New findings in relation to adhesion

Martin Radenberg[1, a], Nina Nytus[1], Ralf Diedel[2], Miriam Miehling[2], Sabine Boetcher[1]

[1] *Lehrstuhl für Verkehrswegebau, Ruhr-Universität Bochum, Bochum, Germany*
[2] *FGK Forschungsinstitut für Anorganische Werkstoffe - Glas/Keramik - GmbH, Höhr-Grenzhausen, Germany*

[a] verkehrswegebau@rub.de

Digital Object Identifier (DOI): dx.doi.org/10.14311/EE.2016.223

ABSTRACT

The adhesion of bitumen and aggregates is a very important factor regarding the durability of pavements. Loss of adhesion results in extensive renewals. Because of the countries' and communes' low budgets, these should be prevented by the right choice of bitumen and aggregate. This paper describes the findings of two projects with the goal of quantifying the main influencing factors on adhesion. Although adhesion is a topic for research since decades, there still are unsolved questions. The first project examined the component bitumen in detail. The physical and chemical properties of 90 different bitumen were analyzed and a selection of 15 bitumen, with significant differences in their properties, were used for adhesion tests with four different aggregates (basalt, rhyolithe, greywacke and diabase). The used test methods were the rolling-bottle-test, the Munich shake test, the determination of the ITSR and the contact angle. All test methods led to the fact that there are "good and bad" mixtures of bitumen and aggregate. The second project analyzes aggregates in detail. The chemical and mineralogical properties and especially the zeta-potential of eight different aggregates were determined and four aggregates with significant differences in their zeta-potential (limestone, greywacke, diabase and quartzite) were chosen for the following adhesion tests with five different bitumen: rolling-bottle-test, the Munich shake test, a kind of a water-boiling-test, determination of the contact angle and a new test method using the DSR. The influence of the zeta-potential (a parameter depending on the pH-value) on the adhesion was proved. The results of both studies helped to quantify the main influencing factors on adhesion and are supposed to lead to more durable pavements in the future.

Keywords: Adhesion, Aggregate, Chemical properties, Durability

This paper resp. this examination is based on parts of two accomplished research projects. First the IGF-project no. 16639 N / 1 with the title “Influence of bitumen properties on the adhesion behaviour of different aggregates”, which was coordinated by the German asphalt institute, Schieffelingsweg 6, 53123 Bonn and financially funded via AiF by the German Federal Ministry of Economics and Technology within the governmental R&D-support-measure “Industrial cooperative research” and secondly the research project with number FE 07.0261/2012/BRB and title “Further investigations to eva-luate the adhesion behaviour between aggregates and bitumen” on behalf of the Federal Ministry of Transport and digital Infrastructure represented by the Federal Highway Research Institute (BASt). The sole responsibility for the content lies with the author.

NEW FINDINGS IN RELATION TO ADHESION

1. INTRODUCTION

The street’s durability depends on the bonding between bitumen and aggregates. This bonding is a research topic since decades; several test methods were used to quantify the main influencing factors without closing success. Standards for bitumen’s chemical composition, which maybe indicate a better adhesion behaviour of the bitumen in combination with the used aggregates, were not clearly expressed so far, because of bitumen’s chemical diversity. The aim of the IGF-project no. 16639 N / 1 [1] with the title “Influence of bitumen properties on the adhesion behaviour of different aggregates” was to quantify the influence of physical and chemical bitumen properties in detail. For that purpose, 90 paving grade bitumen, which were produced in eight different refineries throughout Germany, were analysed physically and chemically. Then 15 bitumen samples with different properties were selected for the adhesion tests with three different types of aggregates. Therefore, different adhesion test methods were assessed.
The results showed that aggregates have a larger influence on the adhesion as bitumen [1]. Therefore, the focus of the research project “Further investigations to evaluate the adhesion behaviour between aggregates and bitumen” was on aggregate’s properties [2].
The research of LABIB [3] shows, that the zeta-potential measurement of aggregates and bitumen can be used to describe the interaction between both asphalt components. According to the electrostatic model, an opposite charge (high potential difference), e.g. the positive loaded charged limestone, has a positive influence on the bonding with a negative loaded charged bitumen emulsion. For the interpretation of the zeta-potential it’s important to express the results as zeta-potential-pH curves because the aggregate surface’s and bitumen emulsions’ charges can be highly changed by the pH value.
Generally it was established, that a better bonding could be achieved between bituminous binders and basic aggregates (e.g. calcium carbonate) than between bituminous binders and acid aggregates (e.g. quartz) [4].
Aggregates with a huge spectrum of their properties (chemistry, mineralogy, zeta-potential and particle surface charge) were chosen to achieve a good basic for the interaction analysis. These aggregates were also typically used for road construction in Germany. The bitumen selection was made, so that the chemical composition also has a huge spectrum and a different adhesion behaviour could be expected. In order to eliminate the viscosity influence only bitumen of the type 50/70 were used.

2. TESTING PROGRAM

2.1 Project “Influence of bitumen properties on the adhesion behaviour of different aggregates”

In order to achieve profound findings, the first part of the research program contained a thorough characterization of the compounds bitumen and aggregates. 90 paving grade bitumen, which were produced in eight different refineries throughout Germany, and seven different kinds of aggregates were analysed physically and chemically. The physical characterisation consists of the classical bitumen tests and the rheological analysis developed in the last decades. The classical bitumen tests needle penetration, softening point ring and ball, fraass breaking point, ductility as well as the rheological bitumen tests using the bending beam rheometer (BBR) and the dynamic shear rheometer (DSR) were realised in accordance with the relevant European (EN)-standards. Regarding the rheological analysis, it has to be mentioned that all cold-temperature-properties detected with the BBR (creep-stiffness S and the m-value) were measured at -16 °C after a load of 60 seconds in order to compare the different bitumen. Using the DSR, the dynamic viscosity μ was detected with a rotating cone-plate-geometry. Moreover, using a DSR with an oscillating plate-plate-geometry, the parameters complex shear modulus G^* and phase angle δ were measured between -20 and +150 °C at a frequency of 10 rad/s. All physical bitumen tests mentioned above are conducted with fresh bitumen and after a short term aging with the Rolling Thin Film Oven Test (RTFOT).
The chemical compositions of the different bitumen were defined with four tests: SARA fractionation, composition of the asphaltenes, simulated distillation and determination of the paraffin-wax-content. Conducting the SARA fractionation, the percentage of saturates, aromatics, resins and asphaltenes was determined by means of a column chromatography. Furthermore, the asphaltenes were divided into low, medium and high volatile compounds, using the chemical analysis procedure developed by ZENKE [5]. By conducting another non-standardised procedure, the simulated distillation developed by THIMM [6], the bitumen is divided into groups according to the sizes of the hydrocarbon molecules. In doing so, the distillation temperatures of the bitumen at a gas-chromatography were

dedicated to the hydrocarbon-molecules-sizes. As the only standardised bitumen test, the paraffin-wax-content was measured in accordance with EN 12606-1 [7].

Then 15 bitumen samples with different properties were selected for the adhesion tests. Their results of the SARA-fractionation is shown in table 1. Conducting the adhesion tests, the interaction between the bitumen samples and three selected most different aggregates were analysed. The results of the following adhesion tests are depicted in this paper.

Table 1: Results of the SARA-fractionation

Bitumen sample	**Paraffin-Wax-Content**	**Saturates**	**Mono-**	**Di-**	**Poly-**	**Sum of**	**Resins**	**Asphaltenes**
			aromatics					
	[% by mass]							
1.2 20/30	0,4	6,8	9,4	9,0	15,8	34,2	37,0	22,0
2.1 20/30	0,5	6,5	10,0	11,5	16,6	38,1	30,7	24,7
2.2 20/30	1,9	4,7	10,8	6,2	9,4	26,3	44,8	24,1
2.R1 30/45	1,0	6,5	8,4	12,5	20,9	41,8	26,4	25,3
6.R1 30/45	0,8	11,5	11,8	11,7	19,3	42,8	21,6	24,1
1.1 50/70	0,5	16,6	12,9	11,6	17,3	41,8	22,6	19,0
1.2 50/70	0,8	6,8	12,0	7,7	11,0	30,6	43,5	19,1
2.1 50/70	0,5	6,2	12,0	12,0	20,7	44,7	30,7	18,4
2.2 50/70	0,5	6,9	12,3	12,1	14,1	38,6	32,5	22,1
2.1 70/100	1,4	7,2	13,1	12,9	16,3	42,2	30,3	20,2
5.1 70/100	1,1	10,4	12,5	13,2	19,9	45,5	25,7	18,4
7.1 70/100	1,6	7,8	13,1	12,8	15,3	41,2	38,3	12,8
1.2 160/220	0,8	14,6	14,9	13,1	15,0	43,0	28,2	14,2
2.1 160/220	1,1	9,0	14,4	13,0	17,4	44,8	29,9	16,4
2.2 160/220	2,2	6,8	7,9	5,9	8,5	22,3	55,3	15,6

Rolling-bottle-test:

The rolling-bottle test was undertaken according to EN 12697-11A [8] with grain fraction 8/11 mm and after 6, 24 and 48 hours the grade of residual coating was determined. The evaluation was supported by a photo optical analysis. For that purpose, photo optical pictures of the aggregates with bitumen coating were analysed with "Image Processing Toolbox" of MATLAB®. With this tool "Cluster-Pictures" were produced and different colour groups were assigned to aggregates, bitumen and background and the parts of these colour groups were determined with this program. Than the residual coating can be calculated as follows:

$$\text{Residual coating [\%]} = \frac{\text{Colour group of aggregates [Pixel]}}{\text{Colour group of residual bitumen [Pixel]}} \qquad \text{(equation 1)}$$

Abrasion test:

The testing device according to EN 12274-7 [9] was used for the abrasion tests. Alternative to EN 12274-7 [9] the test specimens were made with the aggregate fraction 0.5/2 mm of one aggregate type. Because of this the influence of filler and dust was eliminated. The bitumen content was selected during preliminary tests with diabase, so that an air void content of 33 ± 1 % by volume was achieved. For the other aggregate types the bitumen content was adapted with the factor α ($\alpha = \frac{2.650}{\rho_P}$). These adapted bitumen contents are summarized in table 2.

The used specimens were conditioned three days at a temperature of 25 °C with and without water. Then they were exposed to a rotation of 20 r/min for three hours in a drive shaft filled with water at 25 °C. The loss of weight was used as factor of the adhesion.

Table 2: Bitumen content for abrasion test

Aggregate	Bitumen content [% by mass]
Diabase	4,8
Greywacke	5,1
Basalt	4,6
Rhyolite	5,2

Indirect tensile strength (ITSR)

Compacted asphalt specimens according to EN 12697-30 [10] were used for the indirect tensile test. The mixture consisted of aggregates with a maximum size of 8 mm and the target void content calculated with bulk density method d according to EN 12697-6 [11] was 19 % by volume. The bitumen content was adapted for the two chosen types of aggregates, rhyolite and basalt, with the factor α and amounts 7.5 % by mass with rhyolite and 6.7 % by mass with basalt. The aggregate's composition is shown in table 3.

The specimens, analysed with the indirect tensile test, were also conditioned with and without water. The indirect tensile test according to EN 12697-23 [12] was done at a test temperature of 5 °C with a constant rate of deformation of 50 mm/min.

Table 3: Mixture composition for ITSR

	Mixture with Rhyolith	Mixture with Basalt
Percentage by mass passing 11.2 mm sieve	100,0	100,0
Percentage by mass passing 8.0 mm sieve	93,0	94,9
Percentage by mass passing 5.6 mm sieve	26,7	27,7
Percentage by mass passing 2.0 mm sieve	10,6	11,8
Percentage by mass passing 0.125 mm sieve	1,8	1,5
Percentage by mass passing 0.0063 mm sieve	1,0	1,0
Bitumen content by mass	7,5	6,7

2.2 Project "Further investigations to evaluate the adhesion behaviour between aggregates and bitumen"

For this project five bitumen were used. The bitumen selection was based on the different results of the bitumen's chemical composition (SARA fractionation, composition of the asphaltenes) originated from the project mentioned above. Four different bitumen were chosen, which have a huge spectrum of their chemical composition and additionally a bitumen was used, which was produced with nearly 100 % crude oil from Venezuela.

In the investigations of the influencing factors on the aggregates' side four different aggregates should be chosen which have a huge properties' spectrum and are suspected to have an essential influence on the bonding between bitumen and aggregates. Therefore, the following properties were in focus: SiO_2- and quartz content, zeta-potential. At first the following seven aggregate types were characterized in detail: basalt, diabase, granite, greywacke, limestone, rhyolite and quartzite.

For the aggregate's characterization the following tests were done respectively the following specific values were determined: particle size distribution, specific gravity, specific surface BET, surface energy with the contact angle.

The chemical-mineralogical composition was analysed with X-ray fluorescence analysis, X-ray diffraction for a qualitative mineral analysis and a semi-quantitative quartz determination and determination of soluble salts with the leachability method, zeta-potential and specific surface charge.

The zeta-potential measurement was done according to FGK-AV Zeta-Potential [13] with the measurement device DT1200 of the company Dispersion Technology (DT) (electro acoustic method). The samples, used for this measurement, should have a size << 100 µm, so that this method can only be used for ground samples. Due to pulsed ultrasonic waves produced by the measuring sensor the particles in suspension are made to vibrate. This induces displacements of the electrical double layer surrounding each charged particle. Thus, temporary dipole moments are created which add to a measureable alternating current - the colloid vibration current (CVI). Based on the measured CVI the zeta-potential can be calculated. In addition to the basis measurement of the zeta-potential, the zeta-potential was also measured as a function of the pH-value. Therefore, suspension titration with HCl and NaOH was used.

Furthermore the aggregate samples were examined regarding their specific surface charge using the Particle Charge Detector. In contrast to the zeta-potential method both ground and originally coarse samples can be characterized by PCD applying a special back titration technique. Information regarding the charge properties on fine grained as well as coarse aggregates can thus be gained.

For the following adhesion tests, the aggregates limestone, diabase, quartzite and greywacke were chosen. These four aggregates covering the whole spectrum of quartz content (table 4) and zeta-potential depending on pH-value (figure 1) as well as the other aggregate properties (e.g. specific surface charge). The results of the RFA are summarized in table 5.

Table 4: Quartz content of the chosen aggregates

Aggregate's type	Basalt	Diabase	Granite	Greywacke	Limestone	Rhyolite	Quartzite
Quartz content [% by mass]	Below limit of detection	5,5 - 7,5	24 - 35	46 - 52	≤ 1,25	17 - 21	85,5 - 88,3

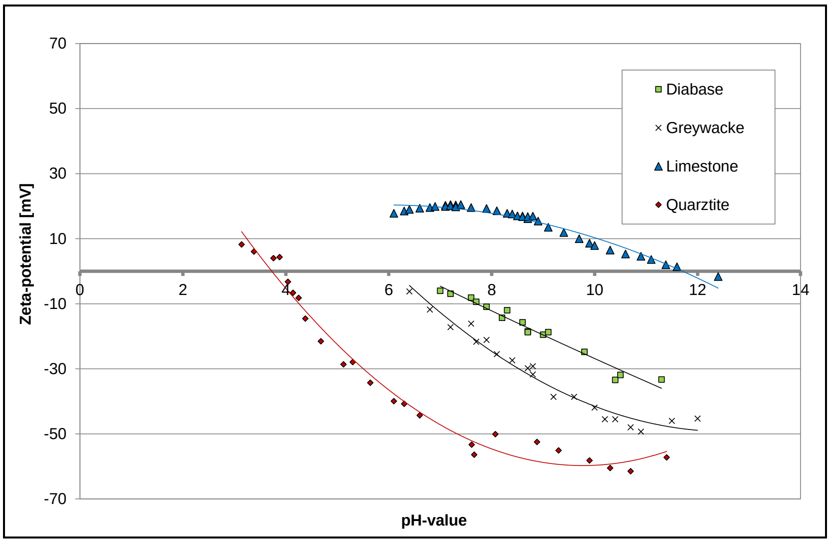

Figure 1: Zeta-potential of the aggregates depending on pH-value

Table 5: RFA results

Elementary oxides		Diabase	Greywacke	Limestone	Quartzite
SiO_2	[% by mass]	50,5	78,1	1,7	92,8
Al_2O_3		14,8	10,0	0,5	3,8
TiO_2		1,8	0,8	< 0,01	0,3
Fe_2O_3		12,3	4,1	0,3	1,6
CaO		6,9	1,8	95,1	0,1
K_2O		0,5	2,4	0,1	1,1
MgO		9,0	1,5	2,2	0,2
MnO		0,2	0,1	0,1	< 0,01
Na_2O		3,4	0,9	< 0,01	< 0,01
Cr_2O_3		0,1	0,0	< 0,01	0,0
P_2O_5		0,2	0,1	0,0	0,0
SO_3		0,2	0,1	0,1	< 0,01
SrO		0,0	< 0,01	0,1	< 0,01
V_2O_5		0,0	< 0,01	< 0,01	< 0,01
ZnO		0,0	< 0,01	< 0,01	< 0,01
ZrO_2		0,0	0,1	< 0,01	0,0
Total:		100,0	100,0	100,0	100,0
Ignition Loss		5,5	3,4	42,8	0,7

The boundary conditions of the adhesion tests, whose results are shown in this paper, are described in the following part.

Rolling-bottle-tests:
The rolling-bottle-tests were conducted according to DIN EN 12697-11 [8]. For the tests, the grain fraction 5/8 mm was chosen, because with this fraction results with a higher accuracy of the PCD-measurements were expected. The grade of residual coating was determined after 6, 24 and 48 h. In addition to the visual estimation, the evaluation was supported by a photo optical analysis.

Abrasion tests:
For this test the same conditions were used as described in 2.1. The adapted bitumen contents for quartzite and limestone are summarized in
Table 6.

Table 6: Bitumen content for abrasion test

Aggregate	Bitumen content [% by mass]
Quartzite	5,1
Limestone	5,1

Drop-shape analysis
For the quantification of the interaction between bitumen and aggregate the contact angle measurement was used with a high temperature dosing system and a high temperature chamber. The used measurement device was a DSA 100 of the company Krüss. The spreading behaviour was determined with the "sessile-drop" method over 120 seconds. The adhesion work was calculated with the results according to the modified equation of Young-Dupré [14].
Furthermore, the influence of water on the contact angle was tested with the method of KORN [15]. Therefore, 140 °C hot bitumen were dropped on the aggregate surfaces at room temperature. Based on the high temperature difference high contact angles occurred. To form a bonding, the samples were kept for 10 minutes in an oven at 30 °C over softening point ring and ball of the respective bitumen. After 5 minutes cooling, the samples were put for 120 minutes in water, which has a temperature 5 °C under the softening point ring and ball of the respective bitumen. With the difference of the contact angle at the beginning and at the end of water immersion conclusions about the bonding behaviour could be drawn.

3. TEST RESULTS AND DISSCUSION

3.1 Project "Influence of bitumen properties on the adhesion behaviour of different aggregates"

The results of the rolling-bottle-tests are shown in figure 2 to 5. The highest decrease in the grade of residual coating was determined with the aggregates rhyolite, diabase and basalt between 6 and 48 h. With the aggregate greywacke the greatest changes were noticed after 48 h for the half of the bitumen-aggregate-combinations. With the rhyolite several agglomerates occurred, so that these results cannot be used. With the other types of aggregates agglomerates occurred in combination with the softer bitumen 70/100 and 160/220.

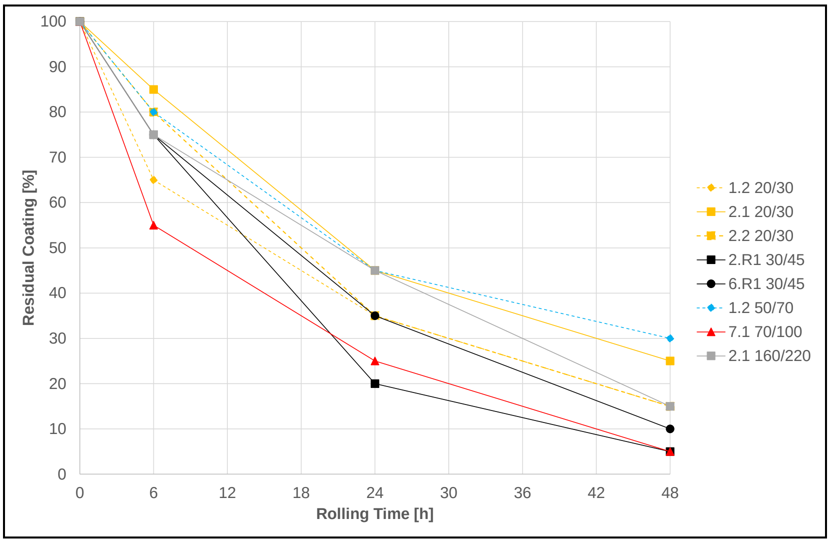

Figure 2: Results of the rolling-bottle-tests with rhyolite

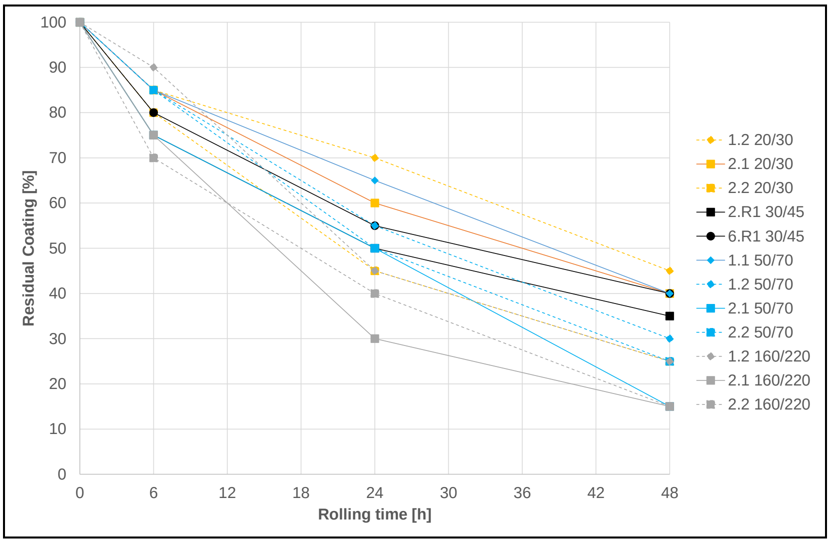

Figure 3: Results of the rolling-bottle-tests with diabase

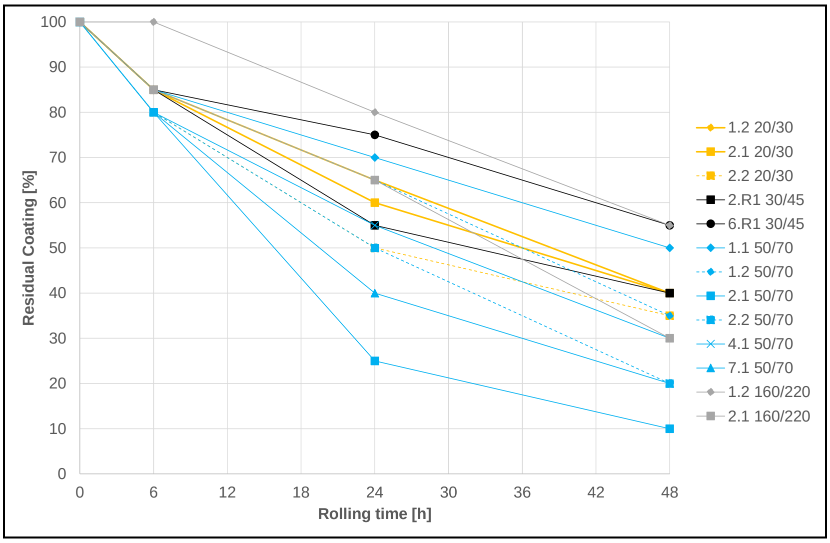

Figure 4: Results of the rolling-bottle-tests with greywacke

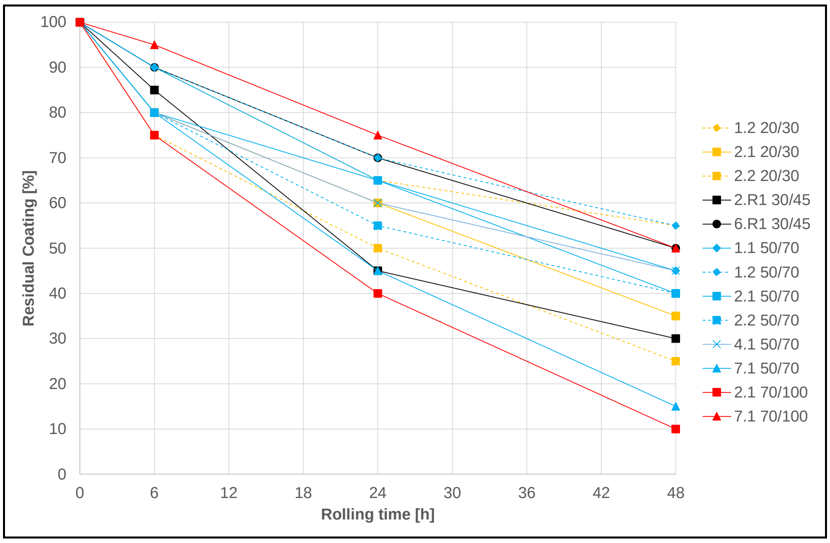

Figure 5: Results of the rolling-bottle-tests with basalt

Useful correlation between the grade of residual coating and the physical and chemical bitumen properties could not be identified.

Figure 6 shows comparatively the results of the abrasion tests with all chosen bitumen and aggregates. The sample mixtures deviated highly from the specification of EN 12274-7 [8] and the samples had huge void contents, therefore, a higher spreading between single values of one variant could be observed than allowed according to EN 12274-7 [8]. A maximal standard deviation of 4 % was defined.
The abrasion test results depends on bitumen hardness; with softer bitumen a lower abrasion was detected and with harder bitumen a higher abrasion. Possibly, the arising mechanic forces were attenuated with softer bitumen.

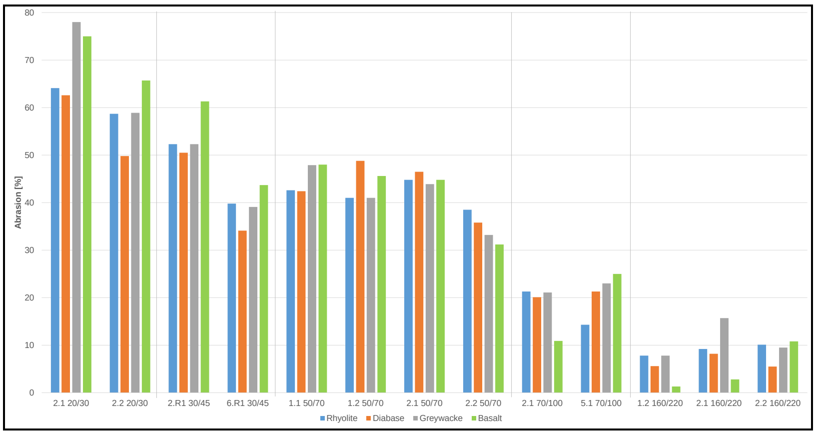

Figure 6: Comparative illustration of the abrasion test's results

The influence of bitumen hardness shows also the correlation between abrasion and physical properties like softening point ring and ball (figure 7). Regarding the chemical properties, a correlation by trend can be seen between abrasion and percentage of asphaltenes (figure 8). This validate again the influence of bitumen hardness and abrasion because the percentage of asphaltenes have an effect on the bitumen hardness. A clear correlation between the other chemical properties, determined in this project, could be shown.

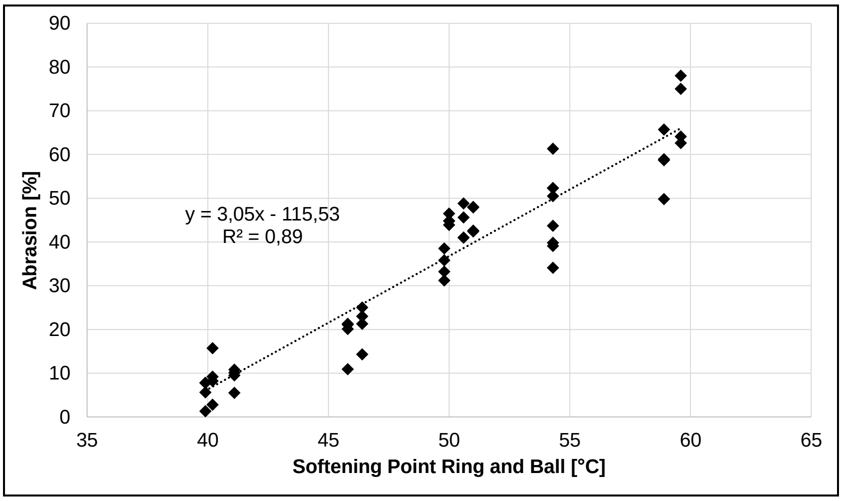

Figure 7: Correlation between abrasion and softening point ring and ball

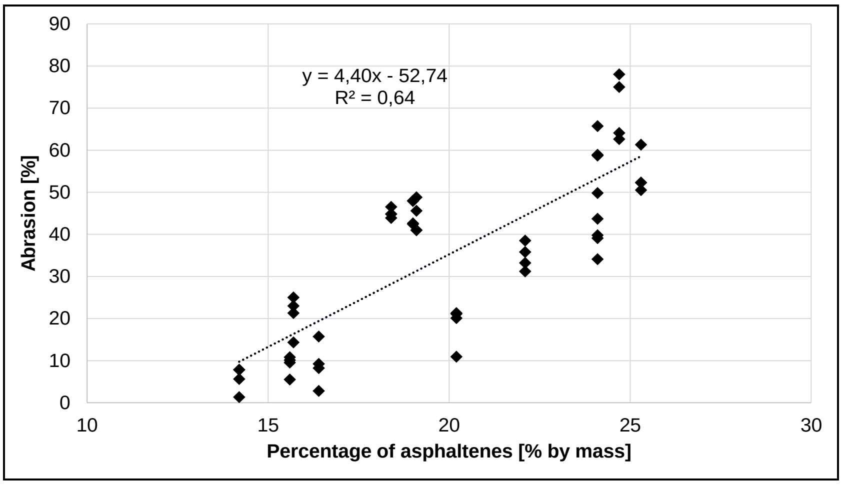

Figure 8: Correlation between percentage of asphaltenes and abrasion

The results of ITSR-value, which were determined in this study, have shown, that this value is unsuitable to describe the adhesion behaviour between aggregates and bitumen [1].

3.2 Project "Further investigations to evaluate the adhesion behaviour between aggregates and bitumen"

The rolling-bottle-test results after 24 h and 48 h are shown in figure 9 and 10. After a rolling time of 6 h a differentiation is not possible. Lowest grades of residual coating were ascertained with quartzite. The results of the other aggregates lie on a comparable level.

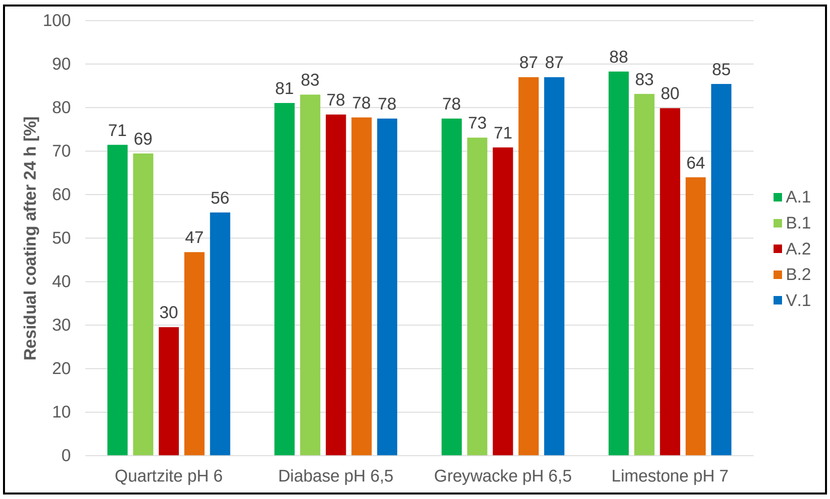

Figure 9: Grade of residual coating after 24 h

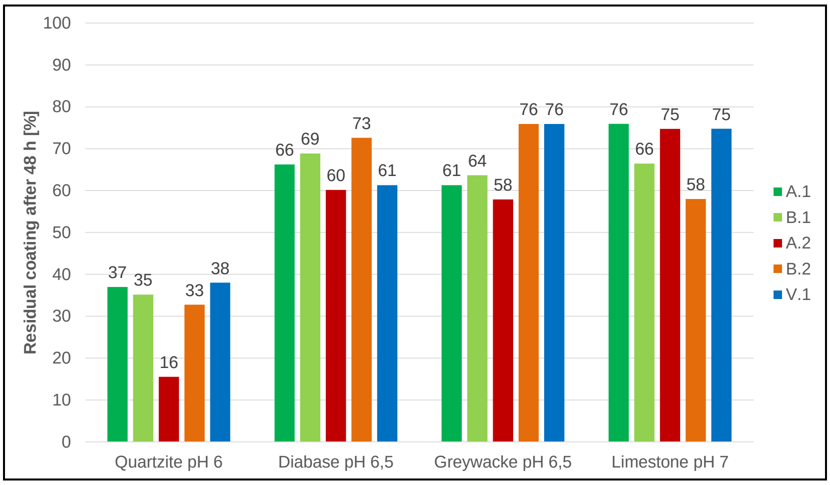

Figure 10: Grade of residual coating after 48 h

With the rolling-bottle-test a significant influence of the aggregates - particularly of the zeta-potential and the soluble salts' calcium carbonate content – on the covering could be determined. A more detailed quantification is possible after a rolling time of 48 h. With the main influencing factors the grade of residual coating after 48 h rolling time can be determined as follows with coefficient of determination of 0.83 (figure 11):

$$U_{48\,h} = 149.11 - 1.63 \times Ca + 0.08 \times Q + 0.53 \times Z \qquad \text{(equation 2)}$$

with U = Grade of residual coating after 48 h rolling time [%],
Ca = Aggregate's Ca- percentage of soluble salts [% by mass],
Q = Aggregate's content of quartz [% by mass]
Z = Aggregate's zeta-potential [mV]

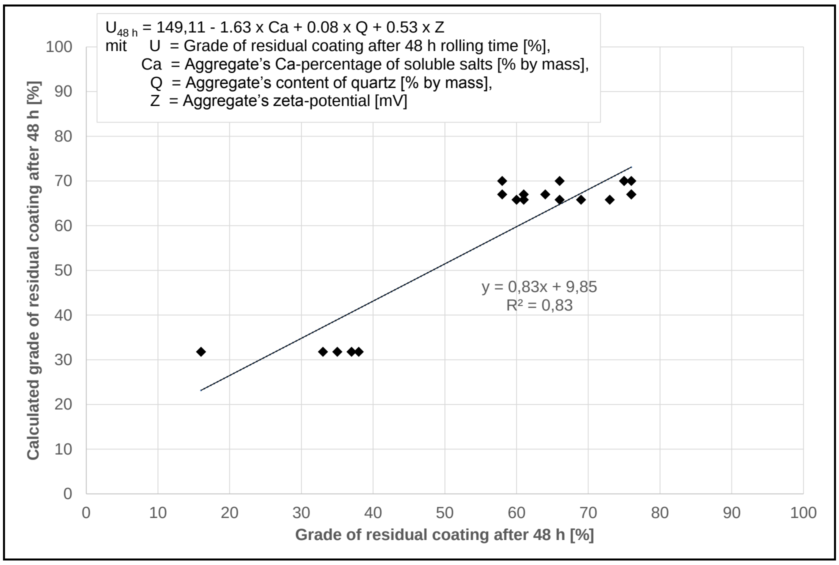

Figure 11: Comparison of grade of residual coating calculated and determined in laboratory after 48 h

The abrasion tests' results can be seen in figure 12. The abrasion test with the used boundary conditions proved to be inappropriate to quantify the possible influencing factors.

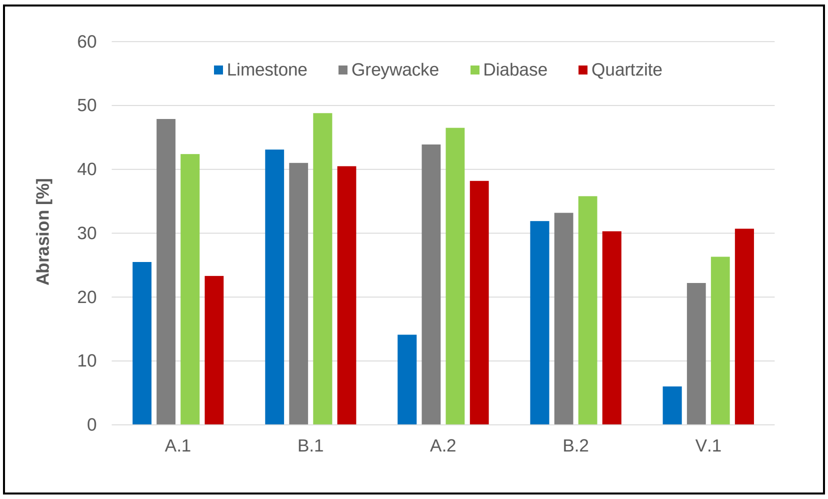

Figure 12: Abrasion test results

Also no influencing bitumen and aggregate properties affecting the contact angle could be found. However, by the contact angle measurement fundamental conclusions about the bonding between bitumen and aggregate can be detected. Al-though the water effect on the bonding can be measured. The results however, were influenced by the roughness and microstructure of the aggregate plates, therefore this should be considered.

Figure 13 shows the used bitumen's contact angles on the chosen aggregates. The contact angles vary from 26.3 ° to 42.3°; the lowest contact angle was measured with diabase and the highest with greywacke. Regardless of bitumen the highest contact angles shaped on greywacke surfaces, which suggest a comparable worse adhesion behaviour. With bitumen V.1, which differs from the other bitumen, the contact angle on greywacke was not significant higher than with the other aggregates. The lowest contact angle was measured with bitumen B.1, A.2 and B.2 on diabase, with bitumen V.1 and A.1 the differences on the different aggregate surfaces are not distinct in that way.
The calculated adhesion work can be seen in figure 14. Values between 55.3 mN/m and 60.9 mN/m were calculated. For all bitumen except V.1 a dependence of aggregate surface was detected. The lowest adhesion work for bitumen A.1, B.1, A.2 and B.2 was measured on greywacke and the highest with bitumen B.1 on diabase. In figure 15 the difference contact angles after 2 h water immersion were diagrammed. On the surfaces of quartzite and greywacke comparable high difference contact angles shaped. These depend on the used bitumen. Bitumen A.2 has the highest value for both aggregates. Difference angles between 10° and 20° stand according to KORN [15] for a bad adhesion behaviour. All difference angles on greywacke surface lie in this range. Good adhesion behaviour (1° to 10°) can be achieved by all bitumen with diabase and limestone. The values of bitumen A.1, B.1 and V.1 on quartzite indicate a "moderate" adhesion behaviour (3° to 10°).

Low adhesion work and high difference angles were determined for greywacke, which suggests a bad adhesion behaviour, but this couldn't be confirmed with the rolling-bottle-tests.

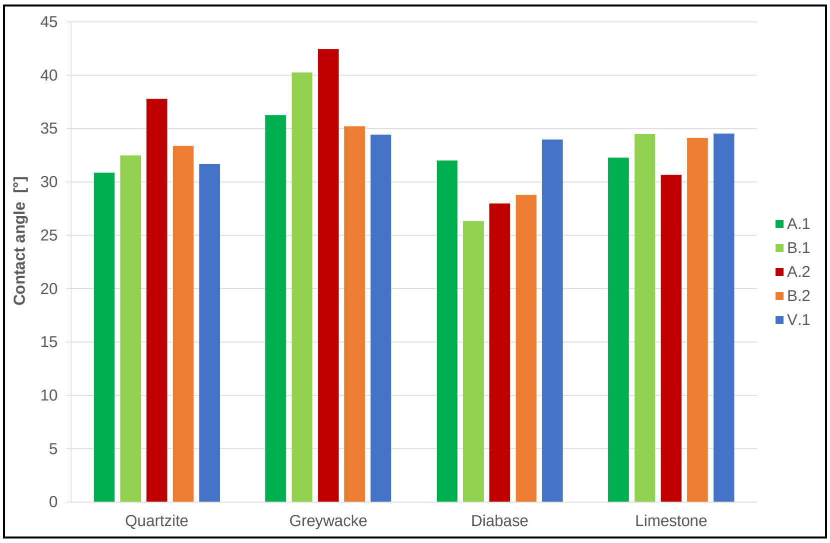

Figure 13: Bitumen's contact angles on the different aggregate surfaces

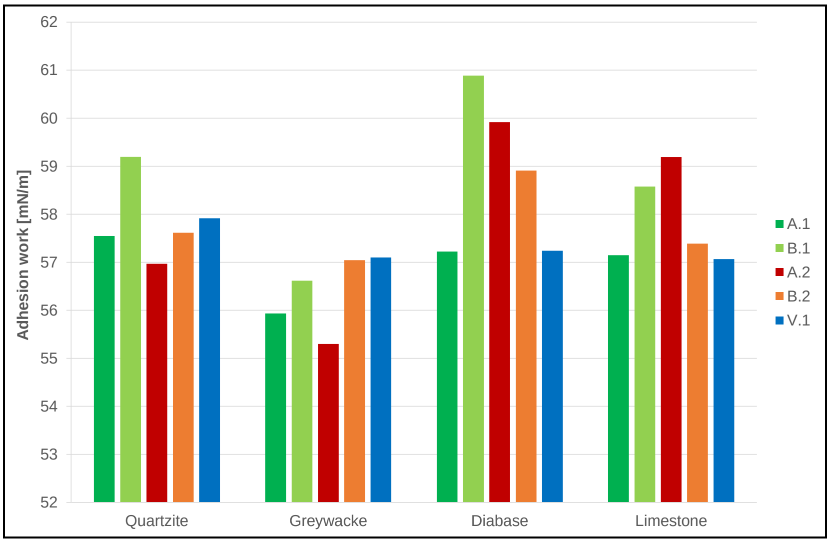

Figure 14: Bitumen's adhesion work on the different aggregate surfaces

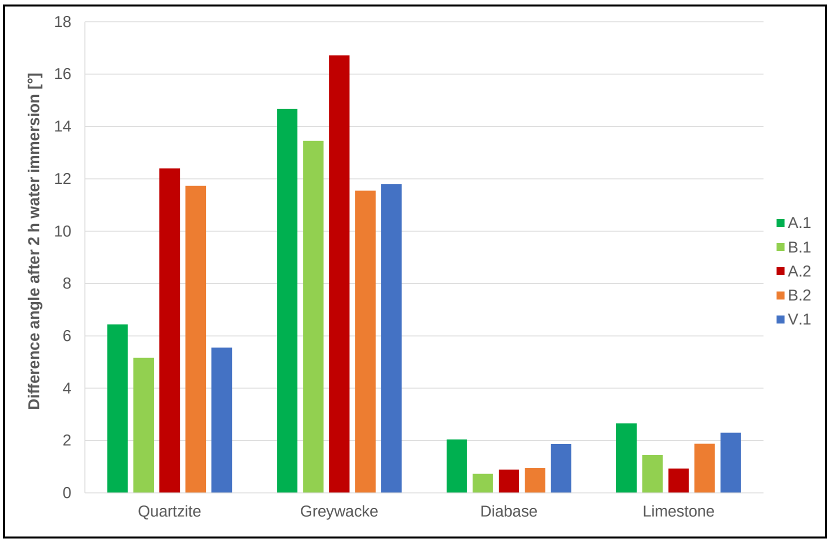

Figure 15: Difference angles after 2 h water immersion

4. CONCLUSIONS

The following conclusions can be drawn from the two research projects reported in this paper:

- A systematic influence of physical and chemical bitumen properties on adhesion behaviour could not be determined. Different bitumen-asphalt-combinations however, have a huge influence on the adhesion behaviour.

- The abrasion test, with the used boundary conditions, is inappropriate to quantify the possible influencing factors. The bitumen hardness is the controlling factor.

- The other test methods show different adhesion behaviour of bitumen and aggregates, but a uniform ranking of the different bitumen-asphalt-combinations could not be detected. With the used test methods the main influencing factors can't be quantified.

- Results of rolling-bottle-tests show a significant influence of the aggregate properties - particularly of the zeta-potential and the soluble salts' calcium carbonate content – on the covering. A more precise quantification is given after a rolling time of 48 h.

- The drop-shape analyses don't show clear influence of aggregate or bitumen properties on the results. They are obviously influenced by micro texture and roughness of the polished stone surface. Exemplary determination of a difference contact angle (changes due to water influence) shows the highest degree of differentiation of contact angle measurements. Therefore, these approaches should be pursued in order to improve the validity of the test procedure.

5. PERSPECTIVES

The huge amount of data should be expanded with more bitumen and aggregate specific values, so that the influencing factors can be quantified with multi variance analysis. Furthermore, new test methods should be developed or the boundary conditions of existing test methods should be adapted, so that they describe the adhesion behaviour independently of bitumen hardness, viscosity and mechanical forces.
Up to now there is no conclusive explanation for the behaviour of greywacke which is classified as less critical concerning adhesion. "Greywacke" however, is a term, which can vary in a wide chemical / mineralogical range because of its ge-nesis. Among other things it could contain carbonates, which could be a reason for a better adhesion behaviour.
Further aggregate tests should concentrate on zeta-potential and charge measurements of more aggregates and first zeta-potential tests of pure rock forming minerals like quartz or dolomite in order to increase the validity of the test methods and to assess the suitability of the aggregates. This should be accompanied by more bitumen tests regarding the adhesion behaviour between bitumen and single minerals.
In order to quantify the influence of porosity, one aggregate (e.g. rhyolite) with different porosities should be examined.
One more object for quantifying the influence solely of the bitumen could be tests with synthetically surfaces having defined characteristics. An appropriate test for that would be drop-shape analysis.

REFERENCES

[1] Einfluss der chemischen, rheologischen und physikalischen Grundeigenschaften von Straßenbaubitumen auf das Adhäsionsverhalten unterschiedlicher Gesteinskörnungen, M. Radenberg, N. Nytus, M. Gehrke, IGF-Vorhaben 16639 N/1, 2014

[2] Weiterführende Untersuchungen zur Beurteilung des Adhäsionsverhalten zwischen Bitumen und Gestein, M. Radenberg, N. Nytus, S. Boetcher, R. Diedel, M. Miehling, Entwurf Schlussbericht 2015

[3] Asphalt-aggregate interactions and mechanism for water stripping, M.E. Labib, Prep. Pap. American Chemical Society, Div. Fuel Chemistry 37 (3), 1992

[4] Fundamentals of Stripping in Bituminous Pavements – State of the Art, Research Report Trita-VT AR 03:01, U. Bagampadde, B.M. Kiggundu, Stockholm, 2003

[5] Zum Löseverhalten von Asphaltenen: Anwendung von Löslichkeitsparameter-Konzepten auf Kolloidfraktionen schwerer Erdölprodukte, G. Zenke, doctoral thesis, faculty of mining, metallurgy and mechanical engineering, University of technology Clausthal, 1989

[6] Validierung eines gaschromatographischen Analyseverfahrens zur Qualitätssicherung des in Deutschland verwendeten Straßenbaubitumens, L. Thimm, diploma thesis, faculty of chemistry, university of Duisburg-Essen, 2003

[7] EN 12606-1: Bitumen and bituminous binders – Determination of the paraffin wax content – Part 1: Method by distillation; German version EN 12606-1:2007

[8] EN 12697-11: Bituminous mixtures - Test methods for hot mix asphalt - Part 11: Determination of the affinity between aggregate and bitumen; German version EN 12697-11:2012
[9] EN 12274-7: Slurry surfacing - Part 7: Shaking abrasion test; German version EN 12274-7:2005
[10] EN 12697-30: Bituminous mixtures - Test methods for hot mix asphalt - Part 30: Specimen preparation by impact compactor; German version EN 12697-30:2012
[11] EN 12697-6: Bituminous mixtures - Test methods for hot mix asphalt - Part 6: Determination of bulk density of bituminous specimens; German version EN 12697-6:2012
[12] EN 12697-23: Bituminous mixtures - Test methods - Part 23: Determination of the indirect tensile strength of bituminous specimens; German and English version prEN 12697-23:2015
[13] Bestimmung des Zeta-Potentials im Initialzustand der Suspension, FGK-Arbeitsvorschrift Basismessung, FGK Forschungsinstitut für Anorganische Werkstoffe - Glas/Keramik – GmbH, Höhr-Grenzhausen
[14] Interfacial Lifshitz-van der Waals and polar interactions in macroscopic systems, C. J. Van Oss, M. K. Chaudhury, J.R. Good, Chemical Review, 88, 927, 1988
[15] Beurteilung der Benetzbarkeit und des Adhäsionsvermögens von Bitumen an Mineralstoffen mittels Kontaktwinkelmessungen, S. Korn, Diplomarbeit. Hamburg. Hochschule für angewandte Wissenschaften, Fachbereich Bauingenieurwesen, 2004