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Influence of the geometrical and physical properties of filler in the filler–bitumen interaction

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HIGHLIGHTS

- Hydraulic and hydrated limes are composed of granulous-shaped particles.
- Cement and natural filler particles are mostly angular with smooth to rough surfaces.
- The Rigden voids of hydrated lime are much larger than those of the other fillers.
- The *RV*, the *PA* and the $\Delta T_{R\&B}$ test results show a strong inter-relation.
- It is proposed the inclusion of the f/b_{max} ratio in filler specifications.

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ABSTRACT

The objective of this laboratory study is to investigate the relationship between the geometrical and physical properties of filler and the consistency of filler–bitumen mixtures and the stripping resistance. Five fillers (3 manufactured and 2 natural) were geometrically and physically characterized with various test methods, including SEM imaging. The filler–bitumen interaction was assessed with two consistency-related mastic tests and a water susceptibility test.

SEM images show significant differences in size, shape and surface texture between tested fillers, and a strong correlation with the stiffening effect. It is proposed the inclusion of the f/b_{max} ratio, obtained from the absorbing capacity test, in the filler specifications to serve as an indicative threshold value of the filler content added.

1. Introduction

The in-service performance of bituminous mixtures is related with the resistance to cracking, rutting, stripping, and other asphalt pavement distresses, and all depend on the cohesion and adhesion properties of the mastic [1–4]. The filler is a key element for the mastic behavior, from the mixture production phase to the construction of pavement layers. This very fine aggregate, be it natural (dust of mineral aggregates) or manufactured (e.g. Portland cement), causes, primarily, the increase of the binder stiffness. Although the mastic-to-bitumen relative stiffening is mainly related with the filler-to-bitumen content ratio (*f*/*b*), many research studies carried out during the 20th century concluded that it depends on the properties of both materials (filler and bitumen) and their specific interaction [5,6]. The literature refers to the importance of the physicochemical nature, the particle size and shape, the surface energy, etc. [7]. The filler–bitumen bond is related to the adsorption and absorption of selected bitumen group elements at the interface. With siliceous fillers, the bond is mechanical (Van der Walls forces), with low strength, while with others types of fillers (e.g. lime) it is chemical, with high strength. In opposition, Clopotel et al. [8] concluded that the adsorption of the bitumen elements is proportional to the surface area of the filler and it is not affected by the chemical composition.

Hesami et al. [6] present a review on the theory of suspensions, dating back to Einstein's studies, and the application to the mastic, a suspension of filler particles in bitumen. Hence, the authors propose a general model that combines the Einstein and Frankel models for hydrodynamic conditions, as follows:

$$\eta_r = 1 + k \cdot \phi \quad \text{if } h \gg r \qquad (1)$$

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$$\eta_r = c'\left[\frac{\left(\frac{\phi}{\phi_m}\right)^{1/3}}{1-\left(\frac{\phi}{\phi_m}\right)^{1/3}}\right] \quad \text{if } 2\delta < h \le 2\zeta \tag{2}$$

and a frictional model

$$\eta_r = \left(\frac{\delta}{r} - h_r\right)\cdot N_c \cdot C_1 + N_c^p \cdot C_2 \quad \text{if } h \le 2\delta \tag{3}$$

where, η_r is the relative viscosity of the suspension (ratio of suspension and fluid viscosity); ϕ is the particle concentration; ϕ_m is the maximum particle concentration which the viscosity tends to infinity; h is the distance between effective particles (particle plus the adsorbed layer of fluid); r is the particle radius; δ is the adsorbed fluid layer thickness; ζ is the thickness of the adsorbed and the influenced layer of fluid; N_c is the number of particles; N_c^p is the number of particles in "primary structure" (filler fraction with more influence on friction); h_r is the relative distance between particles, taken as h/r; k, C', C_1, C_2 are constants related with the filler particles' characteristics and the specific bitumen–filler interaction. This model fitted the mastic viscosity data well from three different studies found in literature.

Following the same objective, Faheem [5] modeled the mastic relative stiffness, G_r (ratio of mastic and bitumen shear modulus), considering two different behavioral regions: (i) diluted, where the stiffness increases linearly with the filler concentration; (ii) concentrated, where filler particles start interacting. The final model is

$$G_r = G_1 + a_1\cdot(\phi_r - \phi_c) + G_2\cdot(a_2 - a_1) \cdot \ln\left(1 + \exp\left((\phi_r - \phi_c)/G_2\right)\right) \tag{4}$$

where, ϕ_r is the filler volume concentration; ϕ_c is the critical volume concentration; a_1, a_2, G_1 and G_2 are constants. The author modeled the values of the constants as a function of the filler and bitumen properties, namely the Rigden voids, the methylene blue value, the calcium oxide content, the bitumen asphaltene content and the bitumen shear modulus.

The stripping phenomenon in asphalt is directly related with the adhesive failure between the bitumen, or the mastic, and the aggregate. In literature, water/moisture penetration in the mastic-aggregate interface is considered the main cause for this distress type. Based on the experimental results of an adhesion test method developed by the author, Jakarni [1] concluded that the mineral filler has an important influence on the mastic adhesion strength. The basic fillers (hydrated lime and limestone) showed to significantly improve the tensile bond strength of the mastic when compared with the bitumen while the acidic filler (gristone) addition resulted in minimum effect.

Most specifications and guidelines for the bituminous mixtures design used worldwide only define a rather broad range for the filler-to-bitumen content ratio and simple requirements for the filler, namely the material source and grading [9]. However, the European standard for aggregates for bituminous mixtures EN 13043 [10] specifies geometrical, physical, chemical and stiffening effect requirements for the filler. Thus, the NCHRP Project 9-45 [9] identified the Rigden voids, the size distribution, the calcium oxide content, the active clay content and the particle density as the filler properties with the closest relation with the asphalt performance. Therefore, the researchers concluded that f/b mass ratio limits are insufficient to describe the effect of filler concentration on mastic and asphalt behavior. The current specifications for paving materials used in Portugal [11] specifies the f/b volume ratio with

$$f/b = \frac{(100 - RV)\cdot \Delta T_{R\&B}}{1021.2 + \Delta T_{R\&B}\cdot v} \tag{5}$$

where, RV is the filler Rigden voids (%); $\Delta T_{R\&B}$ is the softening point increase (from bitumen to mastic) (°C). This equation was developed at the Belgium Road Research Centre (BRRC) to predict the stiffening effect of the filler on the mastic [12], stating that the mastic behavior is optimized when $\Delta T_{R\&B}$ is between 12 and 16 °C.

The concept of the fractional voids, or Rigden voids, was proposed by Rigden in 1947 [13]. The volume of voids in the dry compacted filler sample (fractional voids) corresponds to the volume fraction of bitumen that is fixed and only the fraction in excess of this volume is able to lubricate the filler matrix [5,14]. The larger the Rigden voids of a certain filler, the larger the amount of bitumen needed to make the mixture fluid. The way the filler particles form a denser or looser volume is directly related with the particle shape and size distribution in the filler. Rigden also stated that the chemical differences between fillers were not very important for the mastic behavior.

This paper investigates the relationship between the geometrical and physical properties of fillers and the bitumen–filler interaction. Several fillers from different sources were characterized with various laboratory tests, including SEM imaging, and compared against the anti-stripping resistance and consistency of the mastics. From the results obtained some suggestions are made regarding changes to filler specification for bituminous mixtures.

2. Materials and experiments

2.1. Materials

For this study, five fillers were selected given the materials that are available commercially and the industry practice in the country. These were: hydraulic lime (H); hydrated lime (HL); Portland cement (PC); limestone filler (L); basaltic filler (B). With the exception of hydrated lime, the materials selected are allowed by the Portuguese specifications for paving materials to be used as filler in bituminous mixtures [15]. Limestone-based fillers are the most commonly used in the country, especially with granitic-based aggregates. The selected limestone filler is the reference material for the present study, which is a commercial product obtained from crushed limestone. In contrast, the basaltic filler was obtained directly from the dust collection system (baghouse) of an asphalt plant of a local producer that uses basaltic aggregates. The hydraulic lime is a natural lime produced in Portugal, designated as NHL5 in accordance with EN 459-1 [16]. Grilo et al. [17] state that NHL produced in Portugal, in accordance with EN 459-1 [16], are relatively new binders and there is not sufficient knowledge and (or) experience of their use. The hydrated lime is a calcium lime CL-90S [16], with more than 93% of $Ca(OH)_2$, and is also produced in Portugal.

A paving grade 35/50 bitumen was used in the mastics of this research. Table 1 shows the main properties of the bitumen. The penetration and softening point tests were repeated in the laboratory considering that one of the mastic tests is the delta ring and ball test. Complete information regarding the materials and experiments is available in [18].

Table 1
Bitumen properties.

Property	Test method	Units	Results
Penetration	EN 1426	0.1 mm	44
Softening temperature	EN 1427	°C	55.5
Kinematic viscosity 135 °C	EN 12595	mm^3/s	539
Solubility	EN 12592	%	100.0
Flash point (open cup)	EN ISO 2592	°C	351
Penetration index	EN 12595	–	−1.4
Fraass breaking point	EN 12593	°C	−7
Paraffin content	EN 12605-2	%	<3
Mass variation	EN 12607-1	%	0.1
Retained penetration (resistance to hardening, at 163 °C)	EN 1426	%	77