Testing of bitumen-aggregate affinity by various methods

Krzysztof Błażejowski[1, a], Jacek Olszacki[2, b], Hubert Peciakowski[2, c], Marta Wójcik-Wiśniewska[1, d]

[1] *R&D dept, ORLEN Asfalt, Plock, Poland*
[2] *PKN ORLEN, Plock, Poland*

[a] krzysztof.blazejowski@orlen.pl
[b] jacek.olszacki@orlen.pl
[c] hubert.peciakowski@orlen.pl
[d] marta.wojcik-wisniewska@orlen.pl

Digital Object Identifier (DOI): dx.doi.org/10.14311/EE.2016.397

ABSTRACT

The affinity of bitumen and aggregates is one of the most important factors influencing durability of the asphalt mixture and then the asphalt pavement, especially when the freeze-thaw period occurs. There are many methods of testing this property. Presently, the method according to EN 12697-11 (A: rolling bottles) is one of the most often used. From the other side, the ITSR method)EN 12697-12) with one freezing cycle for asphalt mixtures testing is used. Because there were a lot of problems with fulfilling the ITSR requirements, the applicability of rolling bottle test was questionned. The scope of the research was to recognize an influence of different rolling bottle test variants on the final results (durable bonding between binder and aggregate). Generally, the mode of aggregate-binder samples' preparation for rolling bottle test was similar to behavior of samples in procedure with ITSR. Following parameters were analyzed: preparation mode of bitumen (aged or not), aggregates' soaking with water, freezing of coating aggregates, NaCl solution presence etc. Finally, there were found some conclusions about factors influence to results of rolling bottle test.

Keywords: Adhesion, Aggregate, Antistripping agents, Bonding, Durability

Testing of bitumen-aggregate affinity by various methods

Krzysztof Błażejowski[1], Jacek Olszacki[2], Hubert Peciakowski[2], Marta Wójcik-Wiśniewska[1]
[1]ORLEN Asfalt sp. z o.o., Płock, Łukasiewicza 39, Poland
[2]PKN ORLEN S.A., Płock, Chemików 7, Poland
E-mail: krzysztof.blazejowski@orlen.pl

ABSTRACT

The affinity between aggregate and bitumen is one of the most important factors affecting the durability of asphalt and subsequently the durability and tenacity of the road pavement. The adhesion of bitumen to aggregate also greatly affects the water-tightness and resistance to hardening processes that occur during use of the road. Throughout the world there are many methods of testing the affinity between the grains of aggregate and bitumen. In Poland, the most popular method is the "rolling bottle test" according to EN 12697-11. In 2010, in addition, a new procedure was introduced in Poland for testing asphalt samples according to EN 12697-12 (the ITSR method) which involves the freezing of water-saturated cylindrical samples at -18°C and then thawing them in a water bath at 60°C. A number of research studies carried out in Poland, however, have not shown any link between the results of the rolling bottle method (EN 12697-11) and the results obtained by the ITSR method (EN 12697-12). The aim of the studies presented in this article was to test the hypothesis that the method in which the sample of aggregate coated with bitumen is prepared for the rolling bottle test according to EN 12691-11 makes the adhesion test results different, i.e. whether it affects the durability of the bitumen-aggregate bonding. The authors developed an original method called "BOP", which introduces significant changes into the method of sample preparation with respect to the standard test procedures for rolling in water. In the studies, different options for the preparation of samples of aggregates and bitumen were used, which were to correspond with the procedure of dealing with cylindrical samples in the ITSR method. The studies showed that the proposed methods of sample preparation for the rolling bottle test did not diversify the affinity test results, in addition, a very significant influence was observed of the period of heating the samples on the durability of the adhesion of the bitumen to the aggregate.

---

1. INTRODUCTION

Many factors influence the durability of asphalt used in pavement construction. One of them is the adhesion of bitumen to the mineral aggregate. In asphalt, bitumen acts as a binder bonding the grains of aggregate, which bonding affects, among others, the mechanical strength of the asphalt course, including its resistance to cracking. It can be said that the adhesion of bitumen to the aggregate to a large extent determines the durability of the road pavement, additionally influencing its water-tightness and resistance to the hardening processes that occur during use of the road.

The affinity of bitumen with aggregate depends on the properties of the aggregate such as moisture, dust (cleanness), porosity, ability to absorb bitumen, the micro-texture of the granules, the chemical type (conventionally: acidic, alkaline and in-between), as well as the properties of the bitumen such as viscosity, adhesion, composition group and acid number. Apart from the influence of the particular materials, a large influence on the durability of the bitumen and aggregate binding is exerted by properties of the asphalt, such as bitumen content, the amount of voids, permeability, type of filler, granulation etc. [Read et al. 2003; Abu-Qudais 2007]. A critical analysis of the theories describing the mechanism of loss of affinity between bitumen and aggregate were carried out in a study [Hefer et al. 2005].

In order to improve the adhesion of bitumen to aggregates, adhesives agents are often used, which alter the physicochemical nature of the contact between aggregate and bitumen [Cui et al. 2014]. Adhesive agents reduce the surface tension of the bitumen and reduce the contact angle which forms at the junction of three phases: binder, aggregate and water.

For many years a range of methods has been employed to check the resistance of asphalt to water and frost. Over the years, the most popular method was AASHTO T 283 and then the similar EN 12697-12. Currently, there are also other, new test methods such as SATS (Saturation Ageing Tensile Stiffness) [Airey et al. 2008] are used.

In Poland, as in many European countries belonging to CEN, the adhesion of bitumen to aggregates is tested according to EN 12691-11 (Method A: rolling bottle test). In 2010, a new procedure was introduced in Poland for the testing of

asphalt according to EN 12697-12 (the ITSR method) involving the freezing of water-saturated cylindrical samples at temperatures down to -18°C and then heating them in a water bath at 60° C. Despite the similarities to the US procedures according to AASHTO T 283, the results obtained in the laboratory were significantly lower than those required by national law. A number of research studies carried out in Poland in the years 2011-2014 showed no direct link between the results of studies using the rolling method (EN 12697-11) and the results obtained by ITSR with the procedure of freezing and heating (EN 12697-12) [Krajewski et al. 2012, 2013; Błażejowski et al., 2013; Witczak 2014]. Simultaneously, research studies were in progress to adjust the national ITSR procedure, which ended in 2013 [Sybilski et al, 2013].

2. THE RESEARCH PROGRAMME

The aim of the research programme was to test the hypothesis that the method of preparing the sample of aggregate coated with bitumen for the rolling bottle test according to EN 12697-11 differentiates the test results of adhesion, i.e. affects the durability of the film of bitumen on the aggregate. The authors developed an original method with a working title "BOP", which introduced significant changes in the methods of sample preparation with regard to the standard procedure for the rolling in water test. The different options of sample preparation of samples of aggregate with bitumen were to correspond to the procedure for dealing with asphalt cylindrical samples in the ITSR test.
The research programme involved checking the degree of adhesion of paving grade bitumen 35/50 to acidic aggregate (granite) and alkaline aggregate (limestone). The samples of paving bitumen were subjected to hardening by the Rotating Flask Test (RFT according to EN 12607-3) in two forms: single and triple hardening. Then the aggregate was coated with bitumen, after which there were four options for dealing with the samples thus obtained. Each of the options of the prepared samples was studied using the rolling bottle method according to EN 12697-11, into which additional operations were introduced such as: freezing the bitumen-coated aggregate at -18°C, conditioning the aggregate in water before and after coating in bitumen, rolling in an aqueous solution of NaCl.
The BOP method was particularly different to the standard method in that the bitumen-coated aggregate was heated to 160°C. In the standard rolling test, samples of aggregate are not subjected to heating after coating in bitumen.
The study also used a number of adhesives to improve the adhesion of bitumen to the aggregate.
The authors of the method assumed that, after the introduction of significant changes to the method of sample preparation, they would achieve different adhesion results according to EN 12697-11 (A) which would at a later stage be able to be correlated with the ITSR test results (the planned second stage of the study).

3. THE MATERIALS USED FOR THE STUDY

3.1. Samples of bitumen 35/50

The study used samples of paving grade bitumen 35/50 according to EN 12591 with the parameters listed in Table 1.
Studies of adhesion to aggregate were carried out using non-hardened bitumen, and bitumen following single and triple hardening. The bitumen was subjected to hardening by the RFT method in accordance with the EN 12607-3:2010 standard.

Table 1. Properties of 35/50 bitumen used for the studies

Property		Paving bitumen 35/50 acc. to EN 12591		
		Before hardening	After I cycle of hardening by RFT method	After III cycles of hardening by RFT method
Penetration at25°C, 0.1mm		43	32	27
Change in penetration, 0.1mm		-	-10	-16
Retained penetration after hardening Pen_{25}, %		-	75	63
Softening point, °C		54.2	61.2	65.4
Increase in T_{PiK} after hardening, °C		-	7	11
Dynamic viscosity, Pa·s	60°C	815	1915	6851
	$\Delta\eta_{60}$	-	1100	6036
	90°C	23.98	35.61	83.65
	$\Delta\eta_{90}$	-	11.6	59.7

	135°C	0.91	1.35	2.65
	$\Delta\eta_{135}$	-	0.44	1.74
	150°C	0.41	0.59	1.08
	$\Delta\eta_{150}$	-	0.18	0.67
Hardening index (VAI)		-	2.34	8.40

3.2. Aggregates

Two types of aggregate were used for the adhesion test: crushed limestone, which was alkaline, and crushed granite, which was acidic. In both cases the 8/11 mm. fraction was used.

3.3. Adhesive agents

Four different agents were used in the study to improve the adhesion of bitumen to the aggregate surface. Some basic information about the properties of the adhesives is shown in Table 2. The share of each agent in the bitumen amounted to 0.5% m/m.

Table 2. Characteristics of adhesive agents

Property	A1	A2	A3	A4
Melting temperature, °C	ca. 3	ca. -7	85.4 – 93.3	<-30
Density, 20°C, g/cm^3	ca. 0.96	0.99-1.01	1.10	-
Flash point, °C	>150	174	-	-
Appearance	Light brown oily liquid	Dark yellow oily liquid	Light cream solid wax	Dark brown sticky wax
Composition	Product based on derivatives of fatty amines	Surfactant mixture	Sulphonic acid	Mixture of amidoamines, ethylenediamines and glycerine

4. METHOD OF SAMPLE PREPARATION

0.5% m/m of selected adhesive agent was introduced into the bitumen at a temperature of 150°C.The aggregate was coated with bitumen in accordance with p. 3.1. according to the procedure set out in EN 12697-11.
The further procedure was determined by one of the options of the BOP method.

4.1. BOP IB option

The sample of aggregate coated in bitumen was heated at 160°C for 4 hours, after which it was placed on silicone paper and left at ambient temperature for 24 hours. Next adhesion was determined according to EN-12697 (“rolling bottle test”) in water.

4.2. BOP IIA option

The sample of aggregate coated in bitumen was heated at 160°C for 2 hours, after which it was placed on silicone paper and left at ambient temperature for 24 hours. The sample was then placed in a sealed plastic bag containing 10 ml of distilled water and frozen at a temperature of -18°C for 4 hours, after which the sample was thawed in a water bath at 60°C for 2 hours. Then adhesion was determined according to EN-12697 (“rolling bottle test”) in water.

4.3. BOP IIB option

The sample of aggregate coated in bitumen was heated at 160°C for 2 hours, after which it was placed on silicone paper and left at ambient temperature for 24 hours. The sample was then placed in a sealed plastic bag containing 10 ml of distilled water and frozen at a temperature of -18°C for 4 hours, after which the sample was thawed in a water bath at 60°C for 2 hours. Then adhesion was determined according to EN-12697 (“rolling bottle test”) in an aqueous solution of 7% NaCl.

4.4. BOP IIC option

The sample of aggregate coated in bitumen was heated at 160°C for 2 hours, after which it was placed on silicone paper and left at ambient temperature for 24 hours. The sample was then placed in a sealed plastic bag containing 10 ml of distilled water and frozen at a temperature of -18°C for 4 hours, after which the sample was thawed in a water bath at 60°C for 2 hours. After thawing the sample was again placed in a plastic bag and the cycle of freezing and thawing was repeated once. Then adhesion was determined according to EN-12697 ("rolling bottle test") in an aqueous solution of 7% NaCl. This option was added to check the effect of more than one cycle of freezing and thawing and complete rolling bottle tests were not performed with all the types of adhesive agents.

A summary of all the stages of the treatment of the samples is presented in Table 3.

Table 3. Treatment of samples

Stages of test procedure	BOP-IB	BOP-IIA	BOP-IIB	BOP-IIC
Coating of aggregate with bitumen according to EN 12697-11	+	+	+	+
Heating coated aggregate at 160°C for 4 hours and thorough re-mixing	+	-	-	-
Heating coated aggregate at 160°C for 24 hours and thorough re-mixing	-	+	+	+
Conditioning coated aggregate in air for 24 hours	+	+	+	+
Freezing coated aggregate sample at -18°C for 4 hours, after prior sealing in plastic bag containing 10 ml distilled water	-	+	+	+
Thawing sample in a water bath at 60°C for 2 hours	-	+	+	+
Freezing coated aggregate sample at -18°C for 4 hours, after prior sealing in plastic bag containing 10 ml distilled water (second freezing cycle)	-	-	-	+
Thawing sample in a water bath at 60°C for 2 hours (second thawing cycle)	-	-	-	+
Determining adhesion according to EN-12697-11, rolling time 6 hours ("rolling bottle test")	+	+	+	+
Determining adhesion according to EN-12697-11, rolling time 6 hours in aqueous solution of 7 % NaCl ("rolling bottle test")	-	-	+	+
Photographing aggregate coated with bitumen after testing adhesion	+	+	+	+

5. RESULTS

The adhesion of bitumen to aggregate, determined by the methods described in point. 4.1-4.4, was evaluated visually by specifying the percentage of unwashed-out visible surface of the aggregate sample. The test results were analysed for an assessment of the effectiveness of the "BOP" methods employed.
Figure 1 shows the results of test of the adhesion of bitumen 35/50 to granite and limestone, without the addition of adhesive agents, labelled "standard rolling bottle test" and the BOP IB - BOP IIC methods.

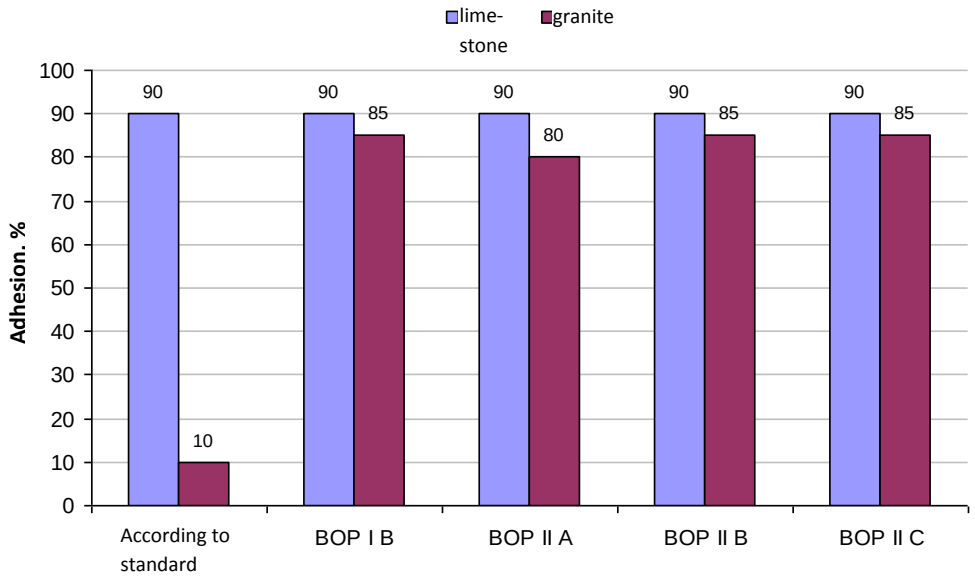

Fig. 1. Adhesion of bitumen 35/50 to granite and limestone labelled "standard rolling bottle test" and "BOP" methods

The adhesion of bitumen to limestone (basic aggregate) for all the samples, determined by both the standard and BOP methods was the same and amounted to 90%. However, the adhesion of bitumen to acidic aggregate - granite, labelled standard "rolling bottle test", was 10% and was significantly lower than the adhesion determined by the BOP methods (80-85%). It should be noted that the samples determined in accordance with EN-12697-11 were not treated by prior heating at 160°C, as was the case in all the samples assayed by the BOP method options.

The optimum heating time for the sample can be taken as 2 hours (methods: BOP IIA, BOP IIB, BOP IIC) as any further extension of the heating time did not significantly affect the degree of adhesion (BOP method IB).

The following graphs show the results of the degree of coverage of the aggregate with bitumen before and after hardening by the RFT method, determined by the BOP IB, BOP IIA and BOP IIB methods using adhesive agents:

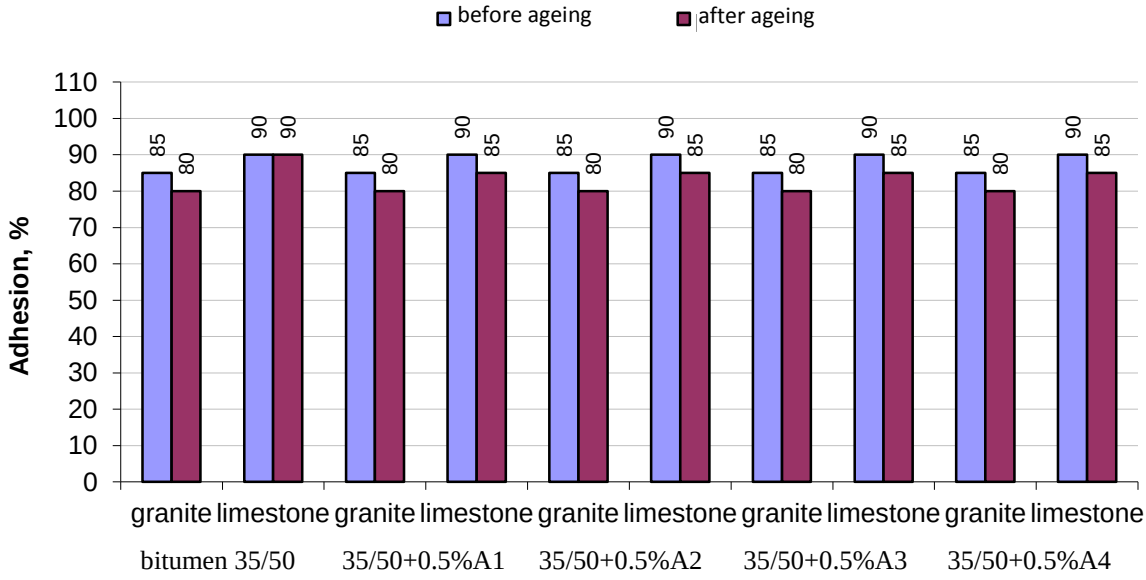

Fig.2. Adhesion of bitumen 30/50 to granite and limestone with the addition of adhesive agents determined by BOP IB method

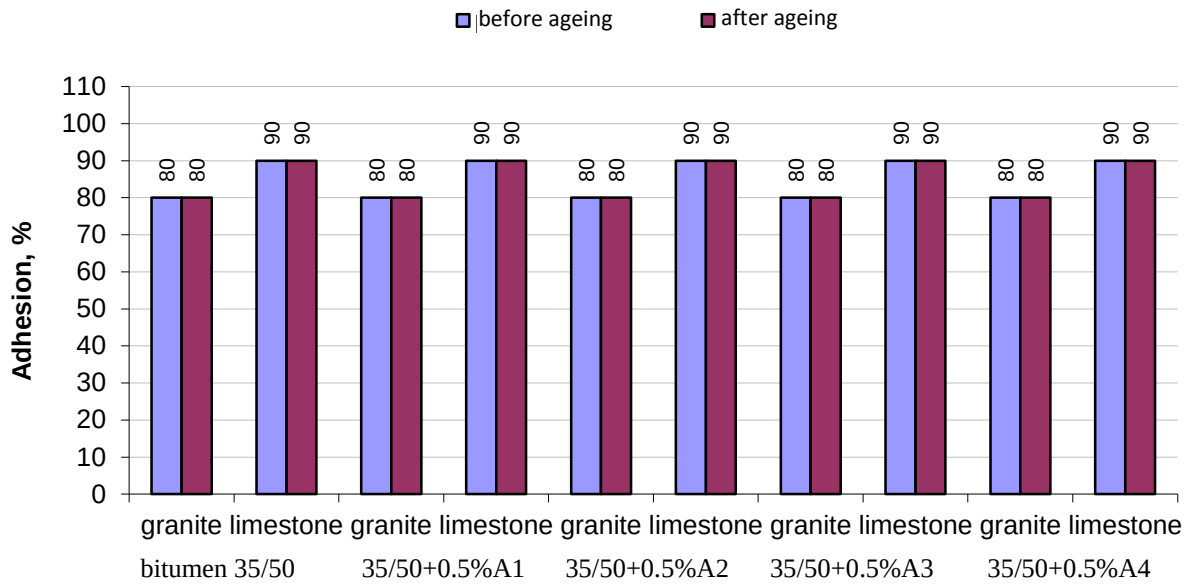

Fig.3. Adhesion of bitumen 30/50 to granite and limestone with the addition of adhesive agents determined by BOP IIA method

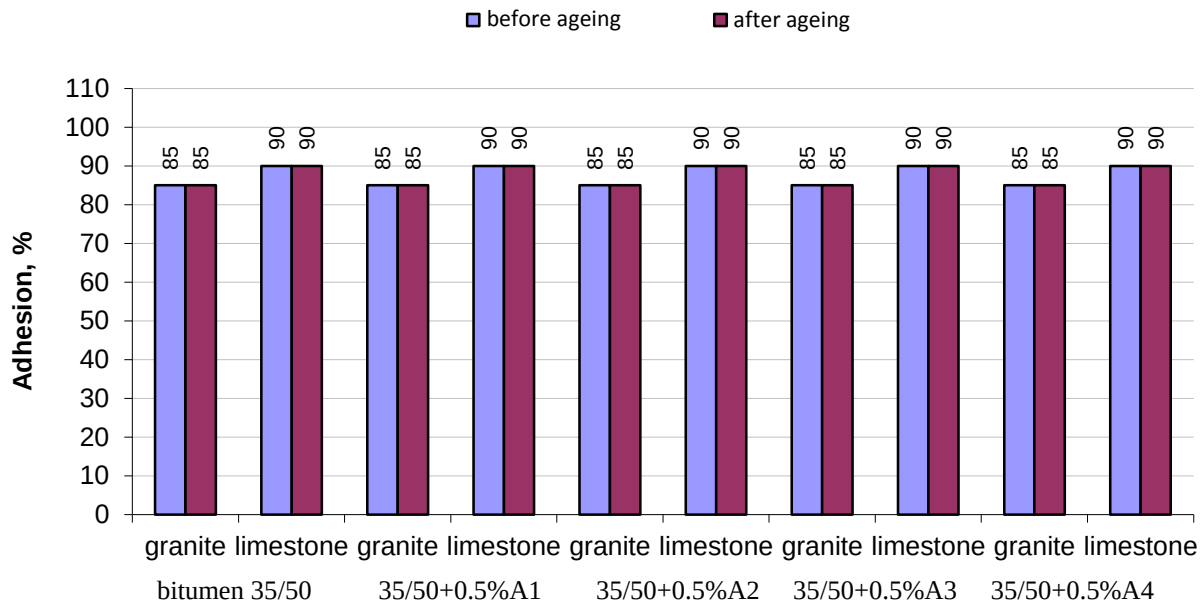

Fig.4. Adhesion of bitumen 30/50 to granite and limestone with the addition of adhesive agents determined by BOP IIB method

The studies showed that different methods of sample preparation ("BOP"), which are a modification of the standard "rolling bottle test", do not significantly affect the degree of washout of the bitumen from the surface of the aggregate and do not affect the results obtained for adhesion.

The adhesion of bitumen to the granite and limestone, determined by all the BOP methods, was comparable. The further freezing of samples, rolling in an aqueous NaCl solution, conditioning of the aggregate in water before and after coating with bitumen, had no effect on the degree of washout of the binder from the surface of the aggregate. It was found that a decisive effect on the adhesion of bitumen to the aggregate was the heating of the aggregate coated with bitumen at 160°C.

The adhesion of paving grade bitumen 35/50 to aggregates using the adhesive agents employed in the study determined by the BOP IB, BOP IIA, and BOP IIB methods was similar and ranged from 80-90%. The addition of an adhesive agent did not significantly affect the results.

The adhesion of bitumen to the alkaline aggregate determined by the BOP IB, BOP IIA and BOP IIB methods was slightly greater than adhesion to an acidic aggregate. However, the difference observed was not as great as was observed when samples were prepared according to standard procedures.

6. CONCLUSIONS

Summing up the range of results, the following basic conclusions can be made:

- The adhesion of bitumen to mineral aggregate is determined by mechanical factors - the degree of moistening and dust of the aggregate, the micro-texture of the aggregate, the granulation of the mineral mix and the physicochemical - chemical nature of aggregate (acidic, alkaline, in-between) as well as the physicochemical properties of the bitumen, such as viscosity and adhesion, which depend on the content of acidic compounds.
- In relation to the original procedure according to EN 12697-1 (method A) the basic difference of the options BOP-IB to BOP-IIC employed was the stage of heating the aggregate coated with a layer of bitumen. For this reason differences arose between the results of the standard procedure and the BOP options:
    - adhesion to limestone in both cases, standard and modified, was very good and was over 90%,
    - adhesion to granite was 10% in the standard method, and in the BOP modified options it ranged from 80-85% (even without the addition of an adhesive agent), which may be explained by the much higher viscosity of the adhesive coating the aggregate grains after heating at 160°C, moreover, the heating probably caused a significantly greater binding of binder to the surface (microstructure) of the grains,
    - adhesion of bitumen to granite and limestone, determined by all the BOP methods was comparable and was 80-90%; therefore, the type of material used to produce the aggregate (acidic, alkaline) did not affect the adhesion of bitumen; all the BOP options offset this impact,
    - additional freezing of samples, rolling in an aqueous solution of NaCl, conditioning the aggregate in water before and after coating with bitumen, had no effect on the degree of washout of the binder from the aggregate surface.
- Where different adhesive agents were used, the phase of heating aggregate coated with bitumen at 160°C for a period of 2 or 4 hours eliminates their impact on the rolling bottle test result.
- There were no significant differences in the degree of adhesion determined by the BOP methods, so the planned ITSR tests were not carried out on the asphalts.

7. SUMMARY

The test results demonstrate that the different methods for the determination of adhesion, which are a modification of the rolling bottle test according to EN 12697-11 (Method A), in which different mechanisms are used for separating the binder from the surface of the aggregate, do not affect the degree of washout of the bitumen from the surface of the aggregate and do not diversify the obtained results. A basic influence on this effect is exerted by the heating of aggregate coated with bitumen at 160°C for 2 or 4 h. This effect is so significant that it eliminates the conditioning with freezing, rolling in NaCl solution or the presence of the adhesive agent.
Taking into account the enormous impact of heating the bitumen-coated aggregate on the durability of the adhesion of the film of bitumen to the surface of the aggregate, including acidic aggregate, one can wonder about the impact of the storage time and temperature of the asphalt in the silo after production (usually at a temperature of about 160°C). The resulting increase in viscosity of the asphalt during this time on the surface of the aggregate should significantly increase the durability of the adhesion of bitumen to the aggregate surface.

BIBLIOGRAPHY

[1] Effect of aggregate properties on asphalt mixtures stripping and creep behavior. Construction and Building Materials, Abu-Qudais S., Al-Shweily H. Volume 21, Issue 9, Pages 1886–1898, September 2007.

[2] The influence of aggregate, filler and bitumen on asphalt mixture moisture damage. Construction and Building Materials, Airey G.D. Collop A.C., Zoorob S.E., Elliott R.C. Volume 22, Issue 9, Pages 2015–2024, September 2008.

[3] Wyniki programu badawczego dotyczącego przyczepności asfaltu do kruszywa (The results of a research programme on bitumen-aggregate affinity) Błażejowski K., Olszacki J., Peciakowski H.. The eSeMA'2013 Rettenmaier Conference, Zakopane, February 2013.

[4] Durability of asphalt mixtures: Effect of aggregate type and adhesion promoters. International Journal of Adhesion and Adhesives, Cui S., Bamber R.K. Blackman, Anthony J. Kinloch, Ambrose C. Taylor, Volume 54, Pages 100–111, October 2014.
[5] A Synthesis of Theories and Mechanisms of Bitumen-Aggregate Adhesion Including Recent Advances in Quantifying the Effects of Water, Hefer A.W., Little D.N., Lytton R.L.
[6] Odporność mma na działanie wody – porównanie procedur i zastosowanie praktyczne (The resistance of asphalt mixtures for water – a comparison of procedures and practical applications), Krajewski M., Błażejowski K.. The eSeMA'2012 Rettenmaier Conference, Zakopane, February 2012.
[7] Test results of adhesion testing, Krajewski M., The eSeMA'2013 Rettenmaier Conference, Zakopane, February 2013.
[8] Weryfikacja i uaktualnienie metody badawczej wodoodporności z cyklem zamrażania mieszanek mineralno-asfaltowych (The verification of ITSR test method with one freezing cycle). Sybilski D., Zofka A., Bańkowski W., Horodecka R., Maliszewska D., Maliszewski M., Wróbel A., Mirski K. Road and Bridge Research Institute Report for General Directorate of Public Roads, Warsaw 2013
[9] The Shell Bitumen Handbook. Fifth Edition. Read J., Whiteoak D, Thomas Telford, 2003.
[10] Prawdy i mity związane z powinowactwem pomiędzy asfaltem i kruszywem. (Truths and myths on bitumen-aggregate affinity), Witczak S., The eSeMA'2014 Rettenmaier Conference, Zakopane, February 2014.
[11] EN 12697-11 Bituminous mixtures. Test methods for hot mix asphalt. Determination of the affinity between aggregate and bitumen.
[12] EN 12697-12 Bituminous mixtures. Test methods for hot mix asphalt. Determination of the water sensitivity of bituminous specimens.
[13] EN 12607-3 Bitumen and bituminous binders - Determination of the resistance to hardening under the influence of heat and air – Part 3: RFT Method.
[14] EN 12591 Bitumen and bituminous binders - Specifications for paving grade bitumens.