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EVALUATION OF POLYAMINOAMIDE AS A SURFACTANT ADDITIVE IN HOT MIX ASPHALT

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Abstract. The phenomenon of breaking the bond between the aggregates and the bitumen is known as stripping. Stripping of asphalt films from the surface of aggregate particles results in premature failure of asphalt pavement. This causes weakening of pavement resistance to rutting and fatigue. Furthermore, moisture damage increases the susceptibility of pavement to reveling, a distress that causes the loss of skid resistance on surface of the road and deterioration of pavement. Surfactant additive or adhesive agent is a surface-active agent that changes (lowers) the surface tension of rock materials. Introduction of surfactant additive results in increased strength of adhesive bond between bitumen and the rock materials surface preventing stripping throughout the service life of the asphalt concrete. Polyaminoamide is an organic water soluble compound that allows waterproofing mineral aggregate surfaces and acts as a bonding agent to bitumen. The objective of this research is to study the effect of polyaminoamide based and poliphosphoric acid based liquid additives on stripping, moisture susceptibility, rutting and fatigue performance of asphalt concrete. In this paper, boiling water test was used to determine the percentage of stripped aggregates after boiling. The moisture susceptibility of asphalt mixtures was investigated by means of testing the retained indirect tensile strength after water immersion using Marshal stability test method. Wheel tracking test was also conducted on asphalt slabs prepared in the laboratory to determine rut resistance. Asphalt concrete with commonly used mineral filler was chosen as a control mixture. It was found that the adhesion additive not only improves stripping resistance, but also slightly improves asphalt rut resistance.

Keywords: polyaminoamide, asphalt, moisture susceptibility, water resistance, rut resistance.

1. Introduction

Environmental factors such as temperature, air and water can have a profound effect on the durability of asphalt concrete mixtures (Attaelmanan *et al.* 2011; Haritonovs *et al.* 2013). In mild climatic conditions where good-quality aggregates and asphalt cement are available, the major contribution to the deterioration may be traffic loading, and the resultant distress manifests as fatigue cracking, rutting and reveling (Terrel, Al-Swailmi 1994; Topal *et al.* 2011). However, when a severe climate is in question, these stresses increase with poor materials, under inadequate control with traffic as well as with water which are key elements in the degradation of asphalt concrete pavements (Topal *et al.* 2011).

There are a variety of material characteristics in both the asphalt binder and aggregate that contribute to moisture damage. There are three mechanisms by which moisture degrades asphalt: loss of cohesion within the bitumen or mastic, adhesive failure between aggregate and bitumen, and degradation of aggregate (Bahia *et al.* 2012; Copeland *et al.* 2007).

There are two main characteristics of asphalt binders that are important to stripping: viscosity and bitumen chemistry. It has been observed that binders of high viscosity are able to resist displacement by water much better than low viscosity binders. However, solving the moisture damage problem by simply specifying highly viscous binders would be detrimental to overall performance of the pavement in terms of workability, low temperature cracking, and fatigue cracking (Hanz *et al.* 2007; Zegeye *et al.* 2012). Bitumen chemistry is dependent on two factors: crude oil source and refining methods. Studies by Peterson cited in Kanitpong identified bitumens containing compounds such as certain forms of carboxylic acids and sulfoxides have displayed more moisture susceptibility (Kanitpong 2004). Different chemical reactions used in refining of the crude oil have the potential to leave undesirable reactants in crude oil which will be presented in bitumen binder.

Mineralogical and physical properties of aggregates also contribute significantly to the bitumen-aggregate bond and its resistance to stripping (D'Angelo, Anderson

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2003). The mineralogical properties of aggregate are most important for stripping in the aggregate affinity for water. Aggregate physical properties such as roughness, porosity and dust coating all greatly affect adhesive strength. Surface roughness increases bond strength by providing more surface area to accommodate the bitumen-aggregate bond. Furthermore, an optimum level of porosity is desirable to allow more interlocking in the bond between the bitumen and aggregate (Punith *et al.* 2012). Finally, the aggregate should be as clean from dust as practically possible. Dust coating the aggregate does not allow the bitumen binder to bond directly to the aggregate, creating a space between the bitumen film and surface of the aggregate (D'Angelo, Anderson 2003).

In order to improve moisture resistance to acidic aggregate (for example granite) the use of surfactant additives such as amine-based liquids, dolomite powder or Portland cement are commonly used treatment methods (Cheng *et al.* 2002; Little, Petersen 2005). These adhesive agents are used to increase the physically chemical bond between bitumen and aggregate as well as to improve wetting by lowering the surface tension of the bitumen (Curtis *et al.* 1993).

The main objective of this study is to evaluate the effect of polyaminoamide based and pholiphosphoric acid based liquid on the stripping properties, moisture susceptibility and performance characteristics of asphalt concrete mixtures. For these purposes boiling water test was used to determine the percentage of stripped aggregate after boiling. The moisture susceptibility of asphalt mixtures was investigated by means of testing the retained indirect tensile strength after water immersion using Marshal stability test method. Wheel tracking test was conducted on asphalt slabs prepared in the laboratory to determine rut resistance.

2. Raw materials

The basic mineral materials used in this study are crushed dolomite aggregate (fraction 8–11mm); crushed granite aggregate (fraction 8–11mm) and crushed diabase aggregate (fraction 8–11mm). In this study bitumen 70/100 from three different sources was used: Orlean Lithuania [OL], BDUS 70/100 Russia [BR] and Nynas Estonia [NE]. These aggregates and bitumens are conventional materials used extensively for local mixes.

2.1. Properties of aggregates

Table 1 contains test results of the basic aggregates used in this study.

The properties of granite and diabase aggregates correspond to the highest category of *LVS EN 13043 Aggregates for Bituminous Mixtures and Surface Treatments for Roads, Airfields and other Trafficked Areas* standard. The aggregate test results show very low flakiness index: granite – 7, diabase – 9 and dolomite – 12. Granite and diabase has excellent resistance to fragmentation (average LA value of 15 and 17) and high frost resistance (average MS value of 0.3 and 0.7). Dolomite showed a slightly lower fragmentation resistance with average LA value of 22 and low fines content (0.9%).

It is important to note, that dolomite is sedimentary carbonate rock, diabase is a basic igneous rock and granite due to high silica content is acid igneous rock. Bitumen together with carbonate and basic rock with low SiO_2 content creates water resistant chemical link, while granite usually has lower bound with bitumen due to high SiO_2 content.

2.2. Bitumen properties

Properties of the bitumens were determined for delivery conditions and after thermal exposure using the rolling thin film oven test (RTFOT) according to *LVS EN 12607-1 Bitumen and Bituminous Binders – Determination of the Resistance to Hardening under the Influence of Heat and Air – Part–1: RTFOT Method*.

The following characteristic values were identified – softening point, needle penetration, Fraas breaking point, aging, dynamic and kinematic viscosities. The results of the tests are summarized in Table 2.

2.3. Surfactant additives properties

Two liquid adhesion agents have been evaluated in this study: polyaminoamide based [PAA] and pholiphosphoric acid based [PPA] additives. The physical and chemical properties of additives are presented in Table 3.

Table 1. Physical and mechanical characteristics of aggregates

Physical and mechanical properties	Unit	Related standard	Value		
			Dolomite aggregate	Granite aggregate	Diabase aggregate
Los Angeles (LA) coefficient	%	*LVS EN 1097-2*	22	15	17
Resistance to wear. Nordic test (A_N)	%	*LVS EN 1097-9*	15.7	12.5	14.0
Flakiness Index (FI)	%	*LVS EN 933-3*	12	7	9
Water absorption	%	*LVS EN 1097-6*	2.7	2.0	2.0
Grain density	Mg/m^3	*LVS EN 1097-6*	2.80	3.02	3.05
Fine content	%	*LVS EN 933-1*	0.9	1.3	1.6
Freeze/thawing (MS)	%	*LVS EN 1367-2*	9	0.3	0.7

Boiling water test was carried out to determine aggregate – bitumen interaction and to ensure for a high performance of the surfactant and to reveal possible problems regarding the susceptibility of the bitumen-aggregate bond to stripping.

3. Evaluation of bitumen-aggregate interaction

Boiling water test is a conventional test method accepted in Latvia. In this test two samples of fully bitumen covered mineral aggregates (fraction 8/11) are prepared a day before the test. The aggregate sample weight is about 600 g and the bitumen weight is around 16 g (aggregate density dependent). Each bitumen coated sample is placed on wire gauze inside glass beaker and distributed homogeneously at the center of the wire gauze. The glass beaker is filled with 650 ml de-ionized water and heated to 100 °C in a period of 1 to 3 min. The sample is boiled for 30 min. After this thermodynamic exposure the wire gauze with sample are removed from the boiling water. It is important that any loose bitumen on the water surface is being removed with filter paper prior to sample extraction, so that the sample is not contaminated with floating bitumen. After taking the wire gauze out of the boiling water it is cooled down in water bath. The mineral aggregates are later carefully removed from the bath and placed on a Teflon pan. The bitumen stripping degree with accuracy of 5% is determined. Figs 1–6 present the visually inspected results of the prepared samples.

The addition of anti-stripping additives has resulted in increased stripping resistance of bitumen mixes with granite and dolomite aggregates. The test results show slightly different adhesion when binders from different refineries are used with the same mineral aggregates, confirming that stripping resistance depends on the binder origin and chemical composition. The diabase aggregates in combination with B70/100 from Orlean Lithuania and BDUS B70/100 from Russia provide 100% adhesion even without addition of any additive.

4. Evaluation of asphalt mixture properties

4.1. Indirect Tensile Strength Test

Marshall Mix design procedure was used for the design of AC 11 mixture. Mix gradation was selected on the basis of the design method recommended by Latvian Road Specifications 2012. The Indirect Tensile Strength Ratio (ITSR) according to *LVS EN 12697-12 Bituminous Mixtures – Test Methods for Hot Mix Asphalt – Part 12: Determination of the Water Sensitivity of Bituminous Specimens* is a common test method to rank asphalt mixtures according to their susceptibility to moisture. In this test the indirect tensile strength of dry specimen is compared with the indirect tensile strength of water saturated specimen after 72 h storage in water at 40 °C. The water saturated specimens were conditioned in an exhausted and water filled vacuum desiccator. The ITSR is calculated as follows:

Table 2. Typical characteristics of the bitumens

Parameter	Origin			Standard
	Orlean Lithuania	BDUS Russia	Nynas Estonia	
Penetration at 25°C, dmm	79.9	80.0	88.0	*LVS EN 1426*
Softening point, °C	45.9	48.3	43.8	*LVS EN 1427*
Fraass temperature °C	–20.0	–19.2	–19.7	*LVS EN 12593*
Kinematic viscosity, mm^2/s	326	288	328	*LVS EN 12595*
Dynamic viscosity, Pa·s	137	115	159	*LVS EN 12596*
Aging characteristics of bitumen under the influence of heat and air (RTFOT method)				
Retained penetration, %	65.7	–66.8	65.0	*LVS EN 1426*
Increase of a softening point, °C	4.7	3.9	5.5	*LVS EN 1427*
Fraass breaking point after aging, °C	–15.0	–	–	*LVS EN 12593*

Table 3. Typical physical and chemical properties of anti-stripping additives

Properties	Polyaminoamide based liquid	Pholiphosphoric acid based liquid
Physical state	Liquid	Liquid
Color	Pale yellow	Brown
Odor	Amine	Ether
Auto ignition temperature, °C	>300	>300
Flash point, °C	>100	>140
Melting point, °C	–5	–10
Density, g/ml	0.96	1.1

$$ITSR = 100\% \frac{ITS_w}{ITS_D}, \% \quad (1)$$

where ITS_w – the average indirect tensile strength of the wet group, kPa; ITS_D – the average indirect tensile strength of the dry group, kPa.

The test specimens were prepared by using Marshall Hammer with 50 blows on each side according to *LVS EN 12697-30 Bituminous Mixtures – Test Methods for Hot Mix Asphalt –Part 30: Specimen Preparation by Impact Compactor Please*. In total 72 specimens were prepared (9 groups of 8 specimens in each group). Air voids content of each specimen was 3% ± 0.5%. The indirect tensile strength was determined using Marshal Stability testing device applying a constant deformation rate of 50 mm/min. The test results are displayed in Fig. 7. Due to the presence of water the adhesion between bitumen and aggregates of the control mix has been reduced. For the granite mixture using PAA and diabase mixtures using PAA as well as PPA the ITSR levels are higher than 100%. This is explained by the chemical and hydraulic characteristics of the mix.

4.2. Wheel tracking test

The resistance against rutting was determined by means of the wheel tracking test on the various mixtures containing different additives. Wheel tracking apparatus is used to

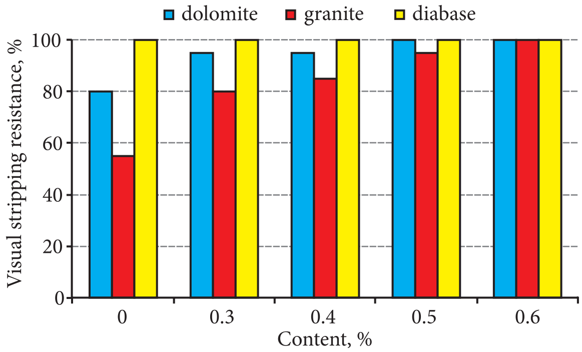

Fig. 1. Stripping resistance with bitumen B70/100 form Orlean Lithuania containing polyphosphoric acid based liquid

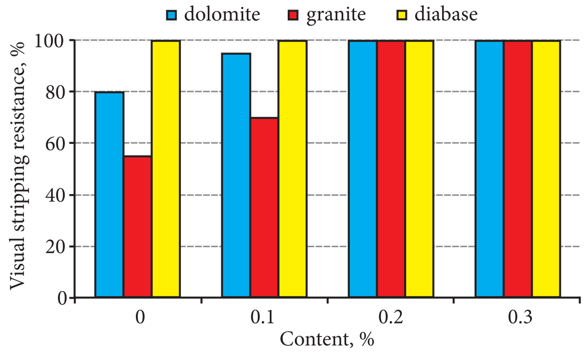

Fig. 2. Stripping resistance with bitumen B70/100 form Orlean Lithuania containing polyaminoamide based liquid

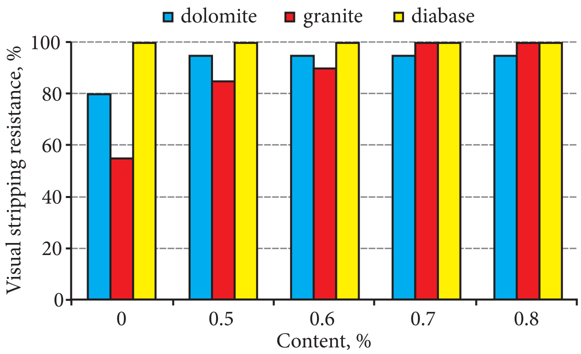

Fig. 3. Stripping resistance with bitumen BDUS 70/100 from Russia containing polyphosphoric acid based liquid

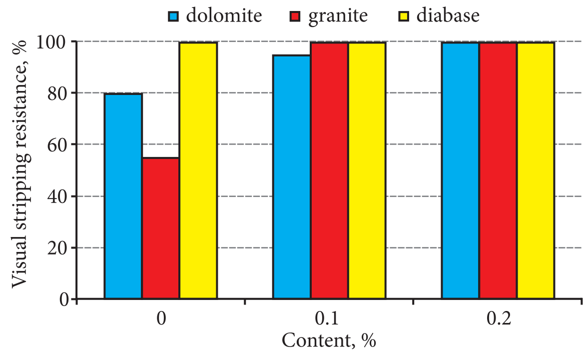

Fig. 4. Stripping resistance with bitumen BDUS 70/100 from Russia containing polyaminoamide based liquid

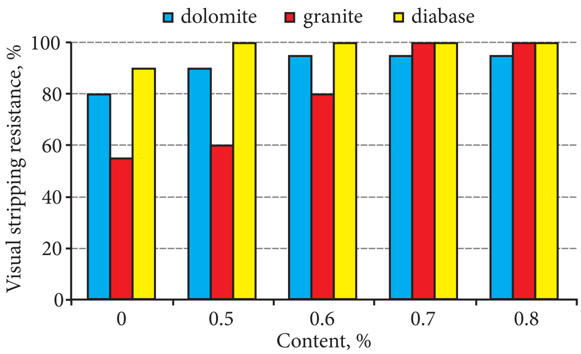

Fig. 5. Stripping resistance with bitumen B70/100 Nynas form Estonia containing polyphosphoric acid based liquid

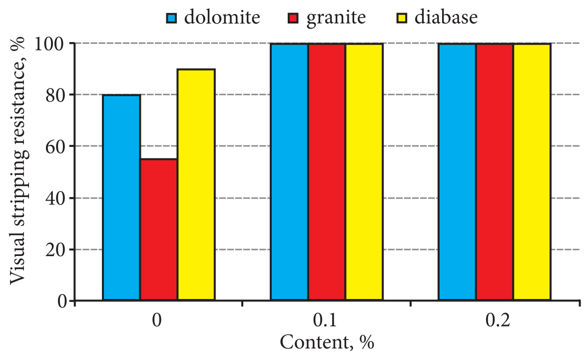

Fig. 6. Stripping resistance with bitumen B70/100 Nynas form Estonia containing polyaminoamide based liquid

simulate the effect of traffic and to measure the susceptibility to deformations of asphalt concrete samples. Tests were performed according to standard *LVS EN 12697-22 Bituminous Mixtures – Test Methods for Hot Mix Asphalt – Part 22: Wheel Tracking* method B (wheel tracking test with small size device in air). This test method is designed to repeat the stress conditions observed in the field and, therefore, can be categorized as simulative. The resistance of asphalt mixture to permanent deformations is assessed by measuring the depth of the wheel track and its increments caused by repetitive cycles (26.5 cycles/min) under constant temperature (60 °C). The rut depths are monitored by means of two linear variable displacement transducers (LVDTs), which measure the vertical displacements of each of the two wheel axles independently as rutting progresses. The wheel tracking slope in mm per 10^3 load cycles is calculated as:

$$WTS_{air} = \frac{RD_{10000} - RD_{5000}}{5}, \quad (2)$$

where WTS_{air} – the wheel tracking slope, mm/10^3 load; RD_{5000}, RD_{10000} – the rut depth after 5000 and 10 000 loading cycles, mm.

The asphalt concrete slabs have been produced by means of roller compactor in accordance to *LVS EN 12697-33 Bituminous Mixtures – Test Methods for Hot Mix Asphalt – Part 33: Specimen Prepared by Roller Compactor* standard. The results of rut resistance test are demonstrated in Fig. 8. The asphalt mixture produced using diabase aggregate with pholiphosphoric acid based adhesion additive has the lowest wheel tacking slope compared to the other types of aggregates and combinations with polyaminoamide based adhesion additive. Asphalt mixtures produced using granite aggregate show a little higher wheel tracking slope and rapid rut resistance improvement compared to control mix using either of the adhesion additives. Amino amide based adhesion additive does not significantly affect the rut resistance properties of mixtures with dolomite aggregates.

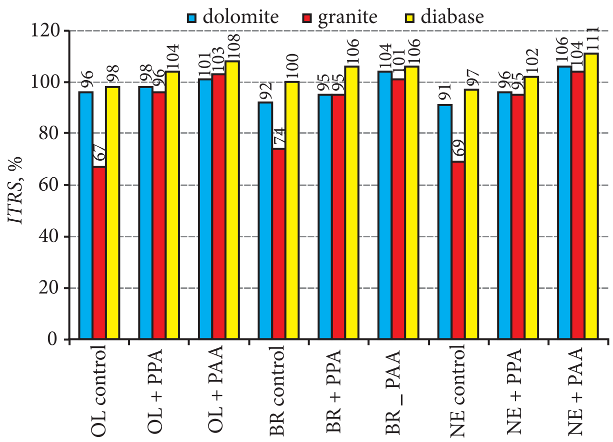

Fig. 7. *ITSR* for different adhesion additives and bitumens

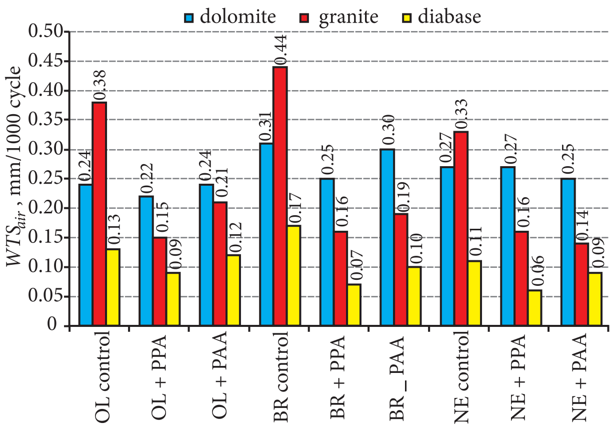

Fig. 8. Wheel tracking test results

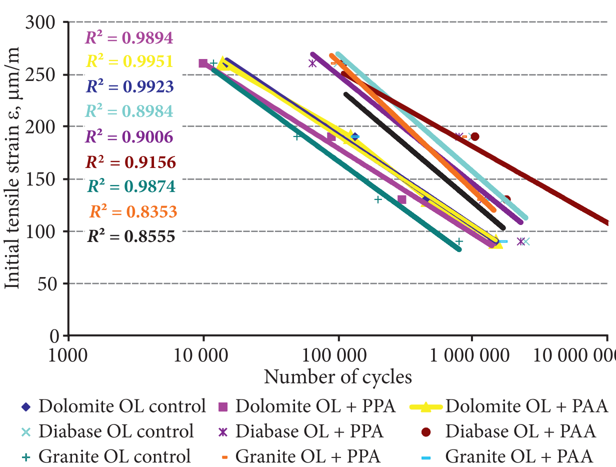

Fig. 9. Fatigue lines

4.3. Fatigue test

To determine the fatigue life of the prepared asphalt concrete mixes, a four point bending fatigue test was conducted according to *LVS EN 12697-24 Bituminous Mixtures – Test Methods for Hot Mix Asphalt – Part 24: Resistance to Fatigue*. The test was run at 10 °C and frequency of 10 Hz. The beams were compacted in the laboratory by using roller compactor and saw cut to the required dimensions of 50 mm wide, 50 mm high and 400 mm long. The failure criterion used in the study is the traditionally applied 50% reduction from initial stiffness. Fig. 9 presents fatigue lines of different aggregates and adhesion additives. Further research will include determination of fatigue life of the other bitumen types.

5. Conclusions

Moisture susceptibility and performance of asphalt concrete with and without adhesion additives on amino amide and pholiphosphoric acid based liquids were investigated. Granite asphalt mixtures without adhesion additives showed poor water resistance with any of the three bitumen types included in the study. Results demonstrated that introduction of adhesion additive on amino amide or pholiphosphoric acid base can significantly improve the resistance to water damage of granite asphalt mixtures. The asphalt mixture produced using diabase aggregate and pholiphosphoric acid based adhesion liquid had the lowest wheel tacking slope compared to other aggregates types and the use of polyaminoamide based adhesion additive. Asphalt mixtures produced using granite aggregate showed a little higher wheel tracking slope and rapid rut resistance improvement compared to control mix when either of the adhesion additives was used. Amino amide based adhesion additive did not significantly affect the rut resistance properties of mixtures with dolomite aggregate. The stripping test results with the same mineral aggregate but bitumen from different refineries showed various results, confirming that adhesion properties strongly depend on the binder origin. The granite asphalt mixtures without any adhesion additive showed very low fatigue resistance what is probably a result of initiation of fatigue micro cracks in the aggregate-bitumen interface. The addition of adhesion additive caused rapid increase in fatigue resistance.

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