THEORY AND MEASUREMENTS OF BITUMEN BINDERS
ADHESION TO AGGREGATE

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The pavement as unit is able to perform services, provided that we ensure good synergy of layers and especially good adhesion between used materials. Adhesion knowledge allows to design suitable technology by preventing defects and to ensure service ability and life of pavement.

In the paper there is a theoretical analysis of problem of bitumen binders adhesion to aggregate, basic methodologies to evaluate this property and specific results achieved at measuring by the method STN 65 7089 and EN 12272-3. At the same time the results are compared from the type of aggregate point of view as one of the most important factors that influenced adhesion.

1. Introduction

The wearing surface of the road is exposed to both vertical and tangential forces caused by the moving vehicle. The bitumen elements and aggregate stripping from the tyre are torn out by the suction effect of the wearing course, thus causing the continual destruction of the wearing course. The loss of bitumen cohesion and adhesion between the bitumen and aggregate is a major cause of defects in bitumen road surfacing. By the traffic effects, ravelling occurs. It is caused by material fatigue and stress pressure in tyre/surface contact area, weather effects, especially water which results in stripping the aggregate grains in the wearing surface (water is forced by the tyre engraving into the tyre/pavement contact area, and arises forces with similar effect as suction) and the ageing of the bitumen binder in the wearing surface which becomes harder and more fragile to the breaking.

The basis for the construction of the high quality wearing course with a long service life is the adhesion between the binder and the aggregate. It is important to achieve a strong bond among all materials during the service life at the varied climatic conditions. Adhesion knowledge allows to design a suitable technology, thus preventing defects and ensuring longer service life of the wearing course.

2 Methodological Approach

2.1 Adhesion

Generally, adhesion is defined as the affinity force between surface molecules of two materials that cause a close contact without chemical change. It depends on the interfacial surface tension and the binder wetting.

The ability of binder to cover aggregate (wetting ability) is called active adhesion. Passive adhesion expresses the binder coat resistance to the stripping from aggregate surface by the water effect. If the conditions of coating are properly designed and performed, there is no active adhesion [1].

Besides the mechanical interaction at the contact surface (liquid bitumen penetration into the pores of solid material), which depends on the physical properties of aggregate and bitumen – also mechanical adhesion exists. It is also necessary to ensure sufficient bond strength of physical and chemical bonds between contacted binder and aggregate – specific adhesion. Incipient chemical cohesion caused by the effect of chemical (interatomic) bonds is more energetically stable than physical cohesion initiated by a physical (intermolecular, van der Waals) bond effect [2].

2.2 Theories of adhesion mechanism

From the general point of view, adhesion is the force that bonds the binder to solid surfaces and prevents its tearing away. Several mechanisms have been used to explain adhesion between

| Theories of adhesion mechanism | Tab. 1 |
|-------------------------------|-------|
| Theory                        | Mechanism                          | Strength of interaction |
| adsorption (Bruyne)           | van der Waals, H-bonding            | moderate to strong      |
| diffusion (Voyutskii)         | inter-diffusion binder molecules to solid material | variable               |
| bonding (Bickerman, mechanical interlock) | mechanical diffusion of binder to aggregate pores, close contact wetting | variable               |
| surface energy theory         | binder and aggregate surface with more critical surface tension, wetting | variable               |
| reactive theory, chemical bonding | covalent bond                        | very strong             |

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If the Dupré equation:

\[ W = \sigma_{SV} + \sigma_{LV} - \sigma_{SL} \]  

(2)

where: \( \sigma_{SV} \) is surface energy of solid phase, 
\( \sigma_{LV} \) is surface energy of liquid phase, 
\( \sigma_{SL} \) is energy of solid – liquid phase interface.

Measurement of surface energies, interface energies and contact angles are difficult under the conditions of conventional laboratories. Thus, the comparison of properties of different surfaces with the binder is difficult because the measurement of the contact angle requires even surface of solid material and then the surface must be modified [5].

Specific value of the surface tension of the binder and aggregate contact above which spontaneous coating does not occur is the critical surface tension. Only binders with surface tension less than critical value will cover solid surfaces spontaneously.

It is necessary that bitumen have good wetting ability to create the stable coat on the aggregate. Wetting and adhesion perform on the phases interface and relate to the intermolecular forces. The bitumen covers aggregate surface well if its surface tension is low. Surface tension of bitumen decreases when temperature increases. The viscosity of bitumen decreases at higher temperature; bitumen covers aggregate well and adheres to the aggregate surface.

3 Experimental measuring

As mentioned above, it is difficult to measure adhesion as the force that bonds bitumen to aggregate therefore methodological techniques have been developed for evaluating adhesion by measuring the properties of the bitumen mixtures. Methods used to adhesion measurement are based on one of following principles:

- visual evaluation by estimation of uncovered surface of the aggregate by the water effect,
- adhesion measurement by the mechanical test (adhesion evaluating as the resistance to mechanical stresses),
- adhesion evaluation as the resistance to water by measuring mechanical property of mixture before and after tempering in water,
- evaluating the chemical adhesion.

The basic test method of adhesion evaluating in the Slovak Republic is the procedure which follows the technical standard STN 65 7089 – Determination of Adhesion of Asphalt Products to Aggregates from year 1982 [6]. According to this standard the adhesion of asphalt products to aggregates is the resistance of the asphalt film on the aggregate surface to the water displacement (passive adhesion). The adhesion influences asphalt mixtures quality and service life. This method also indicates the efficiency of adhesion additives.

The methods able to evaluate adhesion of the binders penetration 35 - 210, cut-back bitumen with the flow time C/5/60 to 170 s and C/5/25 to 70 s and asphalt anionactive emulsions to the aggregates. Dry aggregates and wet aggregates are covered by the...
tested bitumen. After 24 hours of tempering it is exposed to the effects of tempered water (at the temperature of +60 °C res. +40 °C) for one hour. Adhesion is expressed as a mean value of stripping of the asphalt film on the grains of the aggregate surface. Thus, adhesion is accepted if some uncovered points appear on the surfaces of covered aggregate grains or stripped borders and corners on more than two grains of the aggregate. If there are larger stripped areas on the grains of aggregate surface (two and more aggregate chippings), adhesion is not acceptable.

The degree of stripped asphalt on aggregate surface is evaluated visually. In this way, the evaluation becomes subjective depending on the technician performing the test. Methodology, test specimen, and test procedure predetermine this test to the asphalt evaluation and aggregate adhesion used in asphalt mixtures.

The European standard prEN 12697-11 [7] belongs to this test group. Standard measures evaluate the compatibility between the aggregate and bitumen, expressed by visual observation of the loss of adhesion in uncompacted bitumen-coated aggregate mixtures in the presence of the water. Test specimen is also affected by mechanical load with the presence of water.

From the second group of tests that evaluates adhesion by the mechanical test is Vialit test. The test methodology is the base for European standard EN 12272-3 Determination of Binder Aggregate Adhesivity by the Vialit Plate Shock Test Method [8]. This standard specifies test methods for determining the binder – aggregates adhesion as two main components of surfacing.

The standard applies to the measurement of binder – aggregate adhesion and the influence of adhesion agents on adhesion characteristics as the aid to design binder – aggregate systems for surface dressing. This methods allows to evaluate adhesion of:
- hydrocarbon binders used for surface dressings,
- paving grade and modified bitumens,
- cut-back bitumens,
- asphalt emulsions,
- to all aggregate types (with size 6–8, 8–11, 11–16 and 4–6, 6–10, 10–14).

The principle is the measurement of the binder – aggregate adhesion after mechanical exposure. European standard specifies the test method for determining:
- mechanical adhesion of the binder to the aggregate – Vm (property to bond dry aggregate chippings with their natural dust and fine particles)
- active adhesion – Va (the property to bond dump aggregate chippings in their natural state in which they occur in the dumping sites)
- improvement of mechanical and active adhesion using some adhesive agents to the binder or to binder – aggregate interface.

In terms of the standard European requirements prEN 12271-5 [9], the value of mechanical adhesion and active adhesion has to be minimum 95 % res. 90 % to reach value 2 res. 1 (for value 0 there is no adhesion requirement). The standard has three basic divisions:
- active adhesion and mechanical adhesion,
- wetting temperature,
- fragility temperature.

In the first part, the test procedure is defined according to the known test method Vialit and determining the active adhesion. In the second part, the treatment for the determining wetting temperature at the lowest binder temperature before the spreading aggregate is described when the grinding of the aggregate is possible with minimum 90 % adhesion (90 of 100 grains of aggregate remain adherent to the experimental plate). And the other treatment to determine fragility temperature at the temperature at which minimum 90 % aggregate chippings remain bonded to the plate is carried out at Vialit premises.

The binder – aggregate adhesion is expressed by the total number of aggregate chippings bonded to the plate and the aggregate with the binder fallen off after test. The advantage of this test method is that it allows measuring and evaluating the adhesion with different types of binders and aggregates. At the same time, it is possible to verify adhesion agents efficiency. Test results are influenced by binder – aggregate adhesion and also by binder cohesion, especially at low temperatures.

3.1 Experimental measuring - materials

For experimental measurements of bitumen and aggregate adhesion by the above presented test procedures, we used aggregates from different local sources, especially from the northern

| Aggregate content [%] | Varín | Suja | Biely Potok | Dubna Skala | Malužiná | Hanišberg | Kameneck p. Vtáčnikom |
|------------------------|-------|------|-------------|-------------|----------|-----------|---------------------|
| SiO2                   | 1.04  | 0.30 | 0.30        | 70.74       | 56.77    | 56.5      | 59.27               |
| Al2O3                  | 0.21  | 0.20 | 0.07        | 17.31       | 12.96    | 19.5      | 19.72               |
| Fe2O3                  | 0.20  | 0.10 | 0.25        | 5.65        | 4.77     | 6.50      | 5.82                |
| CaO                    | 47.02 | 30.90| 31.60       | 3.49        | 9.06     | 7.50      | 5.81                |
| MgO                    | 6.86  | 21.40| 20.70       | 0.63        | 2.68     | 5.00      | 2.17                |

The aggregate composition Tab. 2
Slovakia, representing these rocks: limestone from the locality Varin, dolomite from two localities (Suja, Biely Potok), granodiorite from the locality Dubná Skala, melaphyre from the locality Malužina and andesite from two the localities (Haníšberg, Kameneck pod Vtáčnikom), and bitumen binders from by-products in Slovnaft oil refinery, both paving grade and modified (with the SBS rubber) [10].

Measured bitumen properties Tab. 3

| Binder          | Penetration at 25 °C in 0.1 mm | Softening point R&B v °C, minimum | Ductility at 25 °C in cm, minimum |
|-----------------|-------------------------------|-----------------------------------|----------------------------------|
| bitumen 70/100  | 83.7                          | 53                                | 114.9                            |
| modified        |                               |                                   |                                  |
| Apollobit MCA-S | 93.9                          | 75                                | 62.2                             |

3.2 Comparison of laboratory results

Experimental measurements of adhesion was performed by two test methods, according to the standard STN 65 7089 and Vialit test [11].

After bitumen and aggregate having been mixed, the process of interaction between aggregate and bitumen begins. These processes are determined by chemical and physical-technical properties of aggregate. The aggregate composition is evaluated from both chemical or mineralogical point of view. The chemical structure has only indirect effect on the aggregate used. From mineral point of view, the aggregate contains mostly one dominant component and some minor mineral components. For example, the limestone rock consists of dominant component limestone, and silica, clay, micaous minerals as minor components. In term of adhesion, the content of SiO2 in aggregate is the most important. The bitumen–aggregate adhesion results by method of STN 65 7089 (tab. 4) and Vialit test (fig. 2) show that the content of SiO2 in aggregate has negative influence on the adhesion of bitumens. The adhesion of acid aggregate and bitumens mostly poor adhesion level.

Regression analysis of relation to content of SiO2 in aggregate shows that there is a linear dependence between adhesion results and aggregate content of SiO2. This dependence obtained from the measurements with all used bitumens and aggregates and also with measuring dry aggregate and aggregate with their natural dust and fines, and also adhesion measuring of specimens aggregate with their natural dust and fines in water. Neutral aggregate (andesites and melaphyre) had average adhesion values 96 to 100 %.

From other mineral ingredients which can be found in the aggregate, some minor minerals that show an increased content of Al2O3 are important. These minerals with SiO2 have an apparent hydrophilic character in contrast with hydrophobic character of limestone. These minerals show very different reactions toward the bitumen binder and water.

The worst adhesion results obtained by the Vialit method show the aggregate with high content of SiO2 70.74 % granodiorite form Dubná Skala locality. On the contrary, the adhesion results with limestones and dolomites with low content of SiO2 showed the best results of adhesion with all used bitumens by Vialit test 98.7 to 100 % adhesion. These results were confirmed by both test methods, adhesion measuring of specimens aggregate with their natural dust and fines and specimens tempering in water. Neutral aggregate (andesites and melaphyre) had average adhesion values 96 to 100 %.

Regression analysis of relation to content of SiO2 in aggregate shows that there is a linear dependence between adhesion results and aggregate content of SiO2. This dependence obtained from the measurements with all used bitumens and aggregates and also with measuring dry aggregate and aggregate with their natural dust and fines, and also adhesion measuring of specimens tempered in water. From the analysis of these dependencies, we can conclude that the adhesion decreases in dependence on the content of SiO2.

From other mineral ingredients which can be found in the aggregate, some minor minerals that show an increased content of Al2O3 are important. These minerals with SiO2 have an apparent hydrophilic character in contrast with hydrophobic character of limestone. These minerals show very different reactions toward the bitumen binder and the water.

From these data it is evident that thickness of bitumen coat around the limestone chipping is essentially higher than around silica (quartz) chipping. On the surface of limestone chipping contents adsorption we can find centers in the form of limestone cations (CaO), magnesia (MgO) and ferrate (Fe2O3), with the intense positive potential. The anion exchange of bitumen binder conducts to the strong binder film bonds.

The adhesion results of bitumens and aggregates by STN 65 7089 Tab. 4

| Binder          | Adhesion |
|-----------------|----------|
| bitumen 70/100  | good - suitable | suitable - poor | poor |
| modified        | good     | good - suitable | poor |
| Aggregate       | dolomite 2 | dolomite 1 | limestone |
|                 | andesite 1 | melaphyre | andesite 2 | granodiorite |
|                 | basic | neutral | acid |
Adhesion results comparison in dependence on the content of limestone (CaO) and magnesia (MgO) show that rocks with high content of these minerals have good adhesion (fig. 3 and fig. 4). The dependence is expressed in regression function. Parameters A and k depend on the type of binder and also the system of measuring (measurement with washed and dry aggregate, aggregate with their natural dust and fines, measurement of experimental samples tempered in water).

Positive effect of CaO and MgO content in the aggregate on the bitumen binder adhesion was confirmed by the adhesion results tested by standard STN 65 7089.

From the measurement results based on the comparison of the observed specimens we can conclude that the best level of adhesion was exhibited with basic aggregates with the high of CaO content and MgO, then with neutral aggregates, and from the adhesion point of view the acid aggregate with the high content of SiO₂ and Al₂O₃ had the worst results with all used binders. The modified bitumen adhesion to aggregate had better results than paving grade bitumen adhesion. Regarding to a limited space of the paper, detailed adhesion results of binders to aggregates at different conditions (wet aggregate, aggregate with dust on the surface, different temperatures, specimens tempering in water, etc.) are not presented in paper.

4. Conclusions

Most workplaces in other countries have developed their own laboratory test methods measuring the adhesion, and their own specific criteria of evaluation of bitumen adhesion to aggregate. At our workplace we evaluated the possibility of using the new adhesion test and evaluation method Vialit. Test results and their comparison with the results obtained from measurements by STN 65 7089 show the suitability of this method to determine adhesion of different binders and aggregates. The test method enables to determine the adhesion and at the same time to study different factors effecting the adhesion: the type of aggregate and binder, surface characteristics of aggregate, temperature sensitivity of binder-aggregate bond, resistance to water, etc. Considering the simplicity of the test equipment servicing, this test method can be included among the general tests of road building materials.

| Binder | Adhesion | bitumen 70/100 | modified bitumen |
|--------|----------|----------------|------------------|
| content | granodiorite | poor | suitable |
| CaO | 3.49 % | 5.81 % | 7.50 % | 9.06 % | 30.90 % | 47.02 % |

| Binder | Adhesion | bitumen 70/100 | modified bitumen |
|--------|----------|----------------|------------------|
| content | granodiorite | poor | suitable |
| MgO | 0.63 % | 2.17 % | 2.68 % | 5.00 % | 6.86 % | 21.40 % |

Fig. 3 The adhesion results of bitumen binder to aggregate in dependence on content CaO in aggregate

Fig. 4 The adhesion results of bitumen binder to aggregate in dependence of content MgO in aggregate

Tab. 5 The adhesion results of bitumens and aggregates by STN 65 7089

Tab. 6 The adhesion results of bitumens and aggregates by STN 65 7089
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