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# Developing enhanced modified bitumens with waste engine oil products combined with polymers

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## HIGHLIGHTS

- Bitumen was partially replaced by waste engine oil or recycled engine oil bottom.
- Different waste and virgin polymers can be used to improve binder performance.
- The new modified binders are very flexible but also stable at high temperatures.
- Low non-recoverable creep compliance values were also obtained with these binders.
- New environmentally friendly solutions were developed for pavement materials.

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## ABSTRACT

The reduction in the use of natural and finite resources and the reuse of waste materials are current concerns of different research areas. Therefore, the main aim of this study is to develop enhanced modified bitumens with waste engine oil and recycled engine oil bottoms combined with polymers (waste polyethylene, crumb rubber and styrene-butadienestyrene), in order to minimize the use of bitumen. After a thermochemical characterization, different compositions were studied with penetration, softening point and viscosity tests. The bitumens modified with waste engine oil products and polymers have similar penetration values, and softening point temperatures higher than those of commercially modified binders. A rheological analysis of the most encouraging solutions showed low thermal susceptibility, high values of high temperature PG and low non-recoverable creep compliance values, which could indicate a promising performance using high amounts of waste materials.



## 1. Introduction

The road paving industry uses a high amount of natural and finite resources, such as bitumen and aggregates, for construction and conservation of road infrastructures [1]. In fact, in the construction sector, the paving industry is one of the highest consumers of fossil fuel, namely for production of bitumen for asphalt mixtures [2]. Furthermore, asphalt plants are considered significant sources of pollution [3], with a high level of greenhouse gas emissions [2].

Thus, waste or recycled materials could partially substitute the raw materials used in asphalt mixtures with some important advantages, namely by reducing the landfill volumes and the amount of natural resources consumption and consequently the need for more extraction [4]. The introduction of several waste materials in asphalt mixtures has already been studied in the past years and their properties were compared to the specification limits, which were usually fulfilled. Moreover, their performance can be improved after waste modification [5], with specific advantages of each solution as presented in the following paragraphs.

Plastic and crumb rubber waste materials have been used for bitumen modification to improve its characteristics [6–9]. The incorporation of plastic wastes in bitumen reduces its penetration values and increases its softening point temperature. The stability and durability of the corresponding asphalt mixtures is improved, namely through the increase of permanent deformation and fatigue resistance [10,11]. Furthermore, some plastic wastes are

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available in large amounts at a reduced price [12], which could be used for bitumen modification. Likewise, crumb rubber incorporation in bitumen increases the softening point temperature and viscosity, reducing the penetration and thermal susceptibility [13,14]. Moreover, the asphalt rubber mixtures can significantly improve the permanent deformation resistance and increase the fatigue resistance at low temperatures [15,16], which could increase the pavements durability, although they typically use higher amounts of binder. This waste material is considered a serious environmental problem, as a result of a fast accumulation and difficult elimination [17], and new solutions for its reuse would be certainly well accepted.

Furthermore, several studies have mentioned the use of waste engine oil and recycled engine oil bottoms or re-refined engine oil bottoms (REOB) as bitumen extenders or modifiers [18–20]. The addition of waste motor oil is used to increase penetration and decrease softening point temperatures and viscosity of a bitumen, thus reducing production and compaction temperatures of asphalt mixtures. However, as a result of its incorporation, the elastic recovery and permanent deformation resistance may be compromised [21], which demands for a careful mix design of these materials. On the other hand, the recycled engine oil bottoms is the non-distillable waste from the recycling of waste engine oil by atmospheric distillation [18,22] and it is used as a binder additive in the United States and Canada due to its low cost and increasing availability over the years [23]. The asphalt mixtures with this waste material showed similar or higher resistance to permanent deformation, water susceptibility and fatigue cracking, when compared with conventional mixtures [24]. Nevertheless, the modified binder with recycled engine oil bottoms exhibited excessive aging, which could lead to premature cracking in pavements [25,26].

Both waste engine oil and recycled engine oil bottoms can be used as binder extenders, but their incorporation rates are usually low in order to guarantee an adequate performance of the asphalt mixtures [18,23,27]. However, according to Jia et al. [21] the waste engine oil products could have a higher commercial value for the road paving industry should they be used as a partial substitute of bitumen, especially if the problems resulting from their incorporation could be minimized or eliminated. Taking into account that polymers are used to solve some of those problems, the bitumen modification with waste engine oil products and polymers could be a promising, environmentally friendly and economic solution for paving industry.

Thus, the main aim of this study is to maximize the use of waste engine oil and recycled engine oil bottoms, combined with different polymers, in order to reduce the amount of base bitumen through the addition of waste materials. Furthermore, these new modified binders should maximize the use of waste materials without compromising their performance, i.e., they should present similar or higher properties than a commercial polymer modified bitumen (PMB) in order to assure equivalent rheological performances.

## 2. Materials and methods

### 2.1. Materials used in this study

Two waste engine oil products were used in this study to partially substitute bitumen. One of them was a waste engine oil (EO) from heavy and light vehicles, which was not subjected to any kind of treatment and was supplied by a certified waste treatment company (*Sogilub, Lda.*). The other product is the waste material generated from the recycling process of the EO, known as recycled engine oil bottoms (RB), and was supplied by the same waste treatment company. This material is an oil with higher viscosity because a large part of the light compounds were removed during the recycling process of the EO. Thus, it basically comprises the heavy compounds of the EO.

The base bitumen used as base for this study is a 35/50 penetration grade binder (B35/50), according to EN 12591 standard, supplied by Cepsa Portugal. This bitumen and a commercial PMB (PMB45-80/60, according to EN 14023 standard) were used as references for comparison purposes.

Three different polymers were used in this work to allow higher rates of EO or RB incorporation in final binder. The selected polymers are a high density polyethylene (HDPE) from plastic waste recycling, a crumb rubber (CR) from waste tires and the most commonly used polymer for bitumen modification, i.e., styrene-butadienestyrene (SBS). HDPE and SBS polymers were supplied by Gintegral S.A., while CR was supplied by Recipneu Lda.

### 2.2. Methods

#### 2.2.1. Characterization of waste engine oil and recycled engine oil bottoms

The waste EO and RB materials were mechanically tested to evaluate their rheological properties. Then, they were thermally characterized in order to assess the loss of volatile compounds when submitted to high temperatures. Finally, they were chemically characterized with the main purpose of identifying their chemical constitution and the presence of heavy metals and other contaminants. These materials were compared to the base 35/50 bitumen, which is the reference material being substituted by EO and RB.

First, dynamic viscosity tests using Brookfield Rotational Viscometer were carried out to characterize the rheology of both EO and RB, according to EN 13302 standard, over a range of temperatures (30–180 °C), with 10 °C increments. The base bitumen (B35/50) was also evaluated in this range of temperatures for comparison purposes, using the same equipment for temperatures above 100 °C, and a dynamic shear rheometer (DSR) equipment for temperatures below 100 °C.

Thermogravimetric analysis (TGA) was also carried out to obtain the mass loss of those materials with the increase of temperature, in order to evaluate the possibility of volatile compound loss at asphalt mixing temperatures. In this test, a sample of 7 mg is placed in a sealed aluminium capsule, which is submitted to a heating rate of 10 °C/min, in a temperature range of 40 °C–750 °C, in a nitrogen atmosphere, which weight is continuously monitored with a high sensitivity balance.

The chemical analysis was carried out through Fourier Transform Infrared Spectroscopy (FTIR) to identify the different compounds and functional groups of the tested materials. Before testing, the EO and RB were placed in a vacuum oven in order to eliminate possible water from the samples. Then, the samples were dropped onto potassium bromide plates and analysed in a FTIR Jasco equipment, in wavenumbers ranging from 4000 to 440 $cm^{-1}$, with 32 scans. The base bitumen was dissolved in chloroform with 10 wt% concentration, and later dropped onto the same plates previously mentioned.

Finally, the analysis of heavy metals and other contaminants in the RB, the EO and the base bitumen was carried out by Inductively Coupled Plasma spectroscopy (ICP), according to D5185-09 standard, in an accredited external laboratory from Canada (AGAT laboratories). This test allowed quantifying the heavy metals and other contaminants present in those materials.

#### 2.2.2. Characterization of polymers

The characterization of the polymers used in this study was made regarding their dimension and thermal behaviour by differential scanning calorimetry (DSC) tests and thermogravimetric analysis (TGA).

The differential scanning calorimetry test identifies melting and/or glass transition temperatures [28]. In this test, a sample of 10–20 mg is placed in a sealed aluminium capsule, which is submitted to a heating rate of 10 °C/min, in a temperature range of −60 °C–160 °C, over two heating and cooling cycles. The thermogravimetric analysis was used to determine the mass loss of the polymers at bitumen mixing temperatures.

#### 2.2.3. Production of modified binders

In a first phase of the study, the partial substitutes (EO and RB) were blended with the base bitumen (B35/50) using a low shear mixer, during 20 min, at the production temperature of the base bitumen (150 °C). Samples were taken at this stage for further characterization. The initial contents of EO and RB used in this phase were 10% each, which is higher than the common contents used for rejuvenating or as extenders. However, higher amounts of these materials were later used together with polymer modification to maximize the use of these waste materials as partial substitutes of bitumen.

After that, the binders with EO or RB were modified with polymers using a high shear mixer, during 20 min, at a temperature of 180 °C. This new mixer was used to obtain a better dispersion of the polymers in the binder. The polymer contents depend on the type of polymer used, bearing in mind the goal of maximizing the use of waste materials. According to the suggestion of different authors [14,29,30], a rate of 20% of crumb rubber incorporation was selected. The most usual contents of the other types of polymers used for binder modification are 5–6% [31]. Thus, the contents of SBS and HDPE used in this study were 5 and 6%, respectively. The amount of EO and RB used in this phase were defined in order to achieve a polymer modified binder with properties similar to those of the control PMB45-80/60 binder, maximizing the use of waste materials.