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Study of the interaction between asphalt and recycled plastics in new polymer modified binders (PMB)

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Abstract

The aim of this study is evaluating the interaction between several base pen grade asphalt binders (35/50, 50/70, 70/100, 160/220) and two different recycled plastics (EVA and HDPE), for a set of new polymer modified binders produced with different amounts of both recycled plastics. After analysing the results obtained for the several polymer modified binders evaluated in this study, including a commercial modified binder, it can be concluded that the new PMBs produced with the base bitumen 70/100 and 5% of each recycled plastic (HDPE or EVA) results in binders with very good performance, similar to that of the commercial modified binder.

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Keywords: Recycled plastics; polymer modified binder; interaction; performance.

1. Introduction

The main material characterized and presented in this study is the asphalt binder, or bitumen. Nearly 95% of the 100 million ton of asphalt binder produced yearly worldwide are applied in paving works, where it serves essentially as binder for the mineral aggregates, in order to produce asphalt mixtures for road paving. Asphalt is traditionally considered as a colloidal system, constituted by molecules of high molecular weight called asphaltenes dispersed in the oil component of the asphalt with lower molecular mass, the maltenes. It is this colloidal structure that defines the rheological properties of the asphalt, ranging from a fluid Newtonian behaviour domain to a solid non-Newtonian behaviour domain [1]. This material derived from crude oil, which was earlier a residue from the petroleum distillation process, is able to

present such different properties. The rheological properties of asphalt also diverge according to the starting crude oil used, the applied refining processes and the possible use of additives [2]. The resulting asphalts are divided into different performance grading categories. Then, the softer asphalts are used for roads in cooler regions, while the stiffer ones are more often used in roads built in warmer regions. However, nowadays the virgin asphalt binders (unmodified) no longer offer enough resistance to support stricter climate demands, the growth in traffic volumes and the increased loads of heavy vehicles. Thus, asphalt modification is the main solution used to solve this problem, assuring the needed durability of world's roads. Among the modified asphalt binders currently used, the polymer modified binders (PMB) are the most widely used because they are less susceptible to temperature changes, and they can improve the performance of asphalt mixtures to meet the road pavement needs [3-5]. However, the use of virgin polymers in PMB can even double the final price of asphalt binders [6]. The use of recycled

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plastics, instead of virgin polymers, can be the answer for that economic issue, being also a better environmental solution for PMB materials. Plastic wastes are already recycled and partly used in new products, but every year millions of ton of plastic are still wasted in landfills [7]. This could motivate the development of new industrial solutions that promote the use of recycled plastics adding value to this wasted material. Thus, the aim of this study is to develop new recycled plastic modified binders with similar or even better properties than the binders currently used (pen grade or commercial PMBs), while evaluating the interaction processes between recycled plastics and different base asphalt binders.

2. Materials and Methods

2.1. Materials

The materials used in this study included different penetration grade asphalts for the modification process, namely 35/50, 50/70, 70/100 and 160/220 asphalts and a commercial polymer modified binder (Elaster® 13/60) used as control material for being the modified binder usually used in Portugal.

The basic properties (penetration, ring and ball temperature and resilience) of the unmodified binders and of the commercial PMB used as control material are presented in Table 1.

Table 1. Basic properties of base asphalt binders used.

Binder	Pen (dmm) (EN 1426)	R&B* (°C) (EN 1427)	Resilience (%) (EN13880-3)
35/50	46	52	9
50/70	61	51	0
70/100	75	49	0
160/220	145	42	0
Elaster 13/60	37	66	21

*Ring and Ball

The recycled plastics selected for asphalt binder modification were the recycled high density polyethylene (HDPE) and ethyl vinyl acetate (EVA). These two materials appear to be promising for binder modification in the literature review, since they presented very good results in previous studies [4]. These recycled plastic materials were also suggested by the supplying waste management company, which had large quantities of HDPE and was only able to supply EVA as an alternative elastomeric waste.

Some important properties of recycled plastics related to their compatibility with the asphalt binder and binder modification level are the melting point (T_m) and the enthalpy of transition (ΔH) [8,9]. These results are obtained with the differential scanning calorimetry (DSC) testing, and are presented in Fig. 1. The melting points of both recycled plastics (EVA 70°C and HDPE 131°C) are below the acceptable temperature for hot mix asphalt production (150°C), which indicates that they are in fluid/liquid state, as needed, at the production temperature. Moreover, the HDPE presents a higher enthalpy of transition (ΔH) than EVA, as it was expected, since it has a more difficult digestion and hence it shows lower compatibility than EVA with the bitumen.

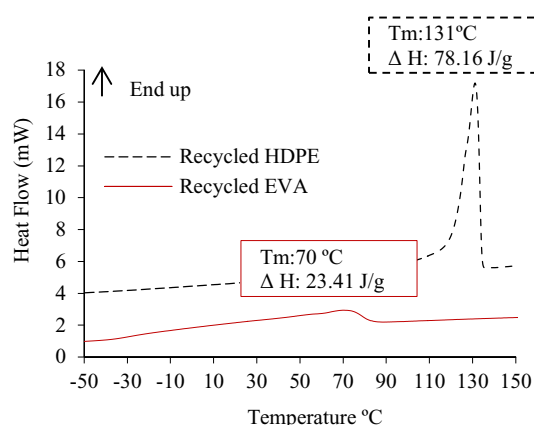


Fig. 1. Calorimetric properties of EVA and HDPE.

2.2. Methods

First, the conventional physical characterization of all binders was performed with needle penetration (Pen) at 25°C (EN 1426), softening point or ring and ball temperature (R&B) (EN 1427) and resilience (EN 13880-3) tests. These tests were carried out in the base and control binders, as well as in the new binders developed during this study.

The differential scanning calorimetry (DSC) tests were performed with the recycled plastics but also with the produced modified binders, in order to evaluate the changes in terms of thermal behaviour of the recycled plastics once dispersed into the asphalt binder. The DSC was carried out in a DSC Diamond Pyris. About 10-20 mg of sample was sealed in an aluminium capsule, and then the following heating and cooling protocol was applied: two heating and one cooling cycles ranging from -60°C to 180°C with a rate of 10°C/min. The first heating cycle provided the same thermal history for all the samples.

The morphological structure of the binders is also very important to evaluate the interaction between the asphalt and the recycled plastics [10] and can be useful to conclude about the storage stability of the binders. Thus, after production, some samples of the binders were stored for 48h at 165°C in an aluminium tube. After cooling, small samples of the binders from the bottom and the top of the tube were recovered, and reheated in a glass slide, to prepare a plane surface for the microscopic assessment. The analysis of the morphologic structure was made by fluorescence microscopy (FM) in an Olympus BH2-RFCA microscope. FM is based on the principle that polymers swell due to the absorption of some fluorescent constituents of the base bitumen, and thus the bitumen-rich phase appears dark, while the polymer-rich phase appears light [8].

Finally, the rheological properties of the asphalt binders were evaluated. Although there are several methods for determining those rheological properties, the oscillatory tests tend to be the best techniques to represent and assess the behaviour of asphalt binder [11] used in road pavements. The rotational rheometer used was the TA instruments AGR2, operating under strain control. The test geometries were plate-plate with 40 mm diameter and 1 mm gap. The dynamic measurements were made at temperatures ranging from 30 to 80°C, while the frequencies ranged from 100 to 0.01 Hz in order to construct the master curves of the dynamic material functions. The maximum strain was kept below the limit of the linear viscoelastic region. The validity of time-temperature superposition principle (TTSP) for rheological simple materials facilitates the production of master curves at the reference temperature (60°C) by shifting the frequency sweeps measured at different temperatures along the frequency axis to form a continuous curve.

3. Results

3.1. Physical characterization

The conventional physical characterization results of the produced binders are presented separately for each polymer. The results obtained after mixing several base asphalt binders and different amounts of EVA are presented in Fig. 2, while Fig. 3 presents the results of binders produced with HDPE. The results of the control PMB material are also presented.

It can be observed that the properties of some of the new asphalt binders produced with recycled EVA are

similar, or even better, than those of the commercial PMB (Elaster®).

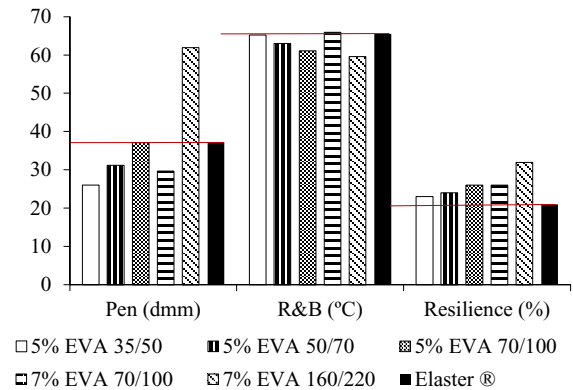


Fig. 2. Basic properties of EVA modified binders.

In fact, EVA based binders have higher resilience (due to EVA elastic properties) and similar softening point, and both properties are related to a good rutting resistance of the road pavement. Since these results are similar to those of the commercial PMB, the penetration values would help to select the best EVA modified binder to use in the next phase of the work. Thus, between the several binders produced, the binder with 5% of EVA and a base binder 70/100 was selected, for being the one with a penetration value nearest to that of Elaster®.

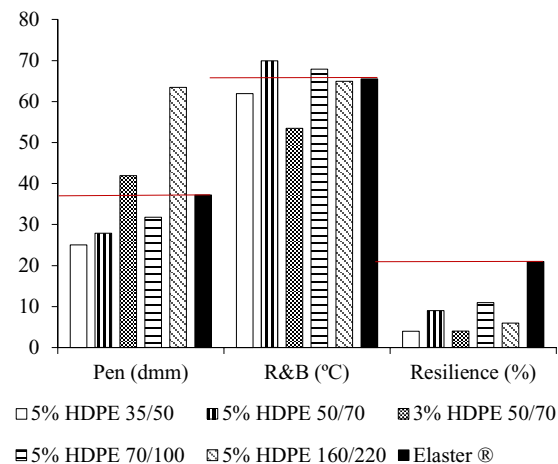


Fig. 3. Basic properties of HDPE modified binders.

Regarding the binders produced with HDPE, and for comparison reasons, the binder produced with 5% recycled HDPE and a base binder 70/100 was also selected. Moreover, among the HDPE based binders produced, this binder presented the highest resilience

and softening point and penetration values similar to those of the commercial PMB.

Summing up, this study will continue with these selected binders:

- 5% EVA with a 70/100 pen grade binder;
- 5% HDPE with a 70/100 pen grade binder.

3.2. Thermal properties

In order to understand the changes induced by the interaction between the 70/100 bitumen and the EVA and HDPE polymers, the resulting modified asphalt binders were tested in DSC. The DSC results of the EVA and HDPE modified binders, as well as the results of the 70/100 base bitumen, are shown in Fig. 4.

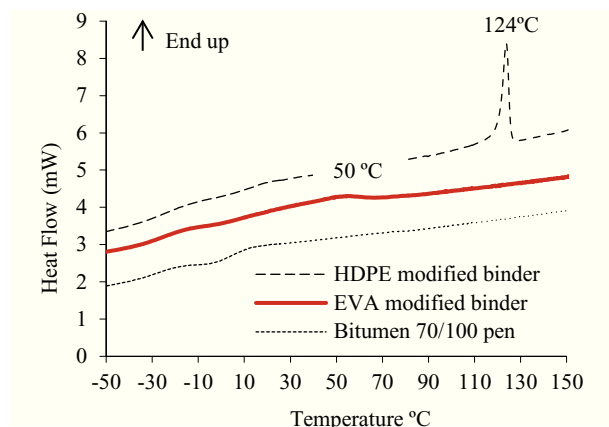


Fig. 4. Calorimetric properties of 70/100 pen bitumen and EVA and HDPE modified binders.

The DSC results show that melting points are perceptible in the modified asphalt binders, as expected, while the base bitumen reaction to heat is typical of an amorphous material (without melting points).

In fact, the fusion of the crystalline parts of the plastics (EVA and HDPE) is visible in DSC results of the new binders. However, comparing these melting peaks with those of recycled plastics before interaction, it can be concluded that the melting point of the crystalline parts of the plastics decreases 20°C for EVA and 7°C for HDPE, which meets with other results found in the literature [12]. This decrease in the melting point is a clear sign of the interaction between the recycled plastics and the base asphalt binder and it is also good for the binder workability in the asphalt mixture production and application.

3.3. Morphological structure

In order to evaluate the variation of the dispersion of different recycled plastics into the 70/100 bitumen, and their storage stability, fluorescent microscopy (FM) observations were carried out. The FM images, related to the EVA modified binder, are presented in Fig. 5, and those related to the HDPE modified binder are presented in Fig. 6.

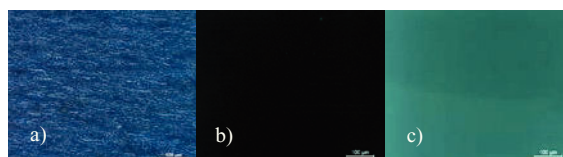


Fig. 5. Fluorescence micrographs of: a) recycled EVA modified binder, b) binder recovered after modification, and c) polymer recovered after modification.



Fig. 6. Fluorescence micrographs of: a) recycled HDPE modified binder, b) binder recovered after modification, and c) polymer recovered after modification.

FM images presented previously are from the whole modified binder (a), and from the bottom (b) and top (c) parts of the binder stored in the aluminium tube (storage stability test).

Comparing the images b) and c) of Figs. 5 and 6, it is clear that phase separation occurs during the storage period, for both binders, meaning that these binders must be produced or remixed immediately before use, otherwise they will not be homogeneous and their performance may be compromised.

The images a) of Figs. 5 and 6 appear to show that the dispersion processes of EVA and HDPE in asphalt are different, although both binders are using the same amount of polymer. In fact, a more homogeneous dispersion can be observed for recycled EVA. As the base bitumen and the polymer amount used in both binders are the same, the binder morphology changes may be related to the different swelling potential of the EVA and HDPE polymers. The different morphology of both polymer modified binders may affect their rheological characteristics.

3.4. Rheological characteristics

The PMB Elaster® and a conventional 35/50 pen grade asphalt binder (the unmodified binder mostly used in Portugal) were used as control materials for rheological evaluation of EVA and HDPE modified binders. Their complex moduli are shown in Fig. 7 and the corresponding phase angles in Fig. 8.

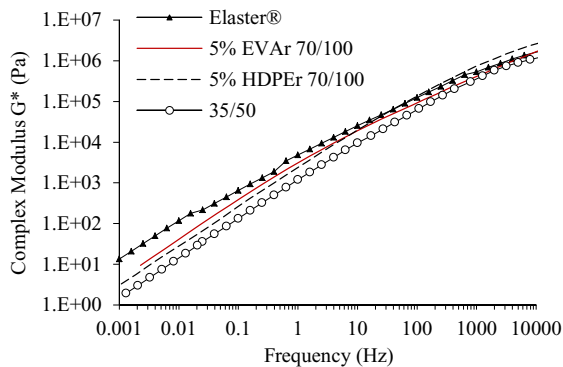


Fig. 7. Master curves of the complex modulus of the binders.

The complex modulus results at high frequencies, or low temperatures, indicate that the HDPE binder has the highest complex modulus, whereby it should be expected that the resulting asphalt mixtures might be very stiff. At low frequencies (high temperatures), all modified binders presented complex modulus values, higher than those of conventional 35/50 pen grade binder, and thus it is expected that asphalt mixtures produced with these PMBs might have a great rut resistance at high temperatures.

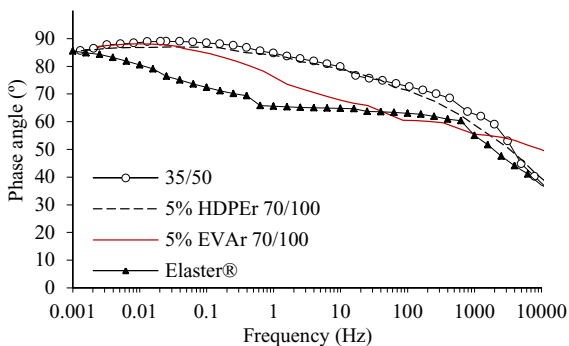


Fig. 8. Master curves of the phase angle of the binders.

The phase angle is an indicator of the ratio between the elastic and viscous properties of a binder. In Fig. 8 it can be seen that Elaster® and EVA binder have lower phase angle values in a wider range of frequencies and temperatures, which means that these asphalt binders may provide a more elastic behaviour

and less viscous deformation under various conditions. The 35/50 and HDPE binders have higher phase angle values resulting in a more viscous behaviour.

4. Conclusions

After analysing all the results of the several polymer modified binders evaluated in this study, including the commercial modified binder, it can be concluded that the new PMBs produced with the 70/100 base bitumen and 5% of each recycled plastic (HDPE or EVA) have a very good performance, similar to that of the commercial modified binder. This result is a consequence of an easy interaction observed between the softer bitumens and both recycled plastics.

According to the DSC results, all the materials used in the binder are in a liquid state at high production temperatures, promoting the needed workability and homogeneity of the binder.

Concerning the morphology of the binders, a more homogeneous dispersion was observed for binders with EVA, but none of these new PMBs are sufficiently stable to be stored at high temperatures.

Finally, and according to the rheological results, it is expected that these new PMBs may result in asphalt mixtures with better rutting performance. Furthermore, HDPE modified binder may result in a very stiff mixture, while EVA modified binder may result in a mixture with better fatigue resistance due to its elastic behaviour.

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