ADHESION IMPROVEMENT BETWEEN BITUMEN AND MINERAL AGGREGATE: THEORETICAL PRINCIPLES AND THEIR IMPACT ON PAVEMENT STRUCTURE LIFETIME

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Abstract

Pavement structures often fail due to insufficient interphase interaction between bituminous binder and mineral aggregate. A thin bituminous film covering aggregate particles is often replaced by water, resulting in bitumen-water-aggregate system, which is firmly linked to adhesion failure between these materials. This paper describes how to avoid this phenomenon using nanomaterial based admixtures - adhesion promoters maximizing the formation of intermolecular interactions between bitumen and aggregate surfaces. Physicochemical and topological principles about interaction between the two materials were described. Regular paving grade bitumen 50/70 was modified by additives based on: (i) alkylsilanes; (ii) phosphorus compounds; and (iii) amines. Mineral aggregate grains (maximal size of 16 mm, pre-heated on 160±5 °C) were covered by the thin film of reference and such modified binders (170±5 °C). In this way prepared samples were exposed to water (60 °C) for 60 minutes. After samples removing from water, their digital images were obtained and then an area of aggregate grains covered by the bituminous thin film was assessed using: (i) bitumen adhesion test; following relevant technical standards; (ii) digital image analysis based on both gray level thresholding and entropy-based segmentation. It was shown that adhesion of the bitumen to aggregate, expressed by aggregate particle coating area, was increased by almost 18 %, if adhesion promoters were used.

Keywords: Asphalt mixture, adhesion promoters, interface interaction, coated area

1. INTRODUCTION

Asphalt mixtures, consisting of bituminous binder, mineral aggregate, and other admixtures, dominate during construction of road pavements worldwide. Their mechanical performance as well as pavement lifetime significantly depends on an interaction between individual phases, namely between bituminous binder and mineral aggregate. If adhesion between the bitumen film and aggregate fails, aggregate stripping usually occurs, resulting in subsequent damage of pavement structure and its integrity lost. Such estimated damages, known as potholes or just stripping, have a strong negative impact on asphalt pavements from the both the operational, traffic safety, and economical point of view [1].

Interphase interactions between bitumen, aggregate and other substances (e.g. dust, water) is complex process resulting in formation of adhesive bonds. Bitumen is usually considered to be a colloid system of intricate chemical composition and of complicated internal arrangement of molecules. Bituminous adhesive forces are promoted by polar components (such as polynuclear aromatics, phenolics, pyridinics, ketones, sulfides, etc.) lowering the interfacial energy between asphalt and aggregate [2]. Similarly, the temperature dependent viscosity reflects the capacity of bituminous material to be absorbed onto the aggregate surface. Furthermore, adhesive bonds are considerably influenced also by physico-chemical properties of the present mineral aggregates (surface morphology - roughness, porosity, surface area, dislocations, geometrical shape, etc. [3]) and the final adhesion is then the result of combination of the above mechanisms. It has to be stressed out that aggregates exhibit a strong affinity for water, in particular, the so-called acidic (hydrophilic) rocks with a high content of silicon-based substances (granite, quartz). Consequently, bitumen is frequently replaced by residual water, followed by formation of interfacial system bitumen-water-aggregate and this stripping effect may be firmly linked to adhesion failure between bituminous material and aggregate surface. This problem can be optimized by addition of adhesion promoters maximizing the formation of intermolecular interactions between bitumen and aggregate surface (apolar Lifshitz-van der Waals, resp. Lewis acid- based components) resulting in plausible values of the final surface free energy. Even a small amount of these promoters, usually between 0.01 and 0.5 wt.% to bitumen, causes significant enhancement in bituminous binder to aggregate adhesion [4,5].

In order to reveal interphase interaction between the two materials, several methods can be employed. Contact angle measurement of hot bitumen droplet applied on the planar surfaces of cut aggregate is able to provide an information about the interphase interaction [6,7]. However, this method cannot take into account a shape factor and the surface roughness of the aggregate. The same problem is typical also for peel-tests, when steel plates, bonded to aggregate by thin layer of the binder, are removed from planar aggregate surfaces [8]. Empirical methods based on visual observations of aggregate coated by the binder are frequently used throughout technical praxis; however, these cannot provide accurate and consistent results due to subjective assessment by evaluators [9,10]. Instead of evaluators, digital and computational technic can be employed to assess the rate of interaction between two materials of different state [11,12].

In this research study, we modified standard paving grade bitumen by several types of adhesion promoters. Such modified binders were used to cover mineral aggregate within test specimens which are defined by a selected aggregate fraction coated by hot bitumen. The area of coated aggregate by binder, pointing to adhesion between the two materials, was assessed using both conventional and primarily digital image analysis based methods.

2. ASPHALT MIXTURES

2.1. Bituminous binder

Paving grade bitumen 50/70 (EN 12591 and ČSN 657204) was used to cover aggregate grains. Reference mix marked as “MR” was produced without any additives. Compared to that, mixtures M1-M5 were made from the same type of bitumen, but these contained selected types of adhesion promoters.

2.2. Mineral aggregate

Crushed aggregate the Brant quarry, where granite porphyry is the key mineral, was chosen with respect to the availability of natural aggregate sources. Moreover, it was targeted to use a less suitable aggregate (hydrophilic) more susceptible to water damage, with poor adhesiveness to the binder and elevated potential of stripping failures shown in the asphalt mix composite. The criterion of less favorite adhesion was fully met by granite porphyry, or porphyry microgranite. A considerable proportion of the rock consists of quartz, potassium feldspar (orthoclase) and mica. The fraction was equal to 8-16 mm.

2.3. Adhesion promoters

To improve adhesion between the bituminous binder and aggregate, liquid-form chemical-based additives were used. In sum, five different products were selected: M1, alkylsilanes-based; M2, phosphorus-based; M3, based on chemical surfactants comprising amines; M4 and M5, based on unsaturated fatty acids combined with diethanolamine. Their amount differed from 0.2 to 0.4 wt. % to binder. Their specification and amount are summarized in **Table 1**.

Table 1 Summarization of asphalt mixture composition

Mixture	Type of mineral aggregate	Bituminous binder	Adhesion promoter	Adhesion promoter (wt. % to binder)
MR	Granite porphyry fraction 8/16 originated from the Brant quarry	50/70	-	0
M1			Additive based on alkylsilanes	0.2
M2			Additive based on phosphorus	0.3
M3			Additive based on chemical surfactants comprising amines	0.3
M4			Additive based on unsaturated fatty acids combined with diethanolamine	0.2
M5				0.4

3. DETERMINATION FOR ADHESION OF BITUMINOUS BINDER TO AGGREGATE

3.1. Conventional visual analysis

Following ČSN EN 73 6161 technical standard, test specimens in a defined glass bowl were prepared. These composed of several individual pieces of aggregate with fraction of 8/16 mm (in sum ca. 300 g, temperature 160±5°C) coated by the bituminous binder (12±0.3 g, temperature 170±5°C). Such prepared samples were stored for 24 hours, then submerged to water (60 °C) and stored additional 60±5 minutes. Then, they were subjected to conventional visual assessment according to cited standard. The conventional analysis is based on subjective assessment of two evaluators, who classify the rate of stripped areas of aggregate.

It is clear from the procedures mentioned above that the subjective assessment cannot exclude "human factor" and provide consistent results. In order to overcome this drawback, the test specimens placed in glass bowls were photographed using digital camera (Canon EOS 70D, focus length 50 mm, Av 8). Such obtained images were subjected to testing by other two assessment methods, as described below.

3.2. Gray level thresholding

The percentage of the aggregate surface coated by the bitumen was assessed using "in-house" developed Matlab software Binder. The main principle is based on identification of areas brighter than the dark bituminous binder. These areas are then considered to be parts of stripped aggregate particles. Such an approach introduced inaccuracies in the case of dark aggregates or light reflections on the bitumen surface. The reflected light causes local over-exposition within images, which the software evaluates as uncovered.

3.3. Entropy-based image segmentation

To overcome the issues connected with dark aggregate or local over-exposition, an alternative approach was addressed. Namely, we employed local entropy calculation in order to assess roughness of the texture. Such a procedure is computationally more demanding but still feasible and with much more accurate outcomes. The areas with high entropy are considered to belong to aggregates while low entropy areas represent the bituminous matrix.

4. RESULTS

Conventional visual analysis of asphalt mixture, executed following ČSN EN 73 6161 standard, revealed that the adhesion between aggregate and reference bitumen (mixture MR) reached only to "unsatisfactory" classification, i.e., the area of aggregate particles covered by bitumen was less than 80 %. According to gray-level and entropy-based analysis, the area was equal to 80.3 % (satisfactory) and 73.9 % (unsatisfactory), respectively. The results acquired using the entropy-based analysis were considered to be more accurate than those obtained from gray-level-based analysis due to problems with proper photographing caused by light reflection from the surface of shiny bituminous binder surfaces (this held true for all results comparison between these two methods).

Covered aggregate surface area of mixture M1 (containing adhesion promoters at the amount of 0.2 wt. %) reached on 91.9 % and 85.9 % (good / satisfactory), depending on the evaluating method (gray-level-based and entropy-based).

If whatever other adhesion promoters were used (M2-M5), the rate of aggregate covered area increased significantly up to ca. 95-99 % (good / excellent) depending on the evaluating method. The results are summarized in **Table 2** and graphically illustrated in **Figure 3.** These finding were comparable to research by Rosi et al. [3,6]. For illustration, the extreme cases (MR and M4) are shown in **Figure 1** and **Figure 2.**

Based on these results, it can be concluded that adhesion promoters significantly contribute to bituminous binder adhesion to mineral aggregate. Moreover, it was found that entropy-based assessment of interphase interaction between the two materials is a potential technique from the practical point of view, providing proper results and regarding shape factor of the aggregate.

Table 2 Assessment of adhesion of bituminous mixtures to mineral aggregate

Mixture	**Visual assessment**	**Gray-level-based assessment (% / classification)**	**Entropy-based assessment (% / classification)**
MR	unsatisfactory	80.3 / satisfactory	73.9 / unsatisfactory
M1	satisfactory	91.9 / good	85.9 / satisfactory
M2	excellent	98.8 / excellent	95.6 / good
M3	good	97.5 / excellent	93.7 / good
M4	good	97.8 / excellent	95.8 / good
M5	good	97.9 / excellent	94.6 / good

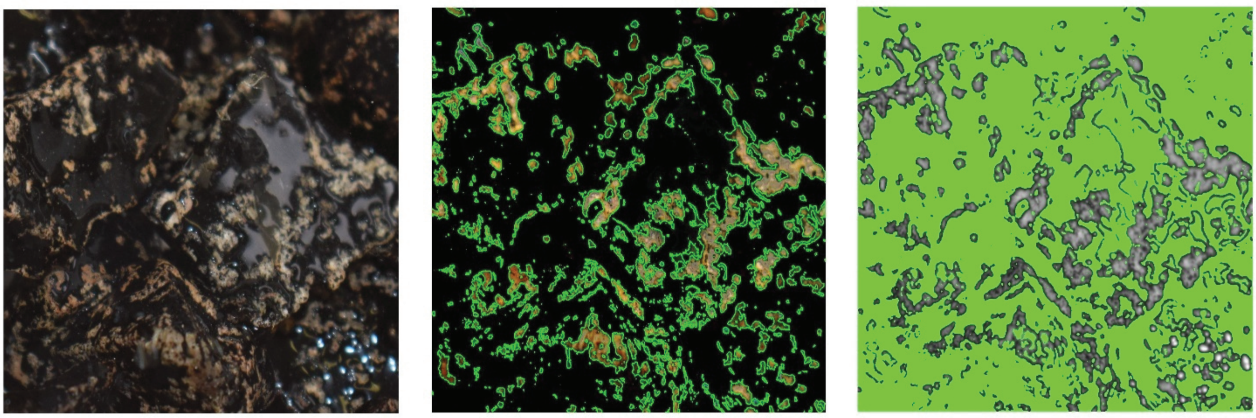

Figure 1 Images of poorly covered aggregate by binder (MR mixture), from left: original photo, gray level thresholding-, and entropy-based image segmentation

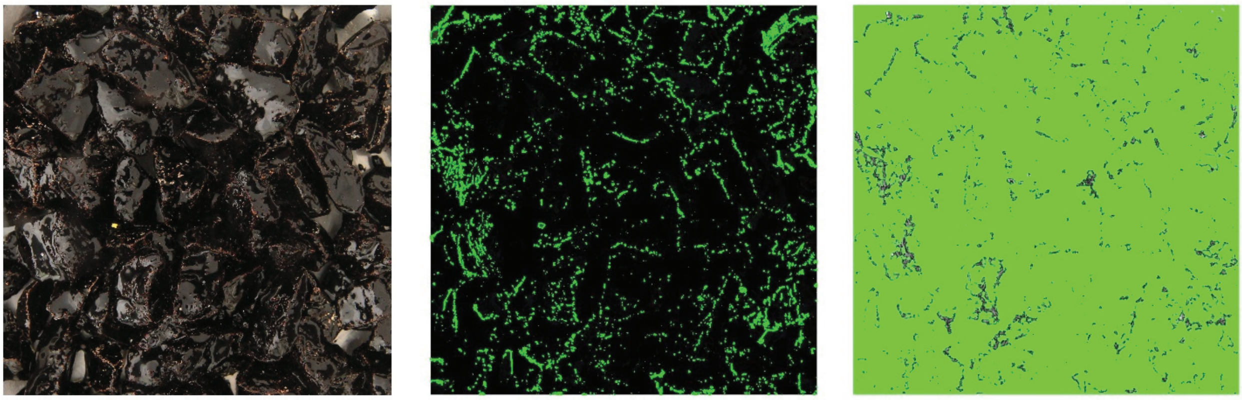

Figure 2 Images of good covered aggregate by binder (M4), from left: from left: original photo, gray level thresholding-, and entropy-based image segmentation

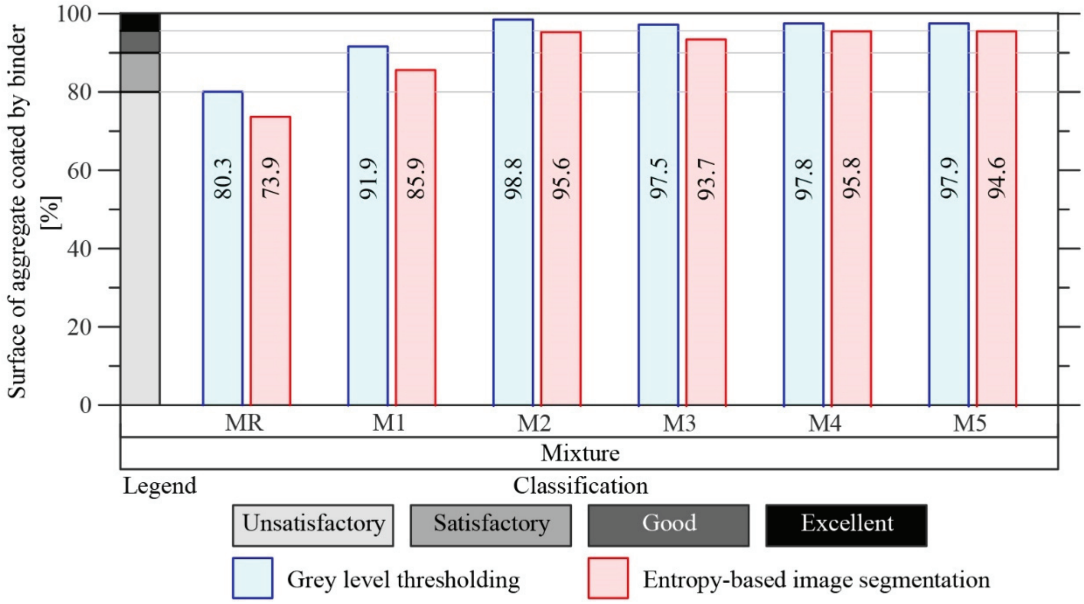

Figure 3 Assessment of adhesion of bituminous mixtures to mineral aggregate

5. CONCLUSION

Interphase interaction between straight-run distilled bituminous binder (50/70) thin film and mineral hydrophilic aggregate (8-16 mm, granite porphyry) from Brant quarry in Central Bohemian region was assessed using conventional and newly developed digital image-based methods. The binder was modified with nanomaterial chemical-based adhesion promoters in order to increase the adhesion between these two materials and thus to ensure creation of aggregate-bitumen interphase system only, with no other materials. For these purposes, chemical-based additives were used at the amount of 0.2-0.4 wt. % (to binder). Asphalt mixtures composed of the aggregate particles coated by the thin film of the bitumen were prepared, photographed and then subjected to the adhesion assessment. Focusing on the surface of these particles, an area coated by the binder was evaluated subjectively, following relevant technical standard and automatically, using grey level thresholding and entropy-based image segmentation analysis. It was shown that the coated area was approx. 74-80 % in the case of reference bitumen (without any nanomaterial based additives, depending on evaluating method. If adhesion promoters were used, the examined area was increased up to almost 100 %. Moreover, it was found that the evaluating method based on entropy-based image segmentation was considered to be more accurate, because it overcame problems relating with light reflection from shiny bituminous binder surfaces, resulting in local over-exposed areas within the photography.

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