RESEARCHING THE EFFECTS OF DIFFERENT AGGREGATES AND BITUMEN TYPES ON ASPHALT MIXTURES

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ABSTRACT
The main purpose of this research is to analyse the influence of different natural aggregate sources, as well as different bitumen types, on the adhesion and affinity between the two materials and on the properties of stone mastic asphalt (SMA). The aggregates used in this study were both fine (sand 0/4) and coarse (4/8 and 8/16), extracted from four different natural rock quarries in Romania, selected after a geographical, geological and geomorphological analysis. From a petrographic point of view, the rocks extracted in the proposed quarries are igneous. Adhesion and affinity tests on bitumen coated aggregates were performed, while also employing an anti-stripping agent in the process. Regular paving grade bitumen 50/70, as well as polymer modified bitumen (PMB) 45/80-65, were used. All tests have shown that using an anti-stripping agent improves both bitumen adhesion to natural aggregates and the affinity between the two. Afterwards, laboratory tests on SMA samples, prepared using the same types of bitumen, aggregates and additive, were performed. Although the results were variable and no general conclusion could be drawn, they were clearly influenced by the shape and nature of the mineral aggregates, as well as by the bitumen types and grades.

Keywords: natural aggregates, bitumen, adhesion, asphalt mixtures, additives.

INTRODUCTION
Asphalt mixtures are composite building materials which consist of a mineral skeleton (natural aggregates and filler) mixed with a bituminous binder, following a recipe and using an adequate technology. Bitumen is a complex and highly viscous colloidal suspension of asphaltenes in a continuous phase of saturated paraffins, aromatic oils and resins, present in natural deposits or produced from residues obtained during crude oil processing [2][7]. Other materials may also be added to the mixture, in order to improve certain characteristics which influence its behaviour in operation. Asphalt mixtures are widely used worldwide as pavement construction materials.

The strength, resistance and durability of an asphalt mixture are influenced by several factors, including: bitumen chemistry, bitumen cohesional resistance, aggregate mineralogy, aggregate surface texture and particle shape, aggregate frictional resistance,
absorption, aggregate surface coating, cyclic loadings and abrasion due to traffic, interaction between constituent materials, and weather conditions [1][2][7]. Most natural aggregates have polar (electrically charged) surfaces. On the other hand, bitumen is a hydrocarbon product with polar activity lower than water, which implies that the latter tends to wet aggregates much more easily than bitumen would. The bitumen-aggregate bond is mainly based on weak dispersion forces and it is much weaker than water-aggregate bonds [3]. However, a good bond should develop between aggregates and bitumen, in which case failure should occur within the binder. Inadequate or incomplete aggregate coating in bitumen during asphalt mixing operation, as well as adhesion loss at the aggregate-binder interface or cohesion failure within the binder during service may lead to pavement distresses (e.g. cracks, potholes, rutting, bleeding) or premature mixture failure [5][8]. The breaking of the adhesive bond between the aggregate surface and the bituminous binder is commonly known as stripping. Usually, the presence of moisture is the common factor to all stripping [8].

Studying and understanding the aggregate-bitumen adhesion mechanisms is important to ensure adequate pavement resistance and durability, with direct effect on its behaviour in service and, consequently, maintenance and rehabilitation costs. Optimum materials selection also plays an important role; however, this procedure is extensively influenced by transportation costs, building costs and other economic aspects.

Natural aggregates with high silica content exhibit acidic behaviour, which has a negative influence on the chemical bonding with bitumen. Various minerals like albite, quartz, and feldspar are detrimental in terms of asphalt moisture sensitivity [10]. On the other hand, siliceous aggregates are harder and more resistant than basic aggregates, with high CaCO₃ content. Aggregates with pores on exposed surfaces, such as limestone, have been found to establish stronger bonds with bitumen and perform better against moisture attack than aggregates with fewer or smaller surface pores, such as quartz [8]. Also, aggregate gradations were found to influence stripping resistance [1].

Mineral fillers (powders with all particles passing the 0.063 mm sieve), such as hydrated lime, Portland cement, limestone or fly ash, have a positive influence on the aggregate-bitumen adhesion [5]. Anti-stripping agents improve the adhesion between the bitumen film and the crushed aggregates, by creating strong chemical links. Applying such additives leads to improved resistance to water and decreased bitumen surface tension [6], increasing the energy required to separate bitumen from the aggregate unit surface [3].

SMA is a mixture with robust stone skeleton, stabilised with bituminous mastic, with filler and cellulose fibres. It is an asphalt mixture recommended and widely used for superior technical class roads.

**MATERIALS**

The natural aggregates used in this study were extracted from four different natural rock quarries in Romania (Table 1), selected after conducting a geographical, geological and geomorphological configuration analysis of Romania’s territory. Furthermore, the selection was influenced by several issues (i.e. low rock density, high Los Angeles abrasion and terrace extraction sites) which restricted the quarry selection options.
From a petrographic point of view, the aggregates used in this study are igneous (Table 1), formed through the cooling and solidification of magma derived from Earth’s superior mantle. They are the result of a sequential natural magma crystallisation, which determines the rock structure and texture, depending on the cooling time.

### Table 1. Natural aggregates

| Source no. | Source abbrev. | Rock nature | Location in Romania |
|------------|----------------|-------------|---------------------|
| 1          | Q_1            | andesite    | North-West          |
| 2          | Q_2            | andesite    | Centre              |
| 3          | Q_3            | dacite      | North-West          |
| 4          | Q_4            | diabase     | South-East          |

Diabase is a fine-grained rock, composed mostly of pyroxene and feldspar. It resembles andesite in several aspects, including hardness, both rocks being much more resistant than dacite. However, they are all durable rocks. The composition of dacite is an intermediate between andesite and rhyolite.

The aggregates used in this study were both fine (sand 0/4) and coarse (4/8 and 8/16), consisting of polyedric stones obtained through crushing and sorting of the extracted mineral deposits. The quarry test reports indicated that the aggregates geometrical and physio-mechanical properties met the standardised technical quality requirements.

The filler used in this study to prepare asphalt mixtures had a minimum of 90% CaCO_3 content and met the qualitative requirements (e.g. gradation, water content, solubility).

The bitumen penetration grade is defined as the distance (in tenths of a millimetre) travelled by a calibrated needle into a bitumen sample, under a known load, at a known temperature, for a known time [2]. Two bituminous binders were used in this study: a regular, straight-run, 50/70 grade bitumen (subsequently referred to as SRB), as well as a PMB 45/80-65. Both were manufactured and provided by Orlen Asfalt Sp. z o.o. Plock, Poland.

With the purpose of improving the bitumen adhesion to natural aggregates and obtaining better asphalt mixture performance characteristics, an anti-stripping additive (provided by Orlen Asfalt Sp. z o.o.) was also employed in this research.

**METHODOLOGY**

Adhesion tests were performed using a spectrophotometer. The procedure involves maintaining the natural aggregate samples and the ones covered with bitumen in contact with a coloured solution, in certain standard conditions. The spectrophotometer is used to determine the different solution concentrations, in the two distinct situations. The adhesion is then calculated by dividing the obtained solution concentrations.

The affinity between aggregates and bitumen was tested using the rolling bottles method. Unlike the boiling water test, which is highly precise, the employed procedure is a simple, yet subjective method, used for routine laboratory tests. The aggregates coated in bitumen are spread on a metallic plate or a silicon-coated paper and kept...
overnight at normal temperature. Then, the sample is divided in three parts, which are transferred in rotating water-filled bottles. This is a direct method to assess moisture sensitivity by visually inspecting the coating-in-bitumen rate, by two independent operators. Other methods have also been used by researchers to evaluate the adhesion between aggregates and bitumen: peel tests [2][4][10], composite substrate peel test [9], energy dispersive X-ray spectrometry [4] or X-ray photoelectron spectroscopy [4].

In this study, SMA specimens were prepared in the laboratory and specific dynamic tests were performed on them in order to assess their physio-mechanical properties.

The void content of SMA was calculated using the true and apparent mixture densities. These parameters were determined on 60-mm-high cylindrical samples, with a diameter of 100 mm, prepared using a gyratory-shear molding press.

The stiffness modulus is one of the most representative characteristic of an asphalt mixture. It is determined through the indirect tensile test, on cylindrical samples 100 mm in diameter and 50 mm high, prepared using a gyratory-shear molding press. The specimens were submitted to deformation in the linear field, under repetitive loads, at a temperature of 20 °C. Effort amplitude and strains were measured, along with the shift between the two.

The SMA susceptibility to deformation (rutting resistance) was assessed through wheel tracking test (Figure 1 right) on laboratory-prepared rectangular asphalt mixture specimens (320x260x40 mm, Figure 1 left). Deformation speed and rut depth caused by the 10,000 passes of a loaded wheel, at 60 °C, were measured. This test simulates the effects of real road traffic on SMA.

The Schellenberg test was performed with the purpose of establishing the SMA susceptibility to segregation (i.e. the bituminous binder lost by the mixture stabilised with fibres at high temperatures).

RESULT AND DISCUSSION

The bitumen adhesion to aggregates tests results (Figure 2) revealed that using anti-stripping agents improves this characteristic. However, the adhesion increase percentage ranges from around 4 to 16 % (Table 2), depending on the aggregate source. The best results were obtained for the andesite aggregates from source no. 2. In this case, the
anti-stripping agent influence on bitumen adhesion is the lowest. Regarding bitumen types, using PMB led to appropriate adhesion values (at least 80 %) even when no anti-stripping agent was added. Diabase aggregates showed an opposite behaviour than the other ones, with improved adhesion for SRB.

Figure 2. Adhesion tests results

Table 2. Changes in bitumen adhesion brought by anti-stripping agent

| Aggregate source | Rock nature | Change in bitumen adhesion with vs. without additive [%] |
|------------------|-------------|---------------------------------------------------------|
|                  |             | SRB 50/70      | PMB 45/80-65                |
| Q₁               | andesite    | + 12.3         | + 15.9                      |
| Q₂               | andesite    | + 4.5          | + 3.9                       |
| Q₃               | dacite      | + 12.5         | + 11.5                      |
| Q₄               | diabase     | + 5.1          | + 6.2                       |

The aggregates extracted from the andesite source no. 2 provided the best affinity tests results (Figure 3). No difference between using SRB and PMB was detected in this case. There is no minimum quality requirement for this parameter. Using anti-stripping agents and/or PMB generally improves this characteristic. However, the affinity of SRB, with no added anti-stripping agent, to diabase aggregates is higher than in the case of PMB.

The wearing course durability is closely related to the SMA void content. As this parameter is reduced, the mixture becomes more impermeable. For upper technical class roads (i.e. motorways, 4+ lane roads), a maximum of 5 % void content is allowed in Romania. However, if the value is reduced too much, bleeding may occur.

In this study, using PMB reduced the void content, irrespective of the aggregates (Figure 4 left). All obtained results were below the maximum allowable limit of 5 %. Dacite provided the lowest void content of mixtures prepared with PMB, whereas andesite from source no. 2 led to increased (but still allowable) values.

Andesite aggregates from source no. 1 led to increased stiffness moduli compared to the other rocks (Figure 4 right). However, tests on SMA prepared with aggregates from the same type of rock, but exploited in source no. 2, resulted in 4 to 8 % lower values. This reduction roughly applies to dacite and diabase aggregates as well. Therefore, the aggregate influence on the results was rather similar, except for the andesite from source
no. 1 in North-Western Romania. Using PMB resulted in 2.4 to 6.8 % more flexible asphalt mixtures.

Regarding the SMA resistance to permanent deformation (Figure 5), the results were rather surprising. Although a low deformation speed was obtained for SMA prepared with dacite aggregates (source Q3), its rut depth was the highest of the analysed mixtures. A significant impact on deformation speed was observed for SMA prepared with andesite aggregates from source no. 1, by replacing SMB with PMB. As in the above-presented tests, this result does not apply for aggregates from source no. 2. Although andesite aggregates from source no. 1 led to increased SMA stiffness moduli, this did not reflect on deformation speed when using SRB. However, rut depths for SMAs prepared with andesite aggregates were the lowest, irrespective of the bitumen that was used. The increased mixture flexibility caused by PMB also implies deeper ruts when using dacite or diabase aggregates. Interestingly enough, the results for andesite aggregates did not exhibit this behaviour.

No significant differences were observed between SMAs prepared with different types of aggregates, regarding the mixtures susceptibility to segregation (Figure 6). In the case of diabase and andesite from source no. 1, a slight increase in drained material was observed for SMAs prepared with SRB, due to its decrease in viscosity at high temperatures. However, during the asphalt mixing procedure, using bitumen with low viscosity may improve aggregate coating and thus improve the mixture resistance to stripping [1].
CONCLUSIONS

This research has shown that the properties of an asphalt mixture, with particular focus on the aggregate-bitumen adhesion, are influenced by the natural aggregates, bitumen types and additives it contains.

Bitumen adhesion to andesite aggregates from source no. 2 was better compared to other aggregates. Using PMB and anti-stripping agent generally improved the aggregate-bitumen bond. An exception was noted in the case of diabase aggregates, for which SRB manifested improved adhesion. The specific reasons for this behaviour was not analysed in this study. The same conclusions apply to the affinity between aggregates and bitumen.

Aggregates in the asphalt mixture composition did not influence the void content reduction brought by replacing SRB with PMB. Dacite (source no. 3) provided the lowest void content (when using PMB), whereas andesite from source no. 2 led to slightly increased values. Dacite aggregates also led to low asphalt mixture permanent deformation speed, but, at the same time, to high rut depth. Deeper ruts were recorded for SMAs prepared with diabase aggregates as well. As Pop et al. [6] have shown, using PMB reduces the asphalt mixture stiffness. In this study, andesite aggregates from source no. 1 led to increased stiffness moduli compared to the other aggregates. This had an effect on rut depths, which were lowest for SMAs prepared with andesite...
aggregates, irrespective of the bitumen that was used. Regarding the mixtures susceptibility to segregation, no significant aggregate source influence was recorded.

Further research should consider testing asphalt mixtures prepared using hydrophobic aggregates, such as limestone or marble, as well as analysing the influence of multiple bitumen types and/or additives on asphalt and the interaction between its components.

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