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Comparative Analysis of the performance of Asphalt Concretes Modified by Dry Way with Polymeric Waste

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Abstract

An asphalt concrete has been modified by adding four polymeric wastes: polyethylene (PE) from micronized containers, polypropylene (PP) from ground caps, polystyrene (PS) from hangers and rubber from end-of-life tyres (ELT). These polymeric wastes were selected according to their availability, homogeneity and economic criteria considering the big amount of material required to build a road.

The dry method has been used to modify the bituminous mixture due to its simplicity and the possibility to be carried out in any asphalt plant without important modifications. This is very important in order to spread the process and recycle the polymeric waste in the same place where it is produced, hence improving the environmental impact.

The reference asphalt mixture and the four modified asphalt concretes have been analysed separately and their performance compared, evaluating their resistance against plastic deformation, stiffness, fatigue resistance and workability. The Master curve and the Black diagram of the mixtures were also calculated.

The results showed that the use of polymeric wastes significantly increased the stiffness of the reference mixture in all cases, but especially when PE, PP and ELT were used. However, none of these materials significantly modifies the fatigue behaviour of the reference mixture. Regarding the resistance against plastic deformation, the use of both PE and ELT led to an increase of the resistance, whereas PP did not modify it and PS decreased it. As for workability, the energy of compaction of the modified mixtures did not suffer any important change. Therefore, according to the results obtained, PE, PP and ELT can be used to modify asphalt mixtures since they improve or do not change their properties. On the other hand, PS should be further studied because of the contradictory results obtained, and only when plastic deformation is not a problem this material could be used.

Keywords: Asphalt concrete; Dry way; Polyethylene; Polypropylene; Polystyrene; Rubber; Polymeric waste; Modified mixture; Asphalt mixture; Plastic waste.

1. Introduction

The properties of polymers have motivated their use in multitude of products and applications. However, this proliferation implies an environmental risk if they are not correctly treated at the end of their useful life.

In recent years, the use of polymers in bituminous mixtures has significantly increased, especially in order to modify bitumen by wet way [1,2]. This is the most common use and it has clear advantages, producing a modified bitumen that improves multiple properties of conventional bitumen [3]. Nevertheless, this process has also disadvantages: in general, it is necessary to use specialized plants where high temperatures and agitation process are required; the method is economically costly and in some cases presents precipitation or compatibility problems [4-8].

In the mixtures considered in this article, the polymeric wastes have been incorporated directly to the mixer drum (dry way). This method is less widespread than the wet way, but it is simpler and it can be carried out in any asphalt plant without important modifications, so the spread of the process is favoured and the reuse of polymeric wastes facilitated.

One of the main projects carried out nowadays with polymeric wastes has been developed by the Thiagarajar College of Engineering Madurai in New Delhi (India), where modified asphalt concrete with a mixture of polymeric wastes by dry way (Polyethylene, PE; Polypropylene, PP; and Polystyrene, PS), have been used in rural roads. The mixture improved the normal performance of conventional mixtures avoiding cracking and potholes [9]. The polymeric wastes were added over the hot aggregate creating a film around them, improving adhesiveness with the bitumen that was afterwards added. This way, the mixture achieved higher values of Marshall stability and the bitumen percentage was reduced [10,11].

Normally, polymeric wastes are used independently due to multitude of polymers present in the market with different properties and also in order to better control the resulting mixture. Similar projects with low density Polyethylene (LDPE) showed an increase of the indirect tensile strength and resilient modulus [12], as well as of the resistance against plastic deformation and fatigue [13]. Virgin fibres of PP have been also used that were mixed with the aggregates before adding the bitumen. Results showed an increase of the mixture Marshall stability and fatigue resistance [14]. Nevertheless, in a study with PP from plastic waste it was concluded that its incorporation improved the resistance against plastic deformation but it did not have any influence on its fatigue resistance [15]. The use of PS as a modifier of bituminous mixtures is less developed due to the fact that it presents more compatibility problems with bitumen [16]. In a project carried out in 2012 with PS from plastic waste, the best results were obtained by dry way when 5% was added to the aggregates before pouring the bitumen, increasing in this

way the Marshall stability by 11%. On the other hand, when the polymer was added after the bitumen it was not completely mixed achieving a lower Marshall stability [17].

The use of rubber is more extended than the plastic wastes. One of the critical parameters that determines the mixture performance is the digestion process, which basically depends on the interaction time between the rubber and the bitumen, the mixture temperature and the rubber particle size [18]. When the digestion has not been correctly done, mixtures can have problems of cohesion, thus decreasing the binding capacity of the original bitumen [19]. To make this process easier, it is recommended that the maximum size of the rubber is 0.6mm, its maximum quantity does not exceed 1% of the aggregates weight and the digestion time takes between 60 and 90 minutes [20,21]. The addition of the rubber using the dry process achieves a better performance against the plastic deformation and cracking [22].

The main difference between the use of the plastic polymers and the rubber is that the former can improve the joining among the aggregates and modify the performance of the mixture, while the latter requires a digestion process to modify the properties of the bitumen, process which is not the aim with the plastic polymers.

This paper gathers a comparative analysis of a modified mixture by dry way, using the most common polymeric wastes. The resistance against the plastic deformation, the stiffness, the fatigue resistance and the workability have been analysed. The results show that the addition of these polymers can be useful to improve the properties of the roads, so this process can convert the roads into a tool to recover big quantities of polymeric wastes and make better also their environmental impact.

2. Materials and methodology

2.1. Bitumen and aggregates

For the research done, a conventional bitumen was used as binder, with a penetration grade of 56.8 (EN 1426) and a softening temperature of 51.1°C (EN 1427). The coarse aggregate was

porphyritic, with a coefficient of Los Angeles of 14 (EN 1097-2) and Flakiness index of 24 (EN 933-3). Finally, the fine aggregate used was limestone with a Sand equivalent of 63 (EN 933-8).

2.2. Polymeric wastes

In terms of their internal structure, the PE and PP are two crystalline polymers while PS is amorphous. All of them are thermoplastic polymers which are softened when the temperature exceeds their melting (or glass) point. On the other hand, the rubber is a thermostable polymer that has been vulcanized and works at higher temperatures of its glass transition temperature in its rubbery state.

The polymeric wastes were supplied and characterized by AIMPLAS (Research Association of Plastic Materials). A first selection was done according to economic and technical criteria. As big quantities are required for its use in road works, the availability must be constant and sufficient, the cost limited, and the polymers should be homogenous enough in order not to change the properties of the mixtures. The selected polymers are shown in Figure 1.



Figure 1. Selected polymeric waste: PE, PP, PS and ELT from left to right.

According to their particle size distribution (Figure 2), the PE is the smallest of the thermoplastic polymers with a maximum size of 2mm, whereas PP and PS have a very similar distribution and a very close maximum size: 6.3mm and 5.6mm, respectively. The ELT is the smallest of all the polymers with a maximum particle size of 1mm.

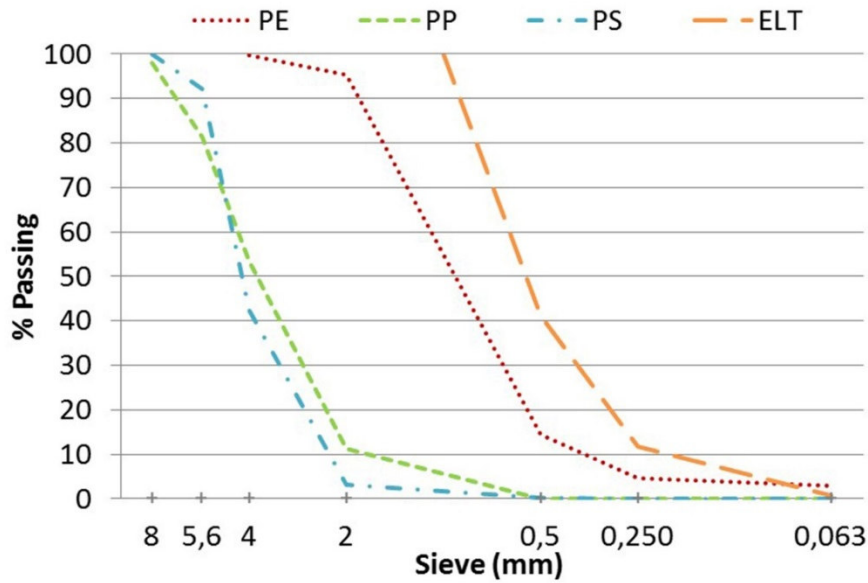


Figure 2. Particle size distribution of the polymeric wastes.

Table 1 gathers the densities of every polymeric waste, which were required to replace the natural filler by volume.

Table 1. Density of selected polymeric wastes [23]

Polymer	PE	PP	PS	ELT
Density (g/cm ³)	0.90	0.94	1.05	1.15

Differential Scanning Calorimetry (DSC) tests were carried out to plastic polymeric wastes to find out their thermal behaviour (this point is required to define the mixing temperature) and their composition (Figure 3). Thus, the melting temperature of the PE sample was 130.72°C, corresponding to a High Density Polyethylene (HDPE). The DSC also showed a minor quantity of Polypropylene. The sample of PP was actually a mix between PP and PE, with melting points at 161.07°C and 132.36°C, respectively. PS is an amorphous polymer with a glass transition temperature of 98.70°C. This DSC also shows traces of other compounds, with a part of PP with a melting point of 162.18°C and a small quantity of HDPE with a melting temperature of 129.56°C. The DSC was not carried out with rubber due to it is a vulcanized material and its glass temperature is below our working temperatures, so it was not necessary.

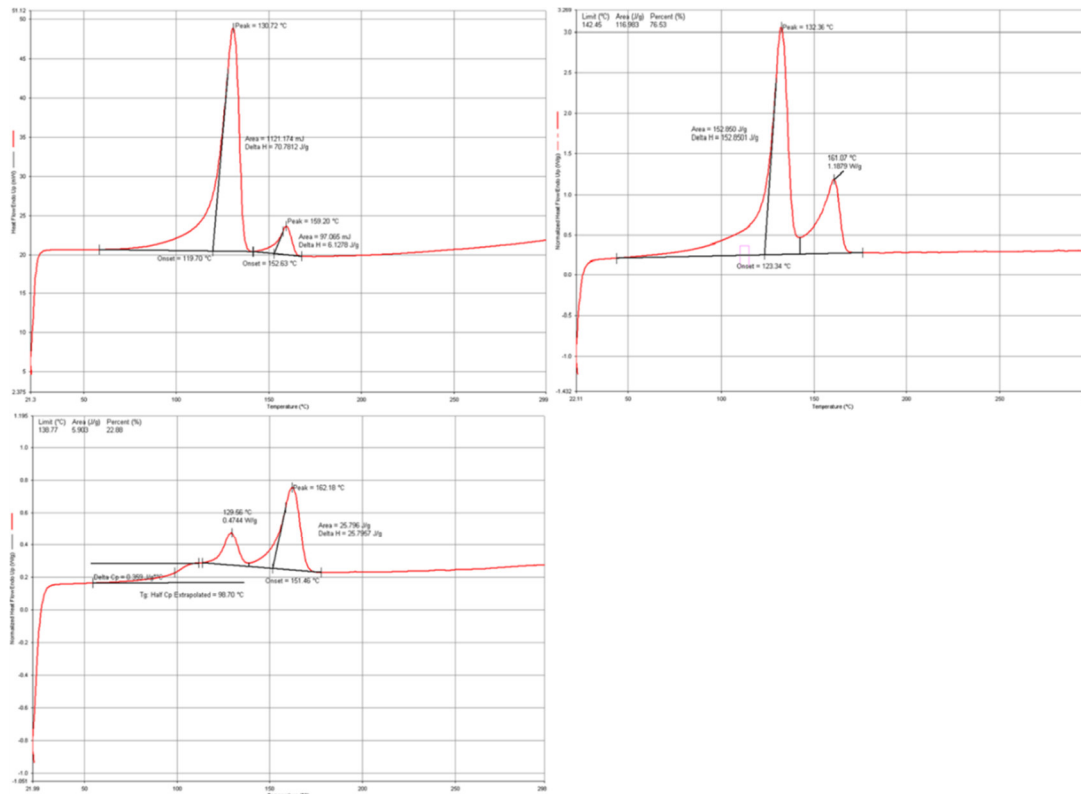


Figure 3. DSC test of the polymeric waste. From left to right, and up to down: PE, PP and PS

2.3. Specimen preparation

The design of the mixtures was performed by ACCIONA [23] in a previous stage of the research. The mixture was a semi-dense Asphalt Concrete for surface layer (AC22 surf 50/70 S) and the amount of bitumen used was 4.8% based on total weight of mix. The temperature of the bitumen and aggregates were 155°C and 175°C respectively. The particle size distribution of the mixture is shown in the following table.

Table 2. Particle size distribution [23]

Sieve (mm)	22	16	8	2	0.5	0.25	0.063
% Passing	96	78	55	29	13	10	6.1

The polymers were added replacing natural aggregate only in the filler fraction, unlike other authors who replaced other fractions or directly added a percentage of the polymeric waste by weight [17,24]. The remainder of the particle sizes stayed unchanged. To achieve this, 1% of aggregates was replaced by polymeric waste corresponding to the volume occupied by this percentage only in the filler fraction. The polymeric wastes were incorporated differently. Thus, the plastic polymers were added directly in the mixer drum with the hot aggregates before the

bitumen was incorporated, while the rubber was added after the bitumen. In the first case, the polymers were softened so that they basically wrapped the aggregates, thus improving the linkage among them. Besides, they also improve adherence between binder and aggregates [10]. In the second case, the rubber has a low influence on the aggregates and it was mainly mixed with the bitumen, modifying its properties although it was added by dry way [25]. The mixtures achieved the properties presented in Table 3, all of the required by the Spanish specifications.

Table 3. Characteristics of used mixtures [23]

Bituminous mixtures	Density (g/cm³)	Voids in mixture (%)	Water sensitivity (%)	Wheel tracking test (mm/10³ cycles)
REF	2.400	4.4	97.8	0.08
PE	2.308	4.7	89.2	0.05
PP	2.346	4.1	90.4	0.06
PS	2.328	4.5	92.8	0.04
ELT	2.430	4.8	94.3	0.07

2.4. Testing program

The resistance against plastic deformation and stiffness were evaluated to know the bearing capacity of the modified mixtures, whereas the fatigue resistance test was carried out to analyse the mechanical useful life. Finally, the workability was also calculated to know the energy of compaction and to evaluate whether the addition of the polymers would require a modification in the compaction process or not.

As the tests have been carried out in the same conditions, the modified mixtures can be compared among them. Therefore, it can be determined which polymeric waste obtains the best performance or which of them is the most suitable to improve a specific property.

The wheel tracking test (EN 12697-22) was done to evaluate the resistance against plastic deformation, which is one of the most important parameters of a bituminous mixture, especially in the southern countries due to the heat and the high axle loads (11.5t for simple axle in the case of Spain). The average slope of the samples between the cycles 5000 and 10000 is a

measure of the accumulated plastic deformations. The lower the average slope, the greater the mixture capacity to resist the plastic deformation.

The test (Figure 4A) was carried out through four 60mm high specimens per modified mixture, which were tested at 60°C. The load used was 700N.

The dynamic modulus was obtained by the four point bending test (EN 12697-26. Annex B). This test evaluates the stiffness of the mixture, which strongly depends on the temperature and the frequency selected to perform the test. This modulus links the applied load and the strain produced, and was performed with the specific devices shown in Figure 4B. The test was done at 20°C with a number of prismatic samples between 15 and 18, depending on the type of mixture. The frequencies went from 0,1Hz to 30Hz.

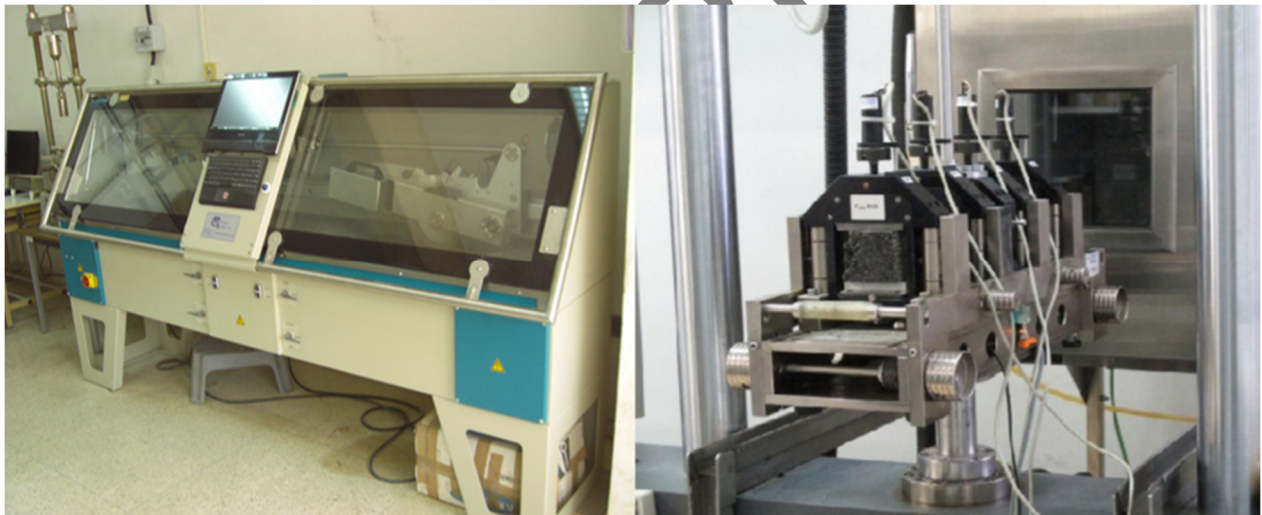


Figure 4. From left to right: A) Wheel tracking test machine B) Experimental equipment to perform dynamic modulus test

The Master curve was obtained in order to analyse the dynamic performance of the mixtures. This curve evaluates the modulus response (M) to different frequencies (f) and temperatures, and it was adjusted to a sigmoidal curve (Equation 1) by the least-squares adjustment[26,27].

$$\text{Log } M \text{ (MPa)} = \delta + \alpha / (1 + \exp(\beta - \gamma \cdot \log f(\text{Hz}))) \quad (1)$$

An equivalent process was carried out with the phase angle of the mixtures, which is a measure of the gap between the moment when the load is applied and when the sample begins to be

strained. It represents the weight of the elastic component, which also depends on the temperature and frequency of the test. In order to know the weight of the elastic performance independently of the frequency or the temperature, the Black Diagram was also calculated. The Black Diagram displays the values of the phase angle (Φ) for a specific value of the dynamic modulus (M); therefore, this ratio will be achieved by the mixtures in different circumstances. This diagram was calculated by adjusting the phase angle and the dynamic modulus to a polynomial function by the least-squares method (Equation 2).

$$\Phi(^{\circ}) = a + b \cdot \log M \text{ (MPa)} + c \cdot \log M \text{ (MPa)}^2 \quad (2)$$

The fatigue resistance must be also analysed since the influence of each polymeric waste should be known, and because when a mixture is stiffer it is normally less flexible at the same time, so it can have cracking problems (though simultaneously a higher load would be needed to achieve the same strain). The four point bending test (EN 12697-24. Annex D) was carried out to evaluate the useful life of a mixture under repeated loads. The test, as in the case of the dynamic modulus, was performed with a number of samples between 15 and 18, at a frequency of 30Hz, a temperature of 20°C. The failure criterion was the cycle (N) for which the sample presented a stress of $\sigma_0/2$, being σ_0 the initial stress for the imposed strain (ϵ). This is equivalent to decreasing the initial Stiffness (S_0) of the material until its half. As the results were obtained, the fatigue laws were calculated with the following equation:

$$\epsilon \text{ (m/m)} = d \cdot 10^{-3} \cdot N \text{ (Cycles)}^e \quad (3)$$

To define the influence of each polymer in the workability of the mixture and to know if any change in the compaction process could be necessary, a workability test was carried out with five samples of each type of mixture. The diameter of the samples was 100mm, the load of the test was 600KPa, the speed of the movement 30 rpm and the angle of rotation 0,82°. The accumulated energy per mass unit was calculated by using the model developed by *del Río*[28]

(Equation 4):

$$\frac{W}{m} = \sum_1^N \frac{W_i}{m} = \frac{2 \cdot \pi \cdot \alpha \cdot A}{m} \sum_1^N h_i \cdot S_i \quad (4)$$

Where W (kJ) is the energy, m (kg) the mass, S_i (kN/m²) is the shear stress measured in each cycle i ; h_i (m) is the height of the sample in each cycle i ; A (m²) is the area of the sample; α (rad) is inclination angle of the cylindrical sample; and N is the total of applied cycles.

The required energy of compaction, which depends on the amount of voids of each mixture, was adjusted to the following straight lines by the least-squares method (Equation 5).

$$\text{Energy (KJ/Kg)} = f \cdot \text{Voids (\%)} + g \quad (5)$$

3. Results and discussion

3.1. Plastic deformation

The results of the wheel tracking test (EN 12697-22) are shown in Table 4. The reference mixture obtained a good value but its use is not recommended for high heavy traffic level in warm areas. The mixtures with PE, PP and ELT considerably increase the resistance against plastic deformation as compared with the reference mixture, and they can be used in any road without limitations of heavy traffic level or climatic conditions. These results are actually very similar to that gathered in Table 3. As for the PS, this case is different since the results obtained were worse than for the reference mixture, while in the design stage its performance was very good. This difference can be due to a lack of homogeneity not detected during the manufacturing of the samples: great quantities of polymeric waste were used and its components could have varied more than expected, or they simply could have been contaminated during the recycling process.

Table 4. Wheel tracking test

Mixture	Average slope (mm/10³ cycles)	Rut depth (mm)
REF	0.08	2.7
PE	0.05	2.6
PP	0.06	2.4
PS	0.13	4.3
ELT	0.06	2.8

The results have been analysed with the software IBM SPSS to find out if there was a real difference between the reference and the modified mixtures. The Shapiro-Wilk test was carried out and, as the results did not follow a normal distribution, the U test of Mann-Whitney was done to check the results. The statistical significances of the modified mixtures in relation to the reference mixture are shown in Table 5. The confidence interval was always 95%, so when a statistical significance is below 0.05 it implies that the analysed results are significantly different. As it can be seen, PE and ELT significantly increase the resistance of the reference mixture against plastic deformation, while PP does not have a significant incidence. Finally, PS worsens the result of the reference mixture.

Table 5. Significance of the average slope

	PE	PP	PS	ELT
REF	0.028	0.289	0.050	0.028

The increase of the resistance against plastic deformation with the thermoplastic polymers can be due to an increase in the internal resistance of the mineral skeleton. Thus, when the polymers are added over their melting temperature, they are softened and work as another binder among the aggregates, linking the mineral structure when the temperature is decreased and finally recovering their solid state. In Figure 5 it is shown how when the PE and PP are added, the aggregates are joined by these links probably improving this way the mixture cohesion.



Figure 5. Coarse aggregates with PE and PP

With the addition of ELT, small elastic particles are incorporated which react with the bitumen, increasing its elastic performance provided that a proper digestion is reached, increasing this way the mixture its resistance against the plastic deformation.

3.2. Stiffness

The results of the four point bending test (EN 12697-26. Annex B) showed that the incorporation of the polymers makes the mixtures stiffer (Figure 6). Nevertheless, the results obtained can be divided in two groups: the mixture with PS in one side, which slightly increases the stiffness as compared to the reference mixture; and mixtures including PE, PP and ELT on the other side, which increase the stiffness with a much higher influence than that of the reference mixture.

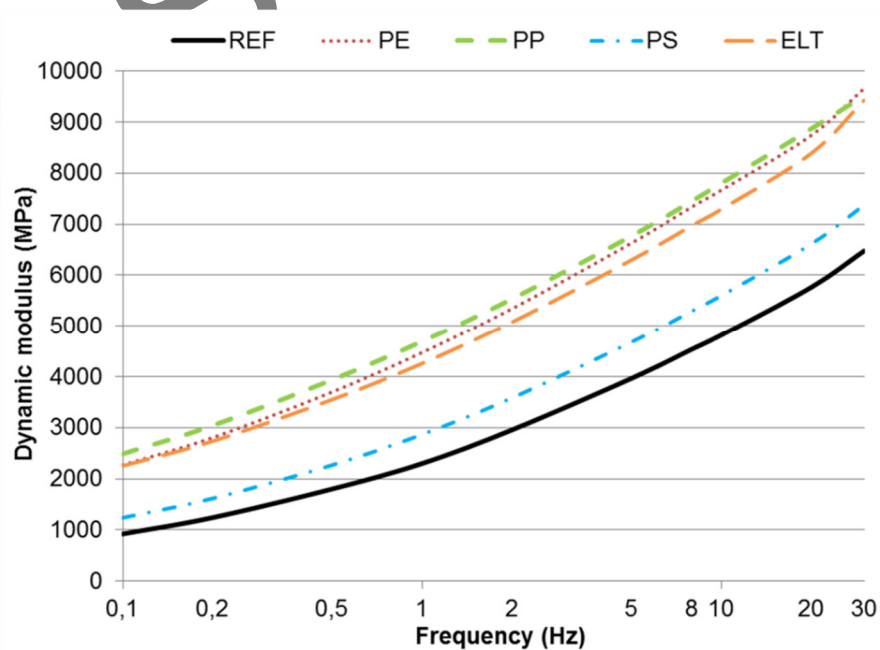


Figure 6. Dynamic stiffness of each type of mixture

Moreover, the increase in the dynamic modulus implies that these modified mixtures will actually have a greater bearing capacity and they will transmit fewer loads to the layers underneath. On the other hand, the mixture including PS was the only mixture whose results do not follow a normal distribution, so to compare the results of the PS and the reference mixtures a Mann-Whitney U test was carried out and the statistical significances are shown in the next table. All the polymers significantly modify the reference mixture.

Table 6. Significance of the dynamic modulus for each type of mixture

	PE	PP	PS	ELT
REF	0.000	0.000	0.004	0.000

From these results the Master Curve of each mixture was developed. Its coefficients, the coefficient of determination, maximum and minimum values are shown in Table 7.

Table 7. Master curves of the reference and modified mixtures

Mixtures	δ	α	β	γ	R^2	M_{min} (MPa)	M_{max} (MPa)
REF	1.096	3.233	-0.849	0.538	0.97	12.5	21330.4
PE	1.914	2.480	-0.840	0.514	0.96	82.0	24774.2
PP	2.046	2.310	-0.846	0.521	0.86	111.2	22698.6
PS	1.333	2.998	-0.883	0.538	0.95	21.5	21428.9
ELT	1.843	2.708	-0.655	0.427	0.96	69.7	35563.1

These Master Curves (Figure 7) verified that all the modified mixtures increase the dynamic modulus as compared to the reference mixture, and depending on the selected frequency the difference is greater.

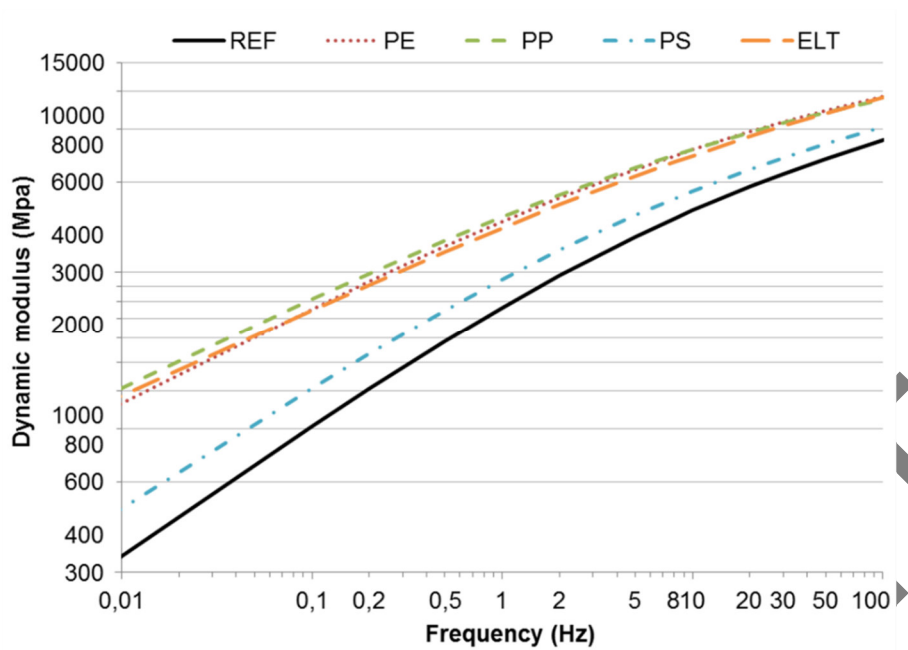


Figure 7. Master curves of each mixture

The frequency is linked with the traffic velocity. When the velocity is low the potential problem is the plastic deformation, while at high speeds the potential problem is the cracking.

Considering low velocities, it has been confirmed that the incorporation of PE, PP and ELT to the mixture makes the resistance against plastic deformation increase, and it is also at low frequencies when the difference between the modified mixtures is greater; meanwhile, at high frequencies the dynamic modulus is still different even though this difference is lower.

The results showed that the mixtures whose modulus increased also saw their phase angle decrease, thus incrementing the elastic performance of the mixture. As it can be seen in Figure 8, the mixtures modified with PE, PP and ELT present the lowest phase angle followed by PS.

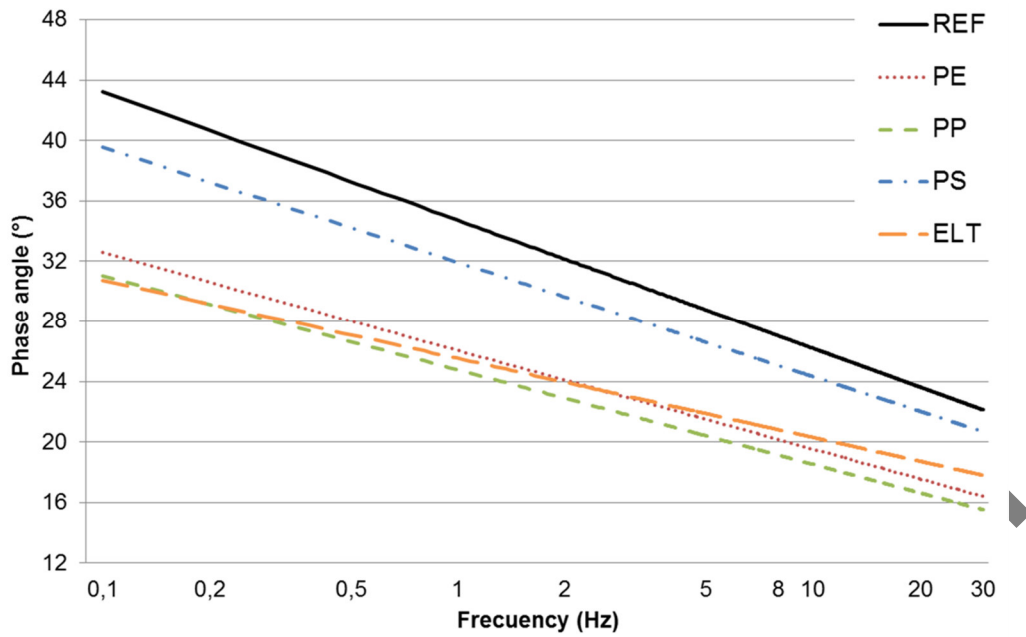


Figure 8. Phase angle of the mixtures related to frequency at 20°C

The Black Diagram was obtained for each mixture, whose coefficients and coefficient of determination are presented in Table 8.

Mixtures	a	b	c	R ²
REF	34.304	24.734	-7.318	0.96
PE	161.470	-49.171	3.254	0.90
PP	150.568	-44.884	2.825	0.88
PS	89.815	-10.090	-1.971	0.93
ELT	66.221	-1.696	-2.637	0.93

It was verified that mixtures including PE, PP and ELT are mixtures with lower phase angle and therefore, more elastic performance. The mixture with PS is between them and the reference mixture, except when the reference mixture achieved its highest modulus, then the mixture including PS presents a higher phase angle, as it is shown in Figure 9.

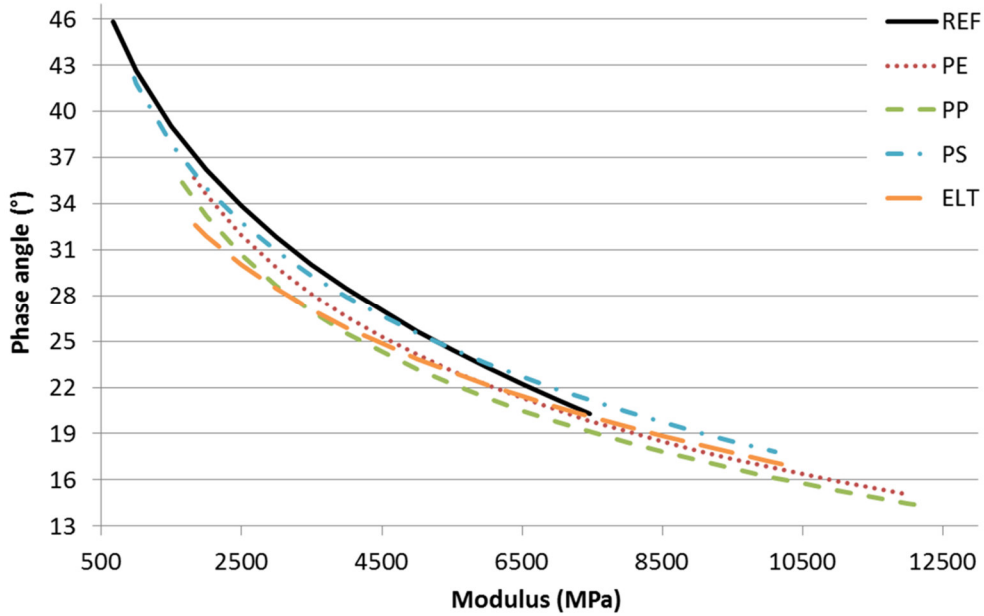


Figure 9. Black Diagram

3.3. Resistance to fatigue

The new links created by the thermoplastic polymers between the aggregates are flexible enough not to punish the fatigue performance of the mixtures. With the addition of rubber the elasticity of the mixture increased and the highest resistance was obtained (Figure 10).

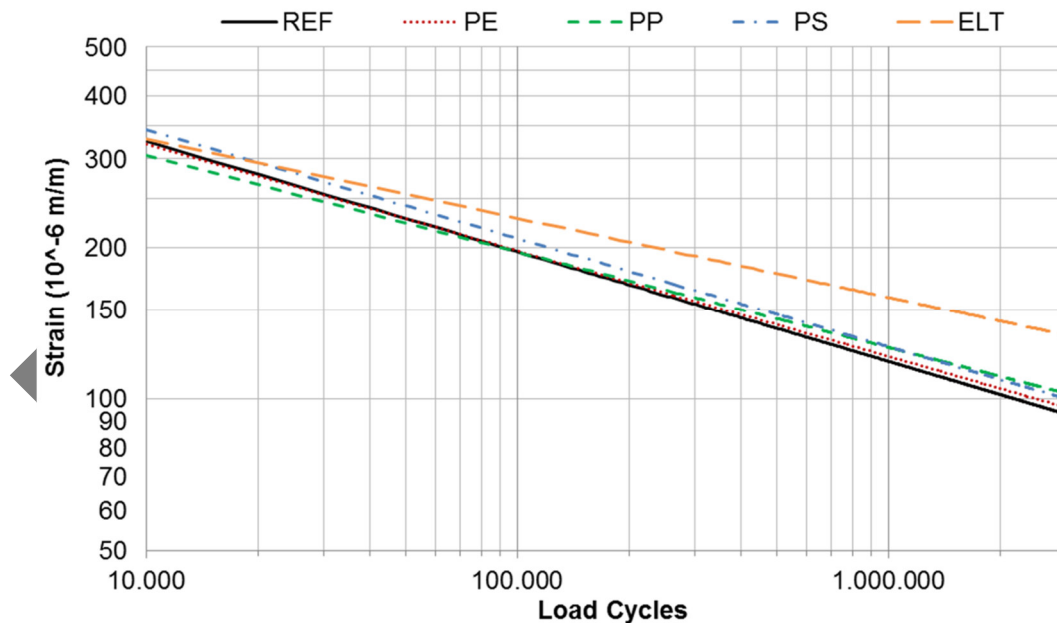


Figure 10. Graphical representation of the fatigue curves: Deformation - Cycles

However, there is no significant difference between the mixtures (the significance of the Kruskal – Wallis test was 0.708). The fatigue laws obtained by means of the four point bending

test (EN 12697-24. Annex D), the initial stiffness (S_0) and the characteristic strain at 10^6 cycles are included in Table 9.

Table 9. Fatigue laws, initial stiffness and characteristic strain

Mixture	S_0 (MPa)	strain-characteristics* ($\mu\text{m/m}$)	d	e	R^2
REF	5645.0	118.5	2.4543	-0.2194	0.96
PE	8580.0	121.2	2.2506	-0.2115	0.83
PP	8205.4	126.3	1.7723	-0.1912	0.90
PS	6205.0	126.7	2.5206	-0.2164	0.93
ELT	8512.0	159.1	1.4083	-0.1578	0.83

* 10^6 cycles

Therefore, the fatigue resistance is not modified by the incorporation of the polymeric wastes here studied, and the modified mixtures, although stiffer, have the same performance than the reference mixture. Thus, these modified mixtures transmit a lower load to the layers below and resist the same number of axles, which implies that the layer thickness could be reduced according to the analysed mechanical parameters. This means an important reduction in the consumption of raw material, although many other parameters must be considered yet.

3.4. Workability

The coefficients of the straight lines and the required energy per mass for 5% of voids in the mixtures are presented in Table 10.

Table 10. Lines of the required energy of compaction and values for 5% of voids

Temperature	f	g	R^2	Energy 5%
REF	-0.14	2.57	0.94	1.87 kJ/kg
PE	-0.15	2.62	0.87	1.87 kJ/kg
PP	-0.16	3.06	0.95	2.26 kJ/kg
PS	-0.16	2.96	0.98	2.16 kJ/kg
ELT	-0.16	2.96	0.95	2.16 kJ/kg

The next graph (Figure 11) represents the accumulated energy of compaction depending on the voids containing each type of mixture.

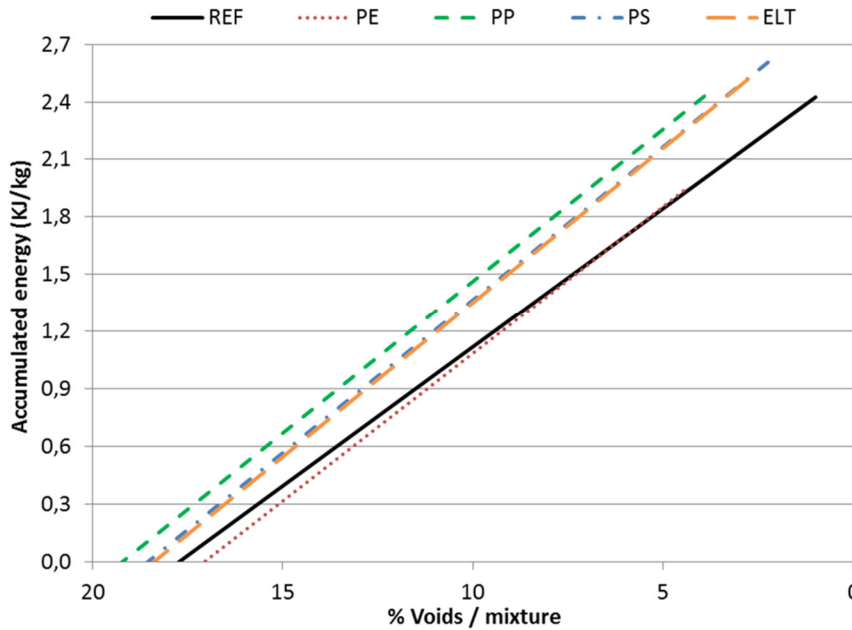


Figure 11. Energy of compaction - Voids

Although in the Kruskal-Wallis test a significance of 0.397 was obtained, which means that there are not significant differences, if results are anyway analysed it seems that for the thermoplastic polymers, the bigger the particle size, the greater the increase of the required energy. The addition of rubber, despite it has the same size than PE, also increases the energy of compaction, probably because it is not melted as the PE when the aggregates are heated up. These differences in the energy of compaction are due to the fact that the polymers have replaced particles of filler, much smaller than them.

4. Conclusions

Four materials were analysed in this paper that modified the composition of a conventional asphalt mixture. PE, PP, PS and ELT were used to replace 1% of the filler fraction by dry way, the performance obtained of each modified mixture has been analysed.

According to the results, the mixtures modified with PE, PP and ELT have a similar performance, increasing the resistance against plastic deformation and the stiffness as compared to the reference mixture, not affecting the stiffness the fatigue resistance at laboratory level.

Any of these polymeric wastes, considering the analysed properties, can be used to modify a bituminous mixture as they increase the capacity of the reference mixture or, in the worst case, they do not change it. The availability and the costs will be probably more important to select one of them than the mechanical performance.

Once each mixture has been independently analysed, it can be concluded that:

- Mixtures including ELT increases 30% the resistance against plastic deformation and 50% the stiffness as compared to the reference mixtures. The mixture has also a more elastic performance with a lower phase angle.
- PP has a very similar performance to the rubber, being that polymeric waste the one that increases the stiffness of the reference mixture the most (60%), and showing also the most elastic behaviour; however, the increase of its resistance against plastic deformation is not statistically significant.
- PE is the polymeric waste which increases the resistance against plastic deformation of the reference mixture the most (60%). This material also increases the stiffness of the asphalt mixture (60%) and its elastic performance as compared to the reference mixture.

Regarding PS, this is a different case because important differences in the results have been obtained. The variability in the performance of the mixtures with PS against the plastic deformation makes therefore necessary to further analyse its behaviour. This variability can be produced by a lack of homogeneity in its composition due to a contamination in its process of recycling.

In any case, it is the polymeric waste that changes the reference mixture the least, this is the point where the PS is the polymer with poorest properties as binder [10]. PS increases the stiffness of the reference mixture 15%. This is the polymer with the lowest elastic behaviour of

all the modified mixtures. Besides, the resistance against plastic deformation is decreased, thus getting its performance worse.

Finally, the required energy of compaction and the fatigue resistance do not have significant differences among any of the mixtures.

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