THE SEARCH FOR SIMPLE TESTS TO ASSESS THE COMPLEX PROPERTIES OF ADHESION AND DURABILITY OF ADHESION

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ABSTRACT

"Adhesion" and "durability of adhesion" are considered essential performance characteristics of asphalt mixtures. As such they are mentioned in the answers to the Mandate M/124 of the CPD for bituminous binders and asphalt mixtures. To agree test methods to assess these properties two special CEN task groups were formed. The CEN Ad-Hoc group adopted the SATS test for "durability of adhesion". The "Industry group" searched for a simple stickiness indicator. This publication reveals the work performed by the "Industry group".

Six bituminous binders, four 50/70 normal paving grades and two specifically modified bitumens, were compared in 9 different test methods to address a stickiness indicator. These test methods were selected following a literature search on procedures deemed to address "adhesion".

The conclusions of this study are:
• There is no easy-to-use method to characterise the adhesion of bitumen

The recommendations of the Industry Group are:
• Test methods that measure a property relying only on a ratio of properties (e.g. before/after treatment) are very likely to be flawed
• Test methods done at a specific temperature should take into account that the ability of the binder to deform will influence the test result rather than the adhesion of the binder itself.
• A reference mineral aggregate material –such as silica sand- can be used in laboratory tests. However, there is no substitute for actual mineral aggregates when lab results are extrapolated to field performance.

Keywords: Comité Européen de Normalisation, Bonding, Best Available Technology, Adhesion

1 INTRODUCTION

This publication describes the work done by the Industry Group Adhesion, as part of the activity of the CEN Ad-Hoc Group Adhesion - Durability, to search for a simple and easy-to-use bitumen stickiness indicator.

2 BACKGROUND

To understand the scope of this work some definitions will be given to reflect the direction during the research. The start of the work is related to the CEN Technical Committee for Bituminous Binders (CEN TC 336) together with the Technical Committee for Road Materials (CEN TC 227).
The Technical Committees of CEN are responsible for harmonization of standards and specifications in Europe. Research and developments are not part of the CEN TC tasks and if required special task groups can be formed with members of a CEN Technical Committee and invited industry experts depending on the topic.

In the Mandate M/124 of TC336 dated 24 September 2001 adhesion was quoted as essential characteristics for bituminous binders. Formal TC336 response to the mandate was: "*The adhesive character of bitumen is heavily dependent on the nature of the aggregate employed in each case, and is therefore not possible to specify it in absolute terms*." In April 2003 two new groups were created involving members from CEN TC 227 WG1 Bituminous mixtures, CEN TC 336 WG1 Paving Grade Bitumens and CEN TC 154 Aggregates, the CEN Ad-Hoc Group Adhesion –Durability [1] and the Industry Group (see table 2).
The CEN Ad-hoc group Adhesion – Durability held its first meeting on 22nd October 2004. The Industry Group, looking at binder adhesion test methods, provided progress reports to the CEN Ad-hoc group.

This paper is related to work of the Industry Group.

Adhesion is the bond strength between two different materials (e.g. bitumen and aggregate or filler) depending on material characteristics on macro-, micro- and nano-scale. **Durability** can be defined as the resistance to the decline of any performance property over time. The decline could be, amongst other, a result of hardening, ageing or exposure to the elements. **Stripping** is defined as the loss of adhesion between binder and aggregate through the action of water.

The following table shows the identified gaps in the specifications framework.

Table 1. Work area on Adhesion and Durability and identified gaps in specification (grey text).

	Bitumen *This is the work area of the Ad-Hoc group Adhesion – Durability*	**Bituminous mixtures** *This is the work area of CEN TC 227 WG1*
Adhesion	Test for binder adhesion ***Industry Group***	Water sensitivity EN 12697-11 and -12
Durability of adhesion	Test for durability of adhesion ***Ad-Hoc Group***	Aging in an oven? Short term ageing using lose un-compacted asphalt? Long Term Ageing?
Cohesion	CEN TC336 WG4: Force Ductility (EN 13589) Tensile Test (EN 13587) Pendulum Test (EN 13588)	Cantabro Deformation Energy until fracture
Durability of Cohesion	Short or Long term ageing with Force Ductility (EN 13589) Tensile Test (EN 13587) Pendulum Test (EN 13588)	Test for durability of cohesion

The working groups contributed each on their relevant identified gaps. Members of the groups were members of the different TC's and invited experts.
The CEN Ad-Hoc Group concluded that the SATS test [2] filled the gap in table 1.

Table 2. Members of the two working groups

Ad-Hoc Group Adhesion - Durability	***Industry Group Adhesion***
Egbert Beuving, *convenor, TC227 / TC336*	Egbert Beuving, *EAPA*
Jeroen Besamusca, *KPC*	Jeroen Besamusca, *KPC*
Jean-Luc Delorme, *TC227*	Tony Harrison / Chris Southwell, *RBA*
Richard Elliot, *TC227*	Colin Loveday, *Tarmac*
Tony Harrison, *TC227 / TC336*	Jim Carswell, *BP* ✝
Jos van der Heide, *TC227*	Carl Robertus, *BP*
Colin Loveday, *TC154*	Jean-Paul Michaut, *Colas*
Alberto Madella, *TC336*	Chris Raynor, *ExxonMobil*
Cliff Nichols, *TC227*	Andy Self, *Shell*
Erik Nielsen, *TC336*	Mike Southern, *Eurobitume*
Chris Rayner, *TC336*	André Stawiarski / Sophie Mariotti, *TOTAL*
André Stawiarski, *TC336*	André Täube, *DAV*
Mike Southern, *TC336*	Wim Teugels, *Nynas*
André Täube, *DAV*	
Wim Teugels, *Nynas*	
Stefan Vansteenkiste, *BRRC*	
Chris Southwell, *TC336*	
John Williams, *TC227*	

3 PROGRAMME

The working procedure for the Industry Group was comparable to the procedure followed by the CEN Ad-Hoc Group Adhesion - Durability:
Literature review of potential test methods and research that is going on that leads to a suitable test method.
Pre-select test methods and evaluate
Recommend the most promising test method to CEN TC 336 and CEN TC 277.

The scope and definition for the Industry Group programme originates from a TC 227 internal discussion:

The background for the need to be able to characterise adhesion is the Initial Type testing of bituminous mixtures. A new (Initial) Type testing of the bituminous mixture is needed if one of the characteristics of the constituent materials changes. So a simple and quick method to characterise the adhesion of the bitumen that can be used alongside other properties to "indicate binder constancy" is desirable.
Therefore:
The adhesion test should characterise the stickiness of bitumen; it should provide relative performance (relative to a standard aggregate) and is not meant to be a performance test for a bituminous mixture.
It should be a tool to check the consistency of adhesion and should be used between the bitumen supplier and the asphalt producer only.

Any outcome was NOT intended to define a specification for adhesion nor would, due to the influence of aggregate, any absolute values or classes be established. This limited the scope of the Industry Group to existing test methods and excluded research programs on, for example, surface energy [3].

3.1 Test methods.

In view of the limited scope, the Industry Group formulated requirements for the evaluation of tests based on the literature review from CEN Ad-Hoc group.
Be simple (no complex equipment)
Differentiate between bitumens
Be reproducible

Be quick (< 7 days)
Samples tested with access to water
Test or samples should be capable to include ageing conditioning

The following tests were selected for the primary evaluation. A complete description is available in reference [4].

Table 3. Test methods selected for first evaluation

1	Vandskak test
2	Munich Shake test
3	Indirect tensile test
4	Duriez
5	Wet attrition in Deval test
6	Cantabro
7	Water stripping test, XPT 66-043
8	PATTI test
9	Visualisation test, MoD stripping test.

3.2 Material selection of Industry group.

A preliminary investigation with only three binders did not show any relation or answers. Therefore the binder set was extended to six, see Table 4, and all binders were to be included in all test methods.

Table 4. Binder materials selected

05-018	50/70 from supplier A
05-064	50/70 from supplier B
05-182	50/70 from supplier C
05-183	50/70 from supplier D
04-351	Polymer modified binder
05-160	50/70 binder designed to have low adhesive properties

The reference aggregate chosen consisted of silica sand 0/1 according to EN 1097-8. As some tests are impossible on sand only, e.g. MoD-stripping and the XPT 66-043, well known reference aggregates were included: Diorite, Gravel, Limestone, Swedish aggregate, Flint (Silex) and Quartzite. For the PATTI test glass was used in addition to the bitumens.
The absolute values of samples with silica sand were low compared with regular asphalt samples due to the high void content.

Samples were tested before and after water immersion. The total number of tests was approximately 150. Each member of the Industry Group contributed to the work by performing at least one test and some members contributed to multiple tests.

4 FIRST RESULTS OF THE INDUSTRY GROUP.

Table 5 provides a summary of the test results. Extracting useful comparative information from different tests is difficult as results are expressed differently. Some tests reveal a property change e.g. tensile strength before and after conditioning, some results are expressed as percentage (e.g. covered aggregate) while others are expressed as a weight loss.

T he results of the attrition tests (e.g. Vandskak, Munich shaking test, Deval and Cantabro) were expressed as percentage with low value best. These test results are indicated in the table by (100-%)with high value therefore representing good performance.

Test results shown in table 5, reveal that each bitumen supplier can claim that its 50/70 bitumen is at least the best of the penetration grade bitumens tested depending on the test method chosen. But results of the same bitumen in another test would show to be weaker or even have the weakest performance. How can that be? All distributed cans of one bitumen type were from the same batch so as to ensure “round robin” requirements of testing. Tests were therefore expected to provide similar ranking of all materials. The ranking should not be dependent on the test method!

Table 5. First results of tests with six different binders, ranked with good properties as high values.

			1	**2**	**3**	**4**	**5**	**6**	**7**	**8**
			Deval test	Cantabro	Vandskak		Munich test	Deval test	Cantabro	Tensile test
			wt loss dry	wt loss dry	wt loss		wt loss	wt loss wet	wt loss wet	before
	Pen	Soft	Silica	Silica	Silica	Swed agg	Silica	Silica	Silica	Silica
	[0.1 mm]	[°C]	[100-%]	[100-%]	[100-%]	[100-%]	[100-%]	[100-%]	[100-%]	[MPa]
04/351	48	62.4	98	86.4	82.0	94.0	88.5	88	88.3	
05/018	67	48.2	93	56.8	33.4	84.6	86.3	79	44.0	1.018
05/064	60	50.0	97	35.7	41.2	71.6	65.3	83	32.6	1.099
05/160	64	71.4	89	58.1	64.6	80.5	75.5	87	61.6	0.755
05/182	57	50.8	90	35.6	35.3	78.3	73.4	84	40.9	1.173
05/183	55	51.0	90	25.8	31.2	72.6	74.5	83	37.8	1.256
	9	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**
	Fracture	Duriez	Duriez	Tensile test	Fracture	Duriez	Duriez	Tensile test	Fracture	Duriez
	before	dry	dry	wet	wet	wet	wet	rel.	rel.	rel.
	Silica	Diorite	Silica	Silica	Silica	Diorite	Silica	Silica	Silica	Diorite
	[N/mm^2/mm]	[MPa]	[MPa]	[MPa]	[N/mm^2/mm]	[MPa]	[MPa]	[%]	[%]	[%]
04/351		11.4	2.3	0.52	2.7	9.4	3.2			82
05/018	10.7	9.4	1.8	0.32	1.6	8.1	2.1	32	15	86
05/064	8.5	11.1	1.7	0.33	1.0	8.9	2.1	30	12	80
05/160	5.6	7.4	1.3	0.43	1.6	6.4	1.8	57	29	86
05/182	7.4	11.5	1.5	0.38	1.2	9.1	2.0	32	16	79
05/183	5.2	11.4	2.2	0.40	1.0	9.7	2.8	31	19	85
	19	**20**	**21**	**22**	**23**	**24**	**25**	**26**	**27**	**28**
	Duriez	7 days @ 60°C		XP T 66 043		16h @ 60°C		Pull off	Pull off	Pull off
	rel.	Immersion	Immersion	Immersion	Immersion	Immersion	Immersion	0 h	24 h	rel.
	Silica	Gravel	Limestone	Diorite	Flint (Silex)	Quartsite	Limestone	Glass	Glass	Glass
	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[psi]	[psi]	[%]
04/351	139	90	97.5	75	75	75	50	230	95	41
05/018	117	38	100	50	35	5	50	290	90	31
05/064	124	88	97.5	90	50	35	25	260	60	23
05/160	138	95	100	90	75	90	75	180	50	28
05/182	133	63	100	75	50	35	35	255	88	35
05/183	127	88	100	90	50	50	25	360	75	21

One important conclusion regarding the apparent inconsistency in results of the four 50/70 bitumens was drawn during the meeting in April 2006. As each test was done by only one lab the inconsistency was mainly attributed to the poor repeatability of the test methods and not so much due to the different origins of the 50/70 bitumens. It was agreed to assume that the adhesion performance of all 50/70 are equal AND the PmB and special binder have different adhesion performance as measured in the test methods. Therefore the set of 50/70 bitumens can be regarded as a statistical group with for each test an average/mean result and a standard deviation. In this way the performance of the PmB and the special binder test results can be evaluated on basis of the (percentage) difference with the average of the four 50/70 bitumens. To ensure 95% confidence the standard deviation is multiplied by 1.6 [5].

For example:
Test 6, Deval with silica. The lowest weight loss is the best performance, and is recalculated to the highest value (X). In this way no weight loss would correspond to 100%. The mean value of the four 50/70 penetration grade bitumens, grey area, is 82. The standard deviation is 2.2 (multiplied by 1.6 = 3.5). In this example the ranking is clear. The PmB and the special binder are significantly better than the 50/70 bitumens. The deviation (Y) for the PmB and special binder is larger than the 95% (3.5) confidence of the 50/70 bitumens.
The percentage variance is relative to the mean value of the four 50/70 bitumens.

Table 6. Example of calculation “high value = best”, deviation from average, variance in percentage.

	Weight loss [m%]	X [100 – value]	Y [X - 82]	(Y / 82) * 100 [%]
04-351	12	88	6	7.3
05-018	21	79	- 3	- 3.7
05-064	17	83	1	1.2
05-160	13	87	5	6.1
05-182	17	83	1	1.2
05-183	16	84	2	2.4

In this way all results can be reported in reference to the average value of the four 50/70 bitumens for each specific test. The relative position of each binder compared with the average of the 50/70 group can now be shown in one graph. Results are shown in figure 1. Test results of the PmB on tensile test and fracture test in dry conditions were not available.

The zero line is of course the 50/70 group average. Some tests show a variance of almost 50%. The special binder (with intended low adhesion properties) is mostly better than then the 50/70 mean value. What have we measured?

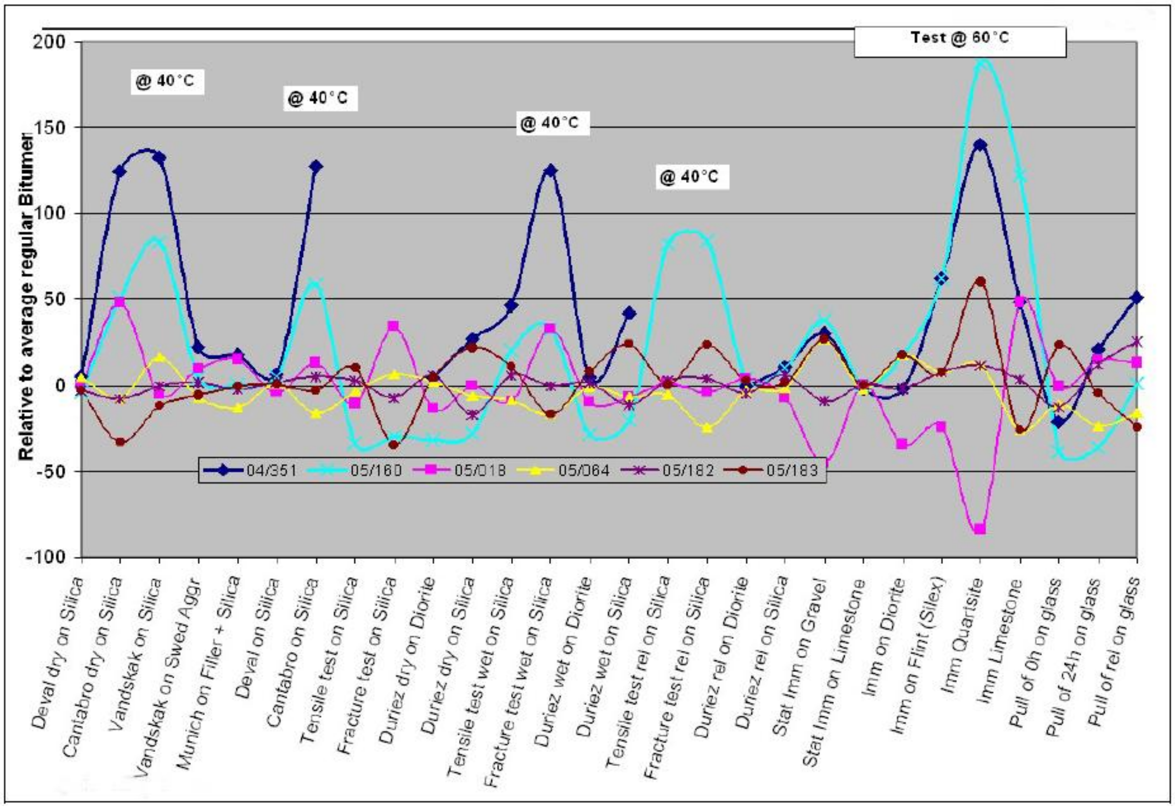

Figure 1. First results of tests, referenced to the average of the four commercial 50/70 binders.

4.1 Discussion of the first results of Industry Group.

The test results lead to two remarkable observations:

The first observation is related to the effect of "ratio" as a measurable property in a particular test. For example, the tensile test measures the strength of an asphalt material which is a function of both the adhesive and cohesive strength of the asphalt constituents. The ratio of tensile strength (after conditioning in water versus initial strength) is intended to be a measure of bitumen adhesion assuming water does not influence the cohesion of the asphalt constituents.

In the tensile test the initial tensile strength is low for the special binder, 05-160, when compared to the other strength values of the other binders. After conditioning only a small change occurs in this test for the special binder resulting in a high ratio of tensile strength before and after conditioning. The drop in tensile strength of the other binders due to conditioning is significantly larger. The absolute tensile strength after conditioning, however, remains larger for the other binders compared to the special binder. The ratio of tensile strength before and after conditioning of the 50/70 bitumens is much lower, rating the adhesion performance for these binders as poorer than that of the special binder.

There are several tests that only look at the ratio before and after conditioning and do not consider the absolute value. The Duriez test, EN 12697-12 part B, is required in the French asphalt design, the Indirect Tensile Strength Ratio, EN 12697-12 / EN 12697-23, is one of the functional requirements in the Netherlands. In both these tests only the value for the ratio are considered. Other examples for "ratio tests" are the Retained Marshall Stability, the Retained Stiffness and Retained Cantabro.

The second observation is related to the high test result values for the PmB and special binder in certain tests. It appears that these high values coincide with a high conditioning temperature and/or a high test temperature, see Figure 2.

The softening point of the penetration grade 50/70 bitumen was approximately 50°C, for the PmB the softening point R&B was ≈ 60°C and the special binder was as high as ≈ 70°C. From a binder stiffness or viscous flow perspective, a test or conditioning a test at high temperature (40°C or 60°C) will affect the penetration grade binder much more than the PmB or special binder. The test results probably reveals differences in flow or stiffness properties and not so much the binder adhesion characteristics.

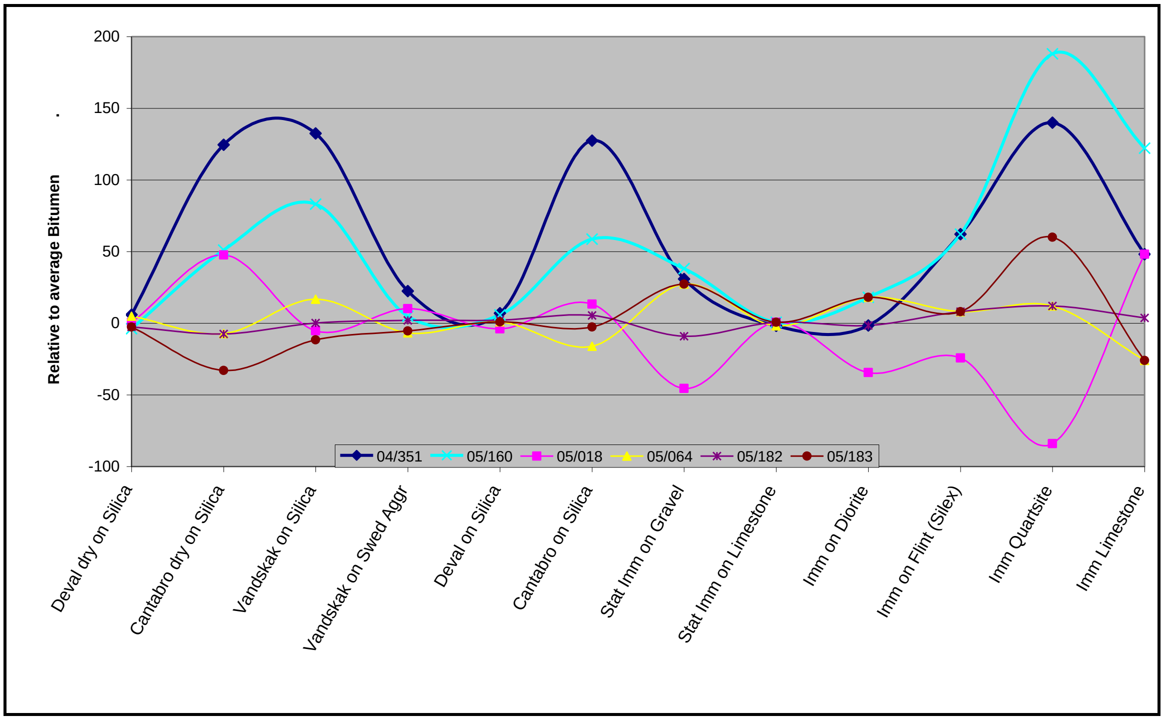

Figure 2. Results of tests with high conditioning and/or high testing temperature, referenced to the average of the four commercial 50/70 binders.

4.2 Follow up tests

Follow up tests were done in order to:

(1) Test the possible effect of the different bitumen stiffness/viscosities at the conditioning temperature in the water stripping test XPT 66-043
(2) Redo a number of tests (Deval/Vanskak/Munich/Duriez and Tensile Test) with silica sand only, i.e. without larger mineral aggregate fractions.
(3) Test the possible influence of binder stiffness at the test temperature by including a hard paving grade binder with a softening point approximately equal to the softening point of the PMB.

4.2.1 Water Stripping at a different temperature

The hypothesis was that the bitumen stiffness/viscosity at the conditioning- and test temperature in the water stripping test affects the test results rather than "adhesion". A high stiffness/viscosity would prevent a bitumen film to retreat from the mineral aggregate surface. To test this the Water stripping test XPT 66-043 was performed at 40°C, 50°C and 60°C for respectively the 50/70 bitumen, the PmB and the special binder (in fact at 10°C below softening point for all binders) with Chailloué Quartzite. When the test was performed at an effective equivalent viscosity temperature, the results are all within the 95% confidence of the mean value of the four 50/70 penetration grade bitumens. This is shown in figure 3, XPT test @ different Temp.

4.2.2 Test with silica sand only

The Deval test, Vandskak test and Munich Shake test were repeated with silica sand only and the Vandskak also repeated with Swedish aggregate. Unfortunately, the Deval and the Munich Shake test were too severe for testing with a one (small) sized aggregate and the sample preparation of the Vandskak was also too complicated with the one sized small aggregate.

Tensile Test and Duriez samples with silica sand only had an average void content of 30% and density of approximately 1640 kg/m^3 revealing the difference with regular asphalt constrcutions with approximately 5% voids content and density of > 2500 kg/m^3. In the initial study the variance of the four 50/70 bitumens in these tests was less than 25% which was smaller than for any of the other test methods.

The results of the 50/70 binders tested with Tensile and Duriez in 2005 and 2008 all fall within the 25% variance.. The EN 12697-23, standard for Indirect Tensile test, cites a repeatability of 17% (which is the standard deviation). Consequently the value of 25% found was regarded as a good reproducibility for the test series performed.

Comparing the results from 2005 with the follow up test results from 2008 shows similar ranking of the materials. The only discrepancy is the Duriez dry result with PmB binder (2005 above average and 2008 below average) but in both cases within the variance of 25%. The PmB clearly shows better performance in the tensile test after conditioning, but is that due to good/better adhesion?

4.2.3 Influence of binder stiffness at the test temperature

To try and answer the question of the influence of stiffness a 20/30 penetration grade bitumen was introduced in the 2008 follow up tests. This bitumen has a softening point of 57°C (comparable to the PmB). This bitumen significantly outperforms all other binders tested in the 2008 series. This is attributed to the fact that in these tests, performed at low temperature, the stiffness (penetration) of the bitumen dominates the test outcome. Alas, a high value in the Duriez and the Tensile Test is therefore more associated with the stiffness of the bitumen and not so much as to the adhesion or stripping properties of the bitumen.

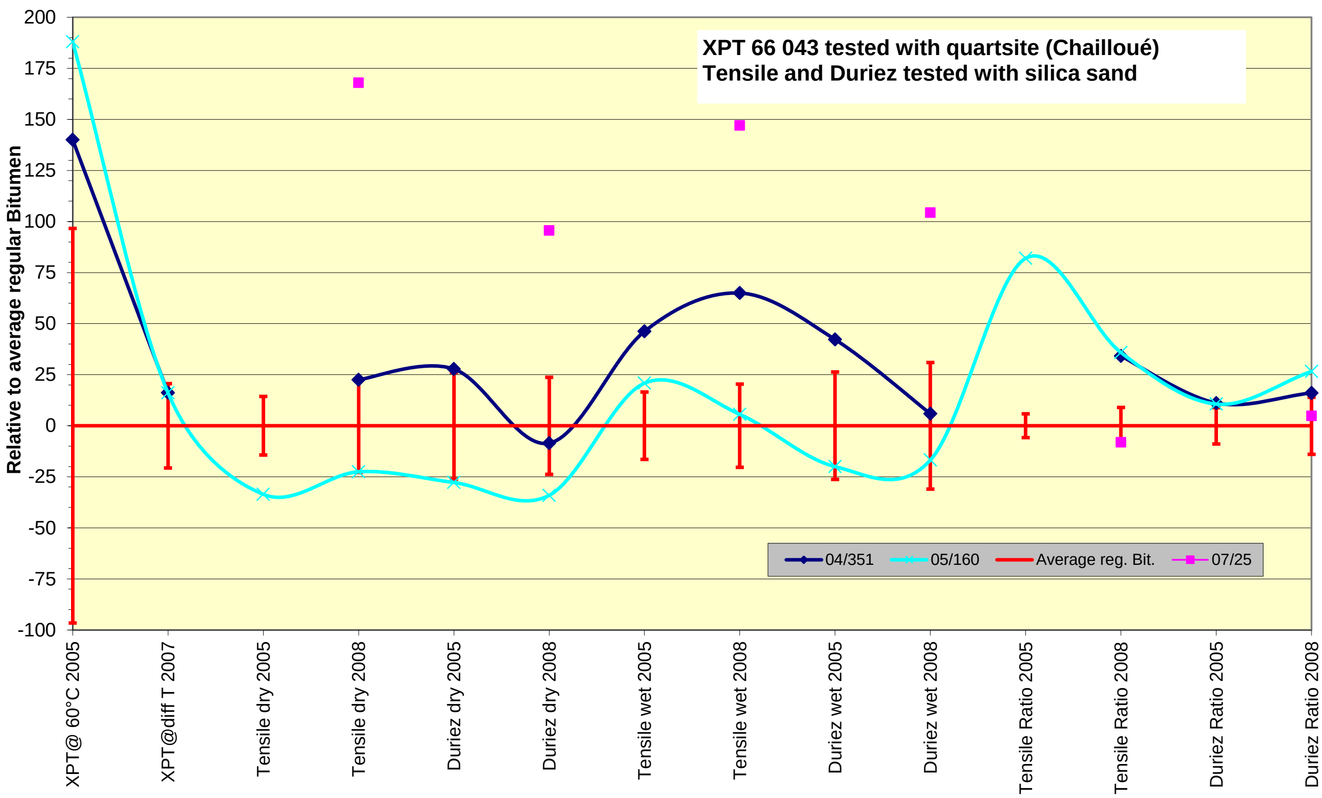

Figure 3. Results of follow up tests, referenced to the mean of the four commercial 50/70 pen grade bitumens.

Looking at ratio the PMB and special binder show, based on tensile test results, better performance than 50/70 and according to Duriez similar or slightly better performance. The performance of 20/30 bitumen is according to the ratio not significantly different from 50/70 bitumen.

4.3 Final discussion

The results of the series of tests in the programme are consistent. There is no simple, easy-to-use test that provides a stickiness indicator for bitumen. Too many factors other than bitumen adhesion influence the test results. This conclusion, based on the wide range of test methods deployed in this study, has been agreed and adopted by the Industry Group and CEN Ad-Hoc group. Consequently, a proposal was made to adapt the response to the Mandate M/124 in the light of this conclusion. The characteristic Adhesion will be declared “not relevant” in the mandate of TC 336.
The essential requirements that remain for binders are

- Mechanical resistance & stability
  - Consistency at elevated service temperature
  - Consistency at Intermediate service temperature
  - Consistency at low service temperature
  - Stress & strain dependency/Loading time dependency
- Durability
  - Short term ageing
  - Long term ageing
- Safety in case of fire
  - Flash point
- Hygiene, health & the environment
- Safety in use
  - Flash point

5 CONCLUSION AND RECOMMENDATIONS

The work of the Industry Group Adhesion revealed some interesting and important issues when studying adhesion in asphalt mixtures. The main aim however, to find an easy test for adhesion characteristics of bitumen and aggregate, was unsuccessful: there is no such easy test. Other confounding factors, e.g. test temperature or penetration (hardness) of the bitumen, could be affecting the outcome of the so called adhesion tests in this study.

Nevertheless some other conclusions can be drawn from the study:

Test methods that measure a property relying only on a ratio of properties (e.g. before/after conditioning) are very likely to be flawed and have limited use to interpretation. We have shown that the absolute values used to calculate the ratio must be taken into account. A very low initial value before water conditioning combined with only a small change after conditioning can lead to very high ratio and consequently overestimating the performance.

There is a potential artefact due to the influence of conditioning temperature or test temperature. Comparing different binders of the same grade at an elevated conditioning- or test- temperature (above room temperature) will potentially influence the outcome due to different temperature dependence of the binders and not due to the actual adhesion parameter tested.

Standard aggregate can be used to evaluate different binders. However, this will not guarantee the same outcome when this specific aggregate is used in the final asphalt pavement construction. Type testing of the asphalt mixture will always be necessary with the all constituents to be used in the actual road pavement construction.

LITERATURE

1. Nicholls, J.C.; Whiteoak, C.D.; Test procedures for durability and adhesion in asphalt, TRL report PPR187, TRL, Wokingham, UK.
2. EN 12697-45, Test method for Bituminous mixtures - Test methods for hot mix asphalt - Saturation Aging Tensile Stiffness (SATS) conditioning test
3. 42nd Petersen conference, Prediction of Pavement Performance Symposium - Adhesion and Cohesion in Asphalt Pavements, Wyoming, June 20-24, 2005
4. N090 CEN Ad-Hoc Group Adhesion Final report.
5. Wijvekate, Statistical analyses