WASTES: Solutions, Treatments and Opportunities
1st International Conference
September 12th – 14th 2011

WASTE POLYMERS RECYCLING IN HIGH PERFORMANCE ASPHALT MIXTURES

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ABSTRACT
Building a road pavement requires large amounts of constituent materials, whose extraction can lead to the devastation of natural resources and causes negative impacts on the environment. As aggregates comprise nearly 90% of asphalt mixtures, their partial substitution by waste thermosetting polymers (12 million tons of waste polymers are presently mislaid into landfills, every year, in Europe) can be considered as a sustainable technology, given that an equivalent performance can be assured. Thus, this study aims on evaluating possible advantages of introducing polyethylene based wastes in asphalt mixtures, namely high density polyethylene (HDPE) and cross-linked polyethylene (PEX), incorporating HDPE as a bitumen modifier and PEX as partial substitute of the aggregates. The laboratory tests carried out in this work showed that the use of HDPE significantly improves the behavior of the asphalt binder/mixture. The use of PEX considerably decreases the density of the mixtures, which can be attractive to lighten structures. The mixtures with PEX have similar performance to the conventional one in terms of water sensitivity, improving the permanent deformation resistance and reducing the temperature susceptibility. In brief, the mixtures incorporating waste polymers could be considered a good technical and environmental alternative for paving works.

Keywords: Waste polymer recycling; Polyethylene; Polymer modified binder; Asphalt performance

INTRODUCTION
Presently, in Europe, 12 million tons of waste polymers are mislaid into landfills every year [1]. Most of these polymers have a very simple process of recycling, but the mechanical recycling generally does not work for thermosetting polymers and elastomers, because these polymers do not melt. Since they cannot be reprocessed, their reuse may be possible by taking advantage of their potential as aggregates that can be incorporated in road pavements.
In fact, building a road pavement requires a large amount of constituent materials, whose extraction can lead to the devastation of natural resources and cause negative impacts on the environment. As aggregates comprise nearly 90% of asphalt mixtures, their partial substitution by waste thermosetting polymers can be considered as a sustainable technology, given that an equivalent performance can be assured [2].
Thus, this study aims on evaluating the possible advantages of introducing polyethylene based wastes in asphalt binders and mixtures by using different processes of incorporation. The materials used were high density polyethylene (HDPE) and cross-linked polyethylene (PEX), the first as a bitumen modifier and the second as partial substitute of the aggregates.
Polymers can be applied in asphalt mixtures as bitumen modifiers or as partial substitute of the aggregates, and they could also be used as an aggregate coating layer [3]. Among polymers that may be used in asphalt mixtures, the most used are thermoplastics and elastomers [4], namely polyethylene based polymers, as well as EVAs, SBS and SBR polymers. According to several authors [5, 6] the use of recycled polymers or waste polymers rather than new is possible and most valuable, which leads to ecological and possibly to economic benefits.

MATERIALS AND ASPHALT MIXTURES PRODUCTION CONDITIONS

Selection of Polymer Wastes

The aim of this study is to use a polymer residue that exists in high amounts in order to replace the aggregates in asphalt mixtures, so that their use is feasible, which presents properties that might be advantageous to the asphalt mixture. According to information gathered from local companies for solid waste collection and processing, cross-linked polyethylene (PEX) wastes are present in large quantities and have low commercial value due to their low demand (Fig. 1a). The polyethylene in PEX was crosslinked in order to become inert, because it is used in hot water pipes or electrical cable sheathing at high service temperatures. Actually, in this study PEX was selected to be used as aggregate in asphalt mixtures, because its crosslinked chains do not melt.
The crosslink process can be more or less effective, thus being important to determine the GEL content of the PEX residue used in this study. The results indicated that the GEL content is 54%, which is the percentage of crosslinked material that does not melt. In fact, some part of the used PEX can melt into the bitumen, thus modifying the binder during the production of asphalt mixtures. Thus, this study also evaluated, comparatively, the bitumen modification with wasted HDPE (high density polyethylene), which is another polyethylene based plastic that melts completely (Fig. 1b).

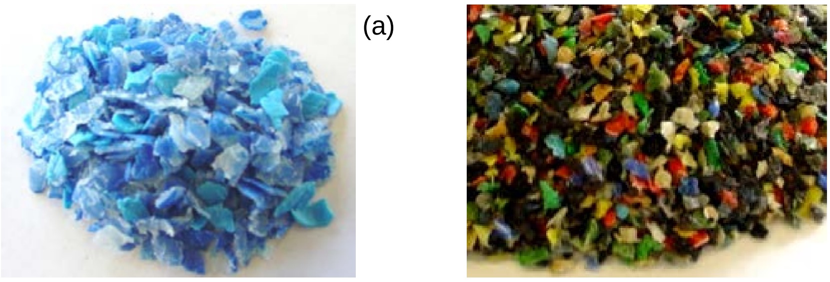
(a) (b)

Fig. 1 – Polymer wastes used in this study: (a) PEX and (b) HDPE

PEX waste is presented in a “flake” shape (Fig. 1a). Its original dimensions, as it was provided, ranged from 0.5 to 10.0 mm. However, due to its lamellar form totally different from that of typical aggregates, which can reduce the mechanical strength of asphalt mixtures, it was decided to mechanically grind the PEX particles (less lamellar particles were obtained with dimensions from 0.5 to 4.0 mm). PEX density was also determined (EN 12697-5 standard), being 938.6 kg/m^3.
HDPE waste (Fig. 1b) is provided with dimensions between 0.5 and 10.0 mm. Since HDPE will be used for bitumen modification, its dimensions can influence de digestion time and the efficiency of the modification process. Thus, the initial dimensions of HDPE were intentionally reduced to 0.5 to 4.0 mm, by using the same mechanical grinder, in order to improve the modification process.

Binder Characterization

All the work of binders and mixtures characterization was developed with a single 50/70 commercial bitumen. The HDPE modified binders were prepared at 180 ºC for two percentages of polymer (3 and 6%) using several digestion times (0.5, 1.0, 1.5 and 2.0 hours).
The PEX waste used in this study is not totally crosslinked, thus being necessary to evaluate its possible interaction with the bitumen during the production of asphalt mixtures. In order to carry out that evaluation, the binder was prepared using the production conditions obtained in the Marshall mix design of the AC 14 Surf 50/70 conventional mixture, i.e. binder content (BC) of 5.0%, together with a PEX percentage of 5.0% (volume ratio between PEX and the all aggregates) determined to properly adjust the grading curve of the mixture with PEX. The binders “modified” with the melted part of PEX were produced for a 2 min “digestion” time (production time for asphalt mixtures), and then a metal mesh was used to filter out the binder, sorting out the crosslinked PEX particles.
In order to classify the binders used in this study, their basic properties were obtained through the EN 12591 standard, namely by carrying out penetration (EN 1426) and softening point (EN 1427) tests. The evolution of the penetration at 25 ºC and softening point of the 50/70 bitumen and the binders modified with 3 and 6% HDPE with the increase of the digestion time is presented in Fig. 2.

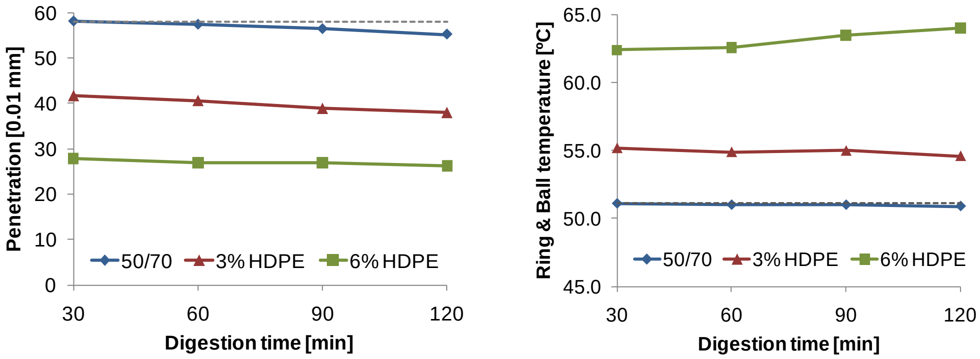

Fig. 2 – Evolution of the penetration and ring and ball temperature with the digestion time

As expected, the higher changes in the properties of the binder were caused by the increase in the quantity of HDPE used (from 3 to 6%). In fact, the binder with 6% HDPE could be classified as a 20/30 binder, increasing the softening point by 13 ºC. The digestion time (30 to 120 min) has little influence in the properties of the binders. The selection of the HDPE modified binder to be used in the following phases (6% HDPE and 60 min of digestion time) of the work was based in these results, as well as on the observation of the binders during their production.
Next, the basic properties of the modified binder obtained by the introduction of PEX as aggregate were determined (Table 1). The filtered binder was obtained after 2 min "digestion" time at 180 ºC.

Table 1 – Comparison between the base properties of binders modified with HDPE and PEX

Type of bitumen	Type and % of polymer	Pen [dmm]	ΔPen [dmm]	ΔPen [%]	A&B [ºC]	ΔA&B [ºC]	ΔA&B [%]
50/70	None	58.1	0.0	0.0	51.1	0.0	0.0
	PEX	37.1	-21.0	36.0	53.7	2.6	5.0
	3% HDPE	41.7	-16.4	28.0	55.2	4.1	8.0
	6% HDPE	27.9	-30.2	52.0	62.6	11.5	22.5

It was observed that only a small part of the SOL content of PEX have melted into the bitumen, probably because the digestion time used was very short (2 min). The penetration results showed that the binder obtained after the mixture with PEX is similar to a HDPE binder modified with 3 to 6% of polymer. The softening point results demonstrated a lower modification grade obtained after the mixture with PEX, even inferior to that obtained with 3% HDPE.

Definition of the Mixtures Production Conditions

The selection of the temperatures to be used in the production of the conventional and modified (PEX and HDPE) mixtures, was based on the reference of EN 12697-35 standard and on the properties of the binders previously presented. Thus, the conventional mixture was produced at 150 ºC, while the polymer modified mixtures were produced at 165 ºC (PEX) and 180 ºC (HDPE).
The conventional mixture (AC14 surf 50/70) was designed according to the grading envelope presented in the national attachment of NP EN 13108-1 standard. By changing the quantities of each aggregate fraction used in the conventional mixture (Table 2), it was possible to define a grading curve that fell inside the limits of the referred envelope (also used in the mixture with HDPE modified binder). However, the grading curve and the quantities used in the mixture with PEX were inevitably adjusted (Table 2) because the PEX waste was applied as a partial substitute of the aggregates (volume ratio of 5%, corresponding to 1.8% in weight due to the low density of PEX).

Table 2 – Materials and quantities used to produce the different asphalt mixtures

Asphalt Mixture	Type of binder	Aggregates used in the mixture [%]				
		Fraction 6/14	Fraction 4/6	Fraction 0/4	Filler	PEX (milled)
Conventional	50/70	46.0	13.0	39.3	1.7	-
HDPE	50/70+6%HDPE	46.0	13.0	39.3	1.7	-
PEX	50/70	46.5	11.3	38.6	1.8	1.8

Next, the optimum binder content of the conventional mixture (5.0%) was determined through the Marshall mix design method, and this value was maintained in the mixtures with HDPE and PEX in order to allow their comparison. Due to the high quantity of polymer waste used in the mixtures with PEX, which partially interacts with the bitumen, it was decided that a complementary study should be carried out according to the national appendix of NP EN 13108-1 standard, namely by using higher binder contents (5.0, 5.5 and 6.0%). In the complementary study, the water sensitivity of the mixtures was evaluated according to EN 12697-12 (Table 3). This test comprises the assessment of the indirect tensile strength (ITS), carried out according to EN 12697-23, of two identical groups of specimens conditioned in different environments (dry and wet), and the evaluation of the ratio (ITSR) between the average strength results of both groups of specimens. The air voids content (EN 12697-8) of the specimens and the deformation on rupture were also determined.

Table 3 – Results of the complementary mix design tests used for the PEX modified mixtures

Evaluated property	BC = 5.0%	BC = 5.5%	BC = 6.0%
Air voids content – Vv [%]	5.0	2.9	1.3
Water sensitivity – ITSR [%]	47.0	57.0	61.0
Indirect tensile strength – ITS_{dry} [kPa]	1655.4	1824.3	1985.6
Mean deformation of dry specimens [mm]	2.4	2.7	2.5

As expected, the mixture with a binder content of 5.0% had higher air voids content, and as result presented higher water sensitivity in comparison with the other mixtures. Although the Marshall mix design have shown an optimum binder content of 5.0%, the results of this complementary study for the mixtures with PEX indicate a higher binder content. Thus, in the following part of the work, two mixtures with PEX will be produced and characterized with binder contents of 5.0 and 5.5%.

PERFORMANCE OF ASPHALT MIXTURES PRODUCED WITH WASTED POLYMERS

This study consists in the comparative evaluation of the mechanical performance of three mixtures with identical composition, which were produced with different constituent materials, namely a conventional mixture, another with HDPE modified binder and the third with PEX substituting part of the aggregates (Fig. 3). The mixtures with PEX were produced with two binder contents (5.0 and 5.5%). Thus, the four mixtures studied in this phase of the work were the following ones:
- Mix A: conventional mixture with 5.0% of a 50/70 bitumen;
- Mix B: mixture with 5.0% of a 50/70 bitumen modified with 6% HDPE;
- Mix C1: modified mixture using milled PEX as aggregate, with 5.0% of a 50/70 bitumen;
- Mix C2: modified mixture using milled PEX as aggregate, with 5.5% of a 50/70 bitumen.

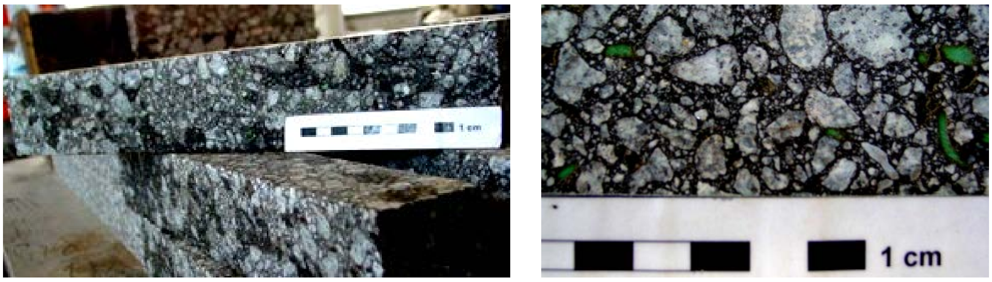

Fig. 3 – Specimens of mixtures C1 and C2 using PEX as aggregates

The maximum density (EN 12697-5) and the bulk density (EN 12697-6) of the asphalt mixtures were determined in order to determine their air voids content (EN 12697-8), as follows:
- Mix A: maximum density of 2442 kg/m^3 and air voids content of 2.0%;
- Mix B: maximum density of 2430 kg/m^3 and air voids content of 3.0%;
- Mix C1: maximum density of 2373 kg/m^3 and air voids content of 3.0%;
- Mix C2: maximum density of 2353 kg/m^3 and air voids content of 2.5%.

The properties of the mixtures with PEX are different from those of the conventional mixture, especially the maximum density that is much lower due to the low density of PEX. The use of this lighten mixtures with PEX can be useful in the transport and in the application on lighten structures (e.g. bridges). Finally, it was observed that the air voids content of the several mixtures is similar.

Water Sensitivity

All mixtures were tested for water sensitivity according to EN 12697-12 standard, previously mentioned, and the results obtained for the mixtures under study can be observed in Fig. 4.

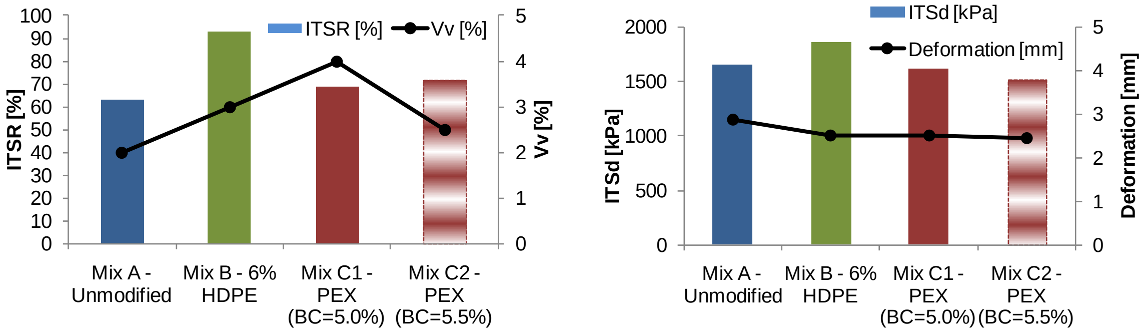

Fig. 4 – Water sensitivity results of the studied mixtures

The mixture with the HDPE modified binder presented better performance than the conventional mixture, whereas the performance of the mixtures with PEX was similar to that of the conventional mixture (mix C2 was slightly less sensitive to water due to its higher binder content).

Permanent Deformation Resistance

In this study, the determination of the resistance to permanent deformation was carried out using the Wheel-Tracking Test or WTT (EN 12697-22). The susceptibility of the mixtures to deform was assessed by measuring the rut depth formed by repeated passes of a loaded wheel at 50 ºC. The evolution of the rut depth (mm) in air of the studied mixtures with the number of load cycles at 50 ºC is presented in Fig. 5, as well as the mean wheel-tracking slope (WTS_{AIR}), in air, which is the result typically used to rank the permanent deformation performance of asphalt mixtures.

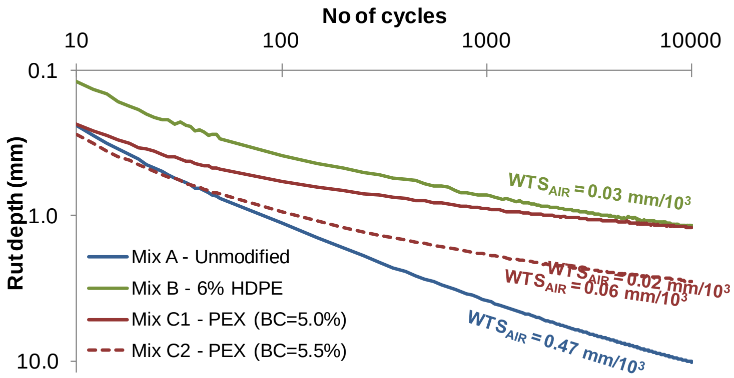

Fig. 5 – Evolution of the permanent deformation of the studied mixtures in the WTT

By analyzing the results of this test it was concluded that the mixtures with polyethylene, used as binder modifier (HDPE) or as partial substitute of the aggregates (PEX) (including mixture C2 with a BC of 5.5%), are much more resistant to permanent deformation than conventional mixture.

Stiffness Modulus

The structural performance of the pavements is directly related to the mechanical behavior of the bituminous mixtures, which can be characterized through the stiffness modulus and phase angle. In this work, these properties were obtained by using the four-point bending beam test, with a repetitive sinusoidal loading configuration, as stated in EN 12697-26. The frequency sweep (0.1 to 10 Hz) tests were carried out at 20 ºC. The variation of the stiffness modulus and phase angle of the studied mixtures with the frequency, at 20 ºC, is presented in Fig. 6.

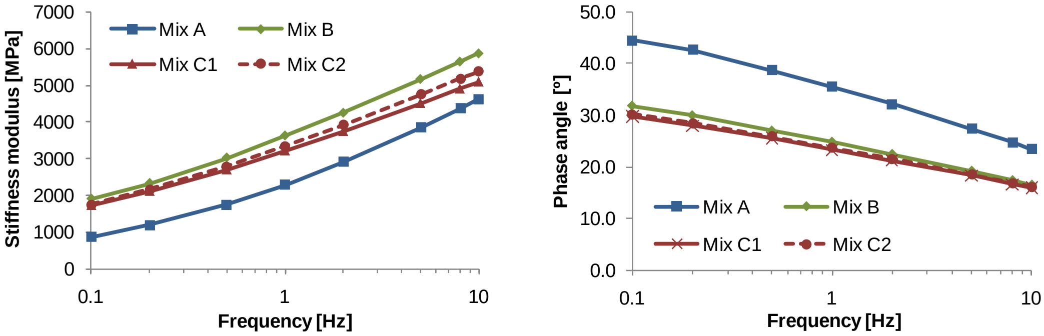

Fig. 6 – Stiffness modulus and phase angle results of the studied mixtures

The evolution of the stiffness moduli with the frequency is similar for all studied mixtures. The mixture with HDPE (mix B) was the one with higher modulus in consequence of the higher viscosity of the modified binder. The conventional mixture was the one with lower modulus, because the use of PEX as partial substitute of the aggregates also increased the stiffness modulus of the mixtures, including in the mix C2 with a higher binder content. The phase angle of the asphalt mixtures with polyethylene based wastes (PEX and HDPE) is clearly lower than that of the conventional mixture, revealing a less viscous behaviour of those mixtures and explaining their higher rutting resistance.

CONCLUSIONS

The main conclusions of this work were the following:

- Several polymer wastes can be used in asphalt mixtures as binder modifiers or as partial substitutes of the aggregates, and the PEX wastes emerged as one of the best solutions due to their low commercial value (HDPE was also selected as a polyethylene based waste alternative);
- The HDPE waste greatly changed the properties of the binder, and the use of PEX waste as aggregates also modifies the bitumen;
- The use of polyethylene based polymers has the advantage of lighten the asphalt mixtures, especially when using PEX, and simultaneously can maintain (water sensitivity) or improve (rutting resistance and stiffness modulus) the performance of those mixture.
- Briefly, the use of PEX / HDPE wastes could be considered a good alternative for paving works.

Acknowledgments

The authors would like to thank Gintegral, Cepsa and Bezerras companies for supplying the materials for this study, which was concluded with the support given by the Portuguese Foundation for Science and Technology (FCT) in the scope of the Strategic Project UI 4047 2011-2012.

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