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Transportation Research Procedia 14 (2016) 724 – 731

Transportation Research Procedia

www.elsevier.com/locate/procedia

6th Transport Research Arena April 18-21, 2016

Evaluation of different test methods for bitumen adhesion properties

Miglė Paliukaitė [a,*], Viktoras Vorobjovas [a], Matas Bulevičius [b], Vitalijus Andrejevas [a]

[a] *Vilnius Gediminas Technical University, Linkmenu str. 28, 08217 Vilnius, Lithuania*
[b] *JSC Problematika, Galvės str. 2, 02241 Vilnius, Lithuania*

Abstract

The adhesion between mineral aggregates and bitumen is an important criterion which describes the quality of the asphalt mixture, asphalt pavement performance and resistance to distress. The lack of bonding can lead to a significant asphalt pavement damage. For the evaluation of the adhesion behavior between bitumen and aggregates, used in road construction, many test methods are known. Therefore, it is important to assess the most appropriate test method for bitumen adhesion properties. In the European standard three testing methods are used to determine the affinity between bitumen and aggregate: Rolling-Bottle-Test, static water storage, detachment in boiling water. Whereas, in the Lithuanian standard – boiling water test method is used. The aim of this research is to determine an effective test method for bitumen and aggregates adhesion properties. Also, to investigate the validity of the test methods for the Lithuanian asphalt binders and to compare the adhesion test methods and parameters used in European standard. The materials used in the experiment research: 5 types of aggregates (granite, dolomite, crushed gravel, and quartz diorite), 6 types of asphalt binders used in Lithuania market (50/70, 70/100, and PMB 45/80-55) and 2 additives – Iterlene 400/IL, Antrocelbond. The test results showed a difference in bitumen adhesion according to test methods used in European and Lithuanian standards, since these methods differ in their methodology, preparation of the samples and results of the measurement unit. Based on the results of this research the conclusions and recommendations were made for determination of bitumen and aggregates adhesion in Lithuania.

Peer-review under responsibility of Road and Bridge Research Institute (IBDiM)

Keywords: Bitumen-aggregate adhesion; adhesion promoters; adhesion testing methods; moisture damage

* Corresponding author. Tel.: +370-5-251-2354; fax: +370-5-273-1020.
E-mail address: m.paliukaite@gmail.com

Peer-review under responsibility of Road and Bridge Research Institute (IBDiM)
doi:10.1016/j.trpro.2016.05.339

1. Introduction

The question of what test method to specify for the effective control of bitumen and aggregate adhesion properties is a difficult one to answer. Numerous research efforts, of which many started long ago, have been conducted to understand the conception of the adhesion and to find the most appropriate test method(s) to determine the affinity between bitumen and aggregate (Rice 1958; Terrel and Shute 1989; Curtis 1992; Robertson 2000; Cui et al. 2014; Liu et al. 2014).

Bitumen and aggregates are considered as the main constituents of an asphalt mixture and their physical and chemical properties have a direct influence on the performance of the mixture. The literature describes four primary theories that explains the concept of bitumen/aggregate interaction: chemical reaction, surface energy, molecular orientation, and mechanical adhesion. The theories explain the adhesive bond between bituminous binder and aggregate (Terrel and Shute 1989; Curtis 1992; Moraes et al. 2011).

A lack of compatibility between bitumen and aggregate is one of the main causes of moisture damage in an asphalt mixture. Moisture damage can manifest itself through various failure mechanisms such as rutting, raveling, potholes and fatigue cracking. For that reason many researches have been trying to define the bitumen-aggregate mechanism and to improve adhesion test methods (Renken et al. 2010; Copeland et. al. 2008; Grenfell et al. 2014; Moraes et al. 2011; Daučík et al. 2011; Liu et al. 2014).

In order to extend the service life of the pavement, several countries have implemented national requirements of mandatory addition of adhesion promoters in asphalt mixes, to secure and maintain the adhesion and durability of asphalt pavements over time. In general, adhesion between bitumen and aggregate depends on the chemical nature of the components and therefore the source of the bitumen and type of aggregate. Other aggregate properties such as surface texture, shape, porosity and absorption will also influence the adhesion. The use of appropriate adhesion additives is a vital importance to ensure the strong bond (adhesion) to the aggregate surface at the very beginning (Liu et al. 2014).

In Lithuanian technical document for asphalt mixes is pointed that the affinity between bitumen and aggregates should be ensured, but the test methods and limit values are not mentioned. Also, in Europe there is not common practice which method to use for determination of the affinity between bitumen and aggregates. Therefore, there are differences of particular methods and it makes a need to determine the appropriate test method for determination of adhesion and limit values.

The first objective of this study is to assess an effective test method for bitumen and aggregate adhesion properties. The second objective is to investigate the validity of the adhesion test methods for the Lithuanian bituminous binders and to recommend the adhesion limit values ensuring the compatibility between bitumen and aggregate.

The scope of this research is to compare different materials used in the experiment research: 5 types of aggregates (granite, dolomite, crushed gravel, and quartz diorite), 6 types of asphalt binders used in Lithuania market (50/70, 70/100, and PMB 45/80-55 from different producers) and 2 additives – Iterlene 400/IL, Adhezin.

The study focused on how to prove the most appropriate testing method effectiveness to asphalt adhesion properties and to recommend the adhesion limit values measured by one of test methods'.

2. Background

2.1. Theoretical review of adhesion

Adhesion between bitumen and aggregate is one of the most important criteria for a highly durable and water damage resistant asphalt pavement. The bitumen-aggregate bond is a surface phenomenon which depends on the contact between the two materials. Terrel and Shute (1989) describe four theories that are often used to explain the adhesion between asphalt and aggregate: chemical reaction, surface energy, molecular orientation, and mechanical adhesion. There are few factors by which these theories are affected: surface tension of the bitumen and aggregate, chemical composition of the bitumen and aggregate, bitumen viscosity, surface texture of the aggregate, aggregate porosity, aggregate cleanliness, aggregate moisture content and temperature at the time of mixing with bitumen (Terrel and Shute 1989, Bagampadde et al. 2004, Ramon-Torregrosa et al. 2008, Grenfell et al. 2014). Chemical reaction is based on the acidic and basic components of both bitumen and aggregate reaction. Robertson (2000)

states that adhesion between bitumen and aggregate arises between the polars of the bitumen and the polar surface of the aggregate. He also claims that polarity alone in bitumen is not sufficient to achieve good adhesion in pavements because bitumen is affected by the environment. Molecular orientation is related with surface energy. It assumes that adhesion between bitumen and aggregate is facilitated by a surface energy since reduction at the aggregate surface as bitumen is adsorbed to the surface (Rice 1958, Tarrer and Wagh 1991). Mechanical adhesion relies on physical aggregate properties, including surface texture, porosity or absorption, surface coatings, surface area, and particle size (Terrel and Al-Swailmi 1994).

The type of aggregate also has a big influence to adhesion properties. Aggregates are commonly classified as hydrophobic (acidic aggregates) and hydrophilic (basin aggregates) which react different in the adhesion process (Tarrer and Wagh 1991). Also, surface texture of the aggregate affects its ability to be properly coated since good initial coating is necessary to prevent stripping (Maupin 1982). Tarrer and Wagh (1991) list a number of factors that influence the bitumen–aggregate adhesion: surface texture, penetration of pores, aggregate angularity, aging of the aggregate surface, adsorbed coatings on the surface of the aggregate, and the nature of dry aggregates versus wet aggregates. Rough surfaces and therefore larger contact area are preferred for better adhesive bond (Moraes et al. 2011). Cui et al. (2014) claimed that acidic aggregates (granite) showed a greater loss of adhesion than basic aggregates (limestone and marble) under wet conditions.

2.2. Basic adhesion test methods

For the evaluation of adhesion behavior of hot mix asphalt many testing methods are known. In the European standard EN 12697-11 three testing methods are described for the determination of affinity between aggregate and bitumen: rolling-bottle-test, static water storage, detachment in boiling water. Whereas, in the Lithuanian standard LST 1362.23 – boiling water test method is used.

In Germany a research was done on the applicability and suitability of the testing methods for bitumen adhesion properties. Tests were done according to European standard EN 12697-11 methods. Also, a direct tensile strength test on asphalt mixtures and the contact angle measurement were carried out. The advantages and disadvantages of adhesion testing methods were investigated. Results showed that the most useable and simple test method is rolling-bottle-test. The final results were interpreted using computer-aided analysis technique based on digital picture analysis (Renken et al. 2010).

Recently the present authors showed that rolling-bottle-test is related to be the most suitable testing method despite disadvantages due to the mechanical strain by the rolling process. Static water storage brought no differentiation of results and boiling water test gave not accurate testing results (Renken et al. 2010). Hugener et al. (2012) analyzed the static method improving some procedures and testing conditions. For example, one of the major impact factors such as mixing conditions and the temperature for water conditioning have been studied in more details. Liu et al. (2014) determined that bituminous mixtures containing limestone aggregates have better moisture resistance than granite aggregates based on results from moisture sensitivity test. Also, it was observed the boiling water test and rolling-bottle-test were the most sensitive while the static immersion test was the least sensitive.

2.3. Adhesion promoters

In order to avoid the problems such as stripping, moisture damage, adhesion promoters can be added in the bitumen binder. They act as a bridge between bitumen and aggregates which resist the displacing effect of water and improve the adhesion of bitumen on the aggregates (Kim et al. 2012). Better understanding of the bitumen–aggregate interactions would help to optimize the use of adhesion promoters. Boulangé et al. (2013) developed a new physicochemical method to characterize the bitumen–aggregate interface. It was found that a porosity effect corresponding to the smallest scale roughness leads to a better resistance to stripping phenomena. Also, chemical effect at the interface bitumen–aggregate, due to the addition of adhesion promoters in bitumen, improved the water sensitivity of the bitumen explained by an increase of the aromatics/resins ratio. Cui et al. (2014) determined the effect of aggregate type and adhesion promoters to asphalt mixture durability. He suggested that the interfacial adhesion in wet conditions can be improved by mixing a silane, amine or rubbery polymer into the bitumen.

3. Materials and testing methods

3.1. Materials

Two different bituminous binders including PMBs from different producers were used for the tests covering the most important range of bituminous binders used in Lithuania. Properties of bitumen are provided in Table 1. The samples were prepared mixing two types of adhesion promoters – Iterlene IN-L, Antrocelbond and compared with original bitumen. The amount of adhesion promoter was used 0.2%, 0.4% and 0.6% (by weight of bitumen).

The adhesion promoter Iterlene IN/400-L consists of alkylamidopoliamine. The adhesion promoter Antrocelbond is patented (No. WO 2012152459 A1) and consists of mixture of amphipathic compounds (60–100%), oil mixture (10–40%), also mixture of aromatic and aliphatic hydrocarbons (1–20%). Further, the adhesion promoters are coded as Promoter 1 and Promoter 2 due to confidentiality.

Aggregates (granite and dolomite) that have been widely used in Lithuania for hot asphalt mixes, were selected in this research. Properties of aggregates are presented in Table 2.

In total, 64 samples were prepared for the bitumen-aggregate adhesion test methods using different kind of additives.

Table 1. Properties of bitumen samples.

Properties	Test methods	Type of bitumen, producer			
		PMB 45/80-55 E Producer 3	PMB 45/80-55 E Producer 2	50/70 Producer 1	50/70 Producer 2
Penetration, mm-1	EN 1426	54	59	63	60
Softening point, °C	EN 1427	59.0	59.0	48.3	49.5
Elastic recovery at 25°C, %	EN 13398	83	82	–	

Table 2. Properties of aggregates.

Properties	Test methods	Aggregate	
		Dolomite 8/11	Granite 8/11
Grading category	EN 933-1	$G_C90/15$	$G_C90/25$
Fines content	EN 933-1	f_1	f_1
Shape index	EN 933-4	SI_{15}	SI_{15}
Water absorption after 24 h	EN 1097-6	$WA_{24}2$	$WA_{24}1$
Freeze-thaw resistance values	EN 1367-1	F_1	F_1

3.2. Methods

The affinity between aggregate and bitumen was determined by three methods:

- boiling water method according to Lithuanian National Standard LST 1362.23;
- rolling-bottle-test according to standard EN 12697-11, clause 5;
- static method according to standard EN 12697-11, clause 6.

Testing methods consist of three stages. In each stage samples were tested using Boiling water and Static methods. Visual determination of the bitumen coverage was carried out by one operator independently. The samples tested using Rolling-bottle-test were carried out by two operators.

According to EN 12697-11 standard and the Lithuanian National Standard LST 1362.23 particles of a specific aggregate size are selected and coated with hot bitumen. After cooling down, the aggregates are immersed in water for a defined time and a specific water temperature or are placed into glass bottle with distilled water for rolling

procedure. If the adhesion strength is not sufficient, water is infiltrating between the thin bitumen layer and the mineral surface, resulting in a weakening or even partial removal of the bitumen film. This can be evaluated by assessing the degree of coating for each mineral aggregate and the adhesion property is characterized by the so called coating index. In table 3 the procedures of the three different test methods for the assessment of the adhesion property are compared.

4. Analysis of results

Surface of the aggregate particles covered by bitumen have been estimated visually. In total, 64 samples were tested. Bitumen coverage of different bitumen, adhesion promoter and granite aggregates are presented in figures 1–3 by the test methods. The coating degree of different bitumen, aggregates and adhesion promoters determined by static method according to standard EN 12697-11 is 100% in all cases.

Table 3. Differences between European and Lithuanian standards for adhesion properties

	EN 12697-11		LST 1362.23
	Rolling-bottle-test	Static method	Boiling water test
Aggregate coating			
Number of samples	3	1 or 4	2
Binder amount	16 g (3.0%)	4% (±x·0.5%)	2.5%
Aggregate size	8/11.2 (5.6/8, 6.3/10) mm	8/11.2 or 6/10 mm	8/11.2 mm
Aggregate amount	510 g for 3 sets	150 particles	50–100 g
Mixing temperature	variable, according to EN 12697-35	variable, according to EN 12697-35 -15°C	variable, according to EN 12697-35
Cooling time	12–64 h	60 min	15 min
Water conditioning			
Conditioning type	rolling bottle	static	static
Water temperature	20±5°C	19±1°C	100±5°C (boiling water)
Conditioning time	24±1h	48±1 h	30±1 min
Assessment of the coating degree			
Condition of the aggregates	wet	dry	wet
Validation of the result	visual estimation of the coating proportion after 6 and 24 h	number of not completely coated aggregates (visual)	visual estimation of the coating proportion with graphical aids
Result	coating coefficient in 5% steps	passing yes/no	coating coefficient in 5% steps

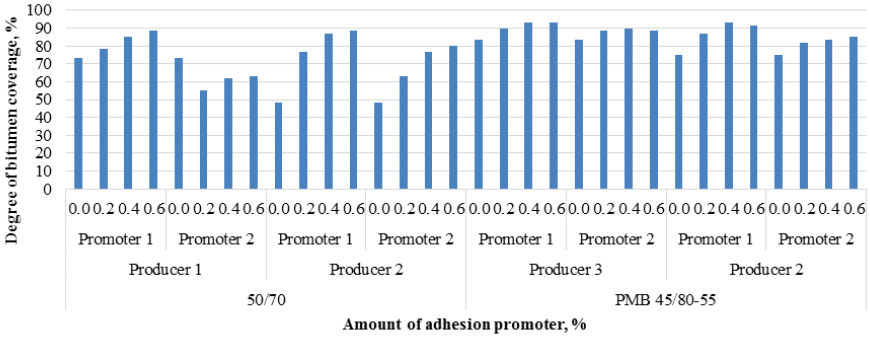

Fig. 1. Bitumen coverage of different bitumen and granite aggregates after Rolling-bottle-test 6 hours.

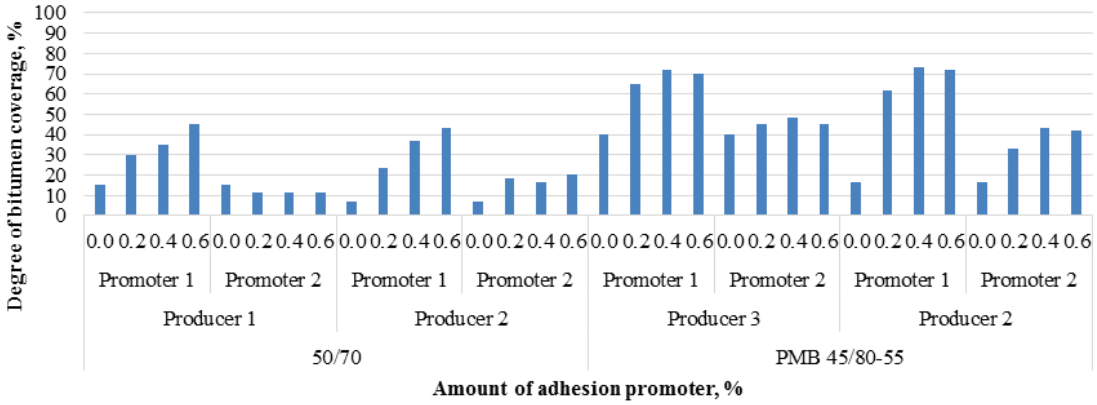

Fig. 2. Bitumen coverage of different bitumen and granite aggregates after Rolling-bottle-test 24 hours.

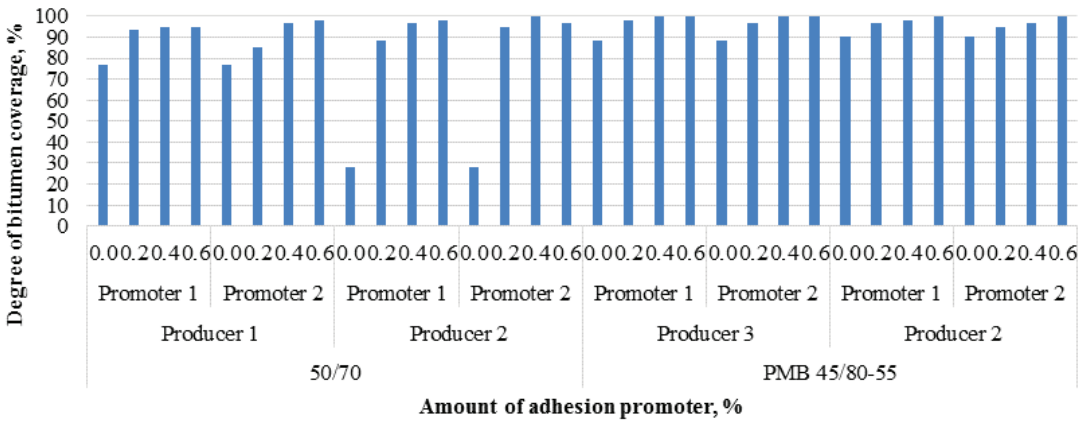

Fig. 3. Bitumen coverage of different bitumen and granite aggregates after boiling water test.

The rolling-bottle-test results after 6 hours show that the coating degree of different bitumen and granite aggregates without adhesion promoters varies from 40% to 83%. Also, mixtures with PMB show the best coverage. The use of 0.4% adhesion promoter increases the coating and the coverage is more than 60% in all cases. The coverage of PMB is about 20% higher than 50/70 bitumen. Also, the results show that coverage is about 20% higher with adhesion Promoter 1 instead of Promoter 2.

The results of the same test method after 24 hours of rolling show that the coating degree of different bitumen and granite aggregates without adhesion promoters varies from 5% to 40%. And the coating increases from 7% to 73% using 0.4% adhesion promoter. Also, mixtures with PMB and adhesion Promoter 1 shows the best coverage.

The bitumen coverage, determined by the boiling water test, according Lithuanian national standard differ from rolling-bottle-test. The results show that the coating degree without adhesion promoters varies from 23% to 90% and in all cases is more than 80% with PMB. The use of 0.4% adhesion promoter increases the coating and the coverage more than 90%. The rolling-bottle-test shows that the coverage of bitumen is higher using adhesion Promoter 2 instead of Promoter 1.

Bitumen coverage of different bitumen, adhesion promoter and dolomite aggregates are presented in figures 4–6 by test methods.

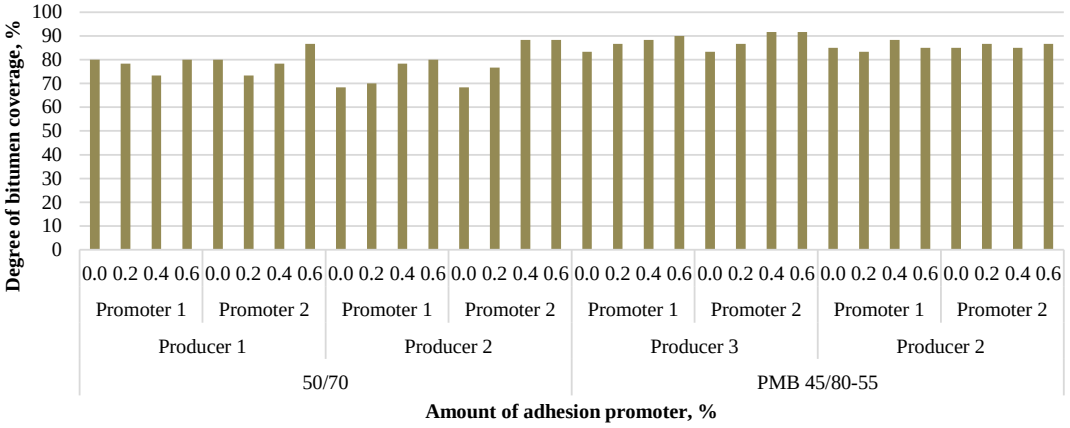

Fig. 4. Bitumen coverage of different bitumen and dolomite aggregates after Rolling-bottle-test 6 hours.

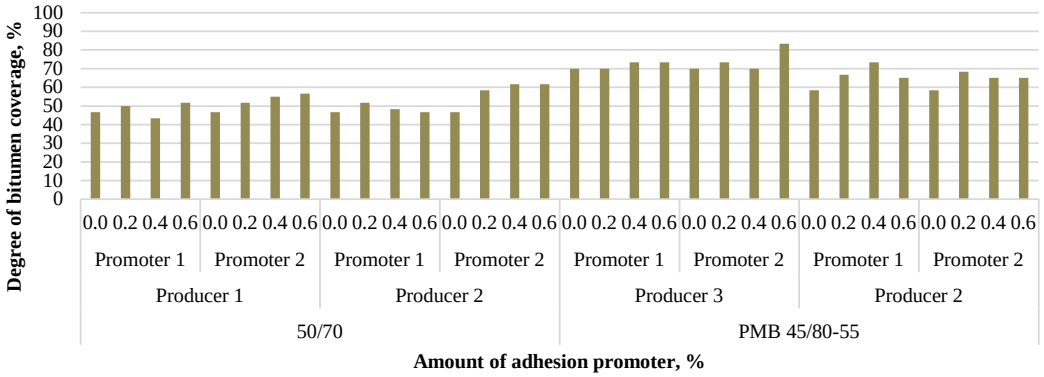

Fig. 5. Bitumen coverage of different bitumen and dolomite aggregates after Rolling-bottle-test 24 hours.

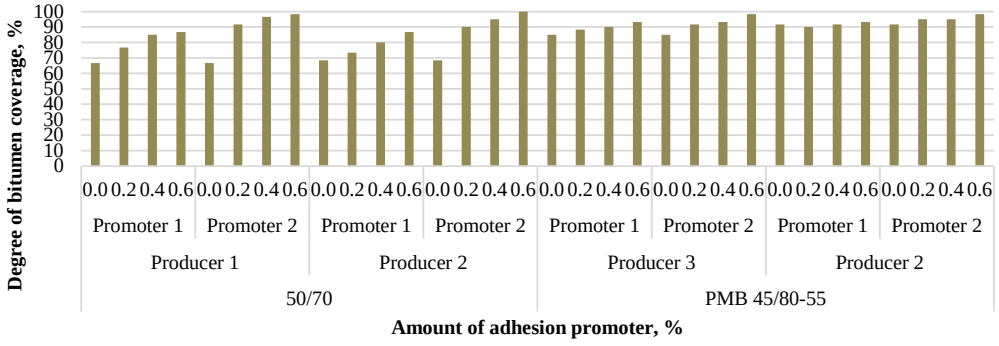

Fig. 6. Bitumen coverage of different bitumen and dolomite aggregates after boiling water test.

The analysis of the rolling-bottle-test results after 6 hours shows that the coating degree of different bitumen and dolomite aggregates without adhesion promoters varies from 65% to 85%. Mixtures with PMB shows the best coverage (> 80%). The use of 0.4% adhesion promoter increases the coating and the coverage is more than 70% in all cases. The coverage of bitumen 50/70 with Promoter 2 is 10% higher than with Promoter 1.

The results after the same test 24 hours rolling shows that the coating degree of different bitumen without adhesion promoters varies from 32% to 70%. And the coating increases from 40% to 73% using 0.4% adhesion promoter. Also, mixtures with PMB and adhesion Promoter 2 shows the best coverage.

The bitumen coverage results determined by the boiling water test made according to Lithuanian national standard shows that the coating degree without adhesion promoters varies from 60% to 92% and in all cases is more than 80% with PMB. The use of 0.4% adhesion promoter increases the coating and the coverage is more than 80%. Comparing to the rolling-bottle-test the coverage of bitumen is 10% higher with adhesion Promoter 2 instead of Promoter 1.

5. Conclusions

Given the results presented in this paper, the following summary and conclusions are offered:

- Test methods show the direct affinity between bitumen and aggregates with some disadvantages such as visual estimation of coating.
- Rolling-bottle-test method is provided as the most suitable test method. The most stable results were determined after 6 hours of rolling.
- The limit value (60%) for bitumen coating of granite and dolomite aggregates was set using Rolling-bottle-test after 6 hours of rolling.
- Rolling-bottle-test after 6 hours of rolling shows that mixtures with polymer modified bitumen increase the bitumen/aggregate adhesion more than 80%.
- The adhesion Promoter 1 is more suitable using granite aggregates and adhesion Promoter 2 – mostly for dolomite aggregates.

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