET has a melting point (approximately 250°C), which is much higher than the mixing temperature (160°C) used in our study. The softened (molten portion likely) improves the aggregate-bitumen bond, and the rigid crystalline portion imparts to the mixes [17].

In the dry process, we have added the waste PET plastic in the final part of mixing, to keep it in its natural state, with minimal change of its properties [11].

It is seen that the best results attain its peak value at 5% PET, and this trend is seen for both PET sizes, but well appears for 2.5 mm – 1.25 mm sizes or when coarser PET size is used, the results are even better than the finer PET size.

In terms of the instantaneous elastic deformation, maximum deformation, and residual deformation, the coarse PET waste size in the asphalt mixture gives better results in the range of 3% to 13% compared to the fine waste PET size in the bituminous mix; at all percentages, coarser PET particles generated higher bulk density than the finer ones; this can be explained in terms of additional surface area to be covered by the binder. When finer PET size is used, the binder coats a larger surface area, resulting in reduced workability during mixing [17].

The effect of lower friction between PET waste aggregates exceeds the good adhesion between the bitumen and waste PET elements.

CONCLUSION

In this study, the effects of adding different percentages of waste PET particles with two sizes at three different temperatures were investigated on the creep recovery of modified asphalt mixture and compared with unmodified asphalt mixture. The results found are as follows:

- Resistances of flow and creep-recovery behavior were changed by adding waste PET particles. It was shown that although stiffness of the modified asphalt mixture initially increased by adding lower percentages of waste PET into the mixtures compared to the conventional mixture, it decreased at a higher amount of PET content (e.g., 7% PET) for both sizes; The optimum content of PET plastic waste is 5% for both sizes where the mixture becomes more flexible and prevents permanent deformation and propagation in mixtures due to temperature application.
- At higher waste PET contents, the bitumen will accumulate on the surface of the waste PET particles, resulting in a decrease in the thickness of the bitumen around the aggregate particles, so a decrease in the adhesion between the aggregate and the bitumen, and eventually an increase in the creep-recovery of the modified asphalt mixture.
- As the temperature increases, the measured deformation increases with time, while at low temperatures, the deformation becomes more rigid; at moderate temperatures, the asphalt mixture loses some rigidity, but still resists creep recovery.
- In general, waste PET modified asphalt mixture with coarser PET size showed comparatively superior performance than fine size for all percentages at different test temperatures.
- Overall, the use of waste plastic PET as an additive in the bituminous mix is encouraged not only as a promising way technique to enhance the performance of asphalt mixes but also is desired in both economic and environmental because this recycled material is cheaper than the original polymer modifiers like the Ethylene-vinyl acetate (EVA), the Styrene-butadiene-styrene (SBS), etc. So, could reduce the cost of road construction and it's easy to obtain this secondary material due to large quantities in the environment.

ACKNOWLEDGEMENT

The authors acknowledge the support provided by the Laboratory of Fiability of Materials in Saharan Regions (FIMAS) and DGRSDT MHESR-Algeria.

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