DETERMINATION OF HEALING AND STRIPPING CHARACTERISTICS OF AC MIXTURES BASED ON THE CHEMICAL PARAMETERS OF MIX COMPONENTS

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ABSTRACT

The calibrated mechanistic surface energy approach (CMSE), developed at Texas A&M University, is one of the fracture damage micromechanical analysis approaches for fundamentally characterizing the properties of asphalt concrete mixtures.

The CMSE approach utilizes the fundamental mix properties including tensile strength, fracture, healing, visco-elasticity, anisotropy, crack initiation and crack propagation to determine fatigue life. This approach requires creep or relaxation, strength and repeated load tests and a catalogue of fracture and healing surface energy components of asphalt binders and aggregates measured separately.

In a cooperation between Delft University of Technology and BAM Wegen, an extensive research program is carried out to verify the findings of the CMSE-approach using Dutch asphalt concrete (AC) mixtures. Both physico-chemical and mechanical properties are determined. In the paper the results of the surface energy measurements on six bitumen and ten aggregates are presented. Based on these results the surface energy components of both bitumen and aggregates and the specific surface area of the aggregates are determined. From these data an estimation of the fracture, healing and stripping characteristics of the resulting asphalt mix can be made.

In the paper the research program is discussed and the findings are presented.

Keywords: Adhesion, Aggregate, Chemical properties, Cohesion, Healing

1. INTRODUCTION

One of the most intriguing properties of an asphalt concrete mix is its ability to recover from mechanical damage. Due to these healing properties, a shift exists between laboratory testing and behaviour of asphalt mixtures in the field. In pavement design procedures, healing is taken into account by the introduction of a healing factor. In the Dutch procedure [1], a healing factor of 4 is used, which implies that due to healing the life span of an asphaltic pavement lasts 4 times longer than calculated with lab testing as input. This factor of 4 was determined decades ago [2], and since that time many things in the asphalt mixture have changed, e.g. new aggregates are used, the properties of the bitumen have been changed or new types of binders are introduced. Due to these changes, the use of the factor 4 for healing is questionable. For this reason, the determination of the healing properties of an asphalt mixture has become an important research topic.

Also in the Netherlands, healing is a popular topic in road engineering. In one of the research programs, the calibrated mechanistic approach with surface energy measurements [3,4] (abbreviated as CMSE), is used as one of the theories to describe the healing process. The approach is based on the theory that an asphalt concrete mixture is a non-linear visco-elastic material and its resistance to fatigue consists of two components: resistance to fracture and the ability to heal. Both of the processes change with time [5]. This approach requires mechanical tests like creep or relaxation, strength and repeated load tests. Also fracture and healing surface energy components of asphalt binders and aggregates are needed [6]. In this paper, the determination of the fracture and healing surface energy components are discussed.

2. THE SURFACE ENERGY PART OF THE CMSE APPROACH

The surface energies play an important role, not only in fracture but also in healing of the AC mixture. The surface energy of the binder and aggregate in the mixture are made up of contributions from non-polar Lifshitz-van der Waals forces and polar acid-base forces mainly associated with hydrogen bonding [7,8,9]. The polar acid-base surface energy is itself a combination of the acid and the base surface energy components.
The difference between the total fracture and healing surface energies lies in the measurement of the individual surface energy components using carefully selected materials with known surface energy component values. Fracture components are found when dewetting, and healing components are determined when wetting occurs [3,7,8,9,10]. In the simplest fundamental theory of energy, if a relatively higher amount of energy is required or must be expended to cause fracture damage (i.e., initiate and propagate a microcrack through the AC layer), then the AC mixture is substantially resistant to fracture damage. If, on the other hand, a higher amount of energy is required or must be expended to repair the fracture damage, then the AC has relatively less potential to self-heal.

Surface energy data constitute input parameters for the healing, crack initiation, and propagation calculations in the CMSE fatigue analysis. The equations (1) through (8) for the SE data analysis required for the CMSE approach, based on an adhesive mode of fracturing under dry conditions, are described hereafter [5].

$$\Delta G_h^{LW} = -\Gamma_{ij}^{LW} + \Gamma_i^{LW} + \Gamma_j^{LW} \tag{1}$$

$$\Delta G_h^{AB} = -\Gamma_{ij}^{AB} + \Gamma_i^{AB} + \Gamma_j^{AB} \tag{2}$$

$$\Delta G_h = \Delta G_h^{LW} + \Delta G_h^{AB} \tag{3}$$

$$\Delta G_f = -\Delta G_f^{LW} + \Delta G_f^{AB} = \left(-\Gamma_{ij}^{LW} + \Gamma_i^{LW} + \Gamma_j^{LW}\right) - \left(-\Gamma_{ij}^{AB} + \Gamma_i^{AB} + \Gamma_j^{AB}\right) \tag{4}$$

$$\Gamma_{ij}^{LW} = \left(\sqrt{\Gamma_i^{LW}} - \sqrt{\Gamma_j^{LW}}\right)^2 \tag{5}$$

$$\Gamma_i^{AB} = 2\left(\sqrt{\Gamma_i^+ \Gamma_i^-}\right) \tag{6}$$

$$\Gamma_{ij}^{AB} = 2\left(\sqrt{\Gamma_i^+} - \sqrt{\Gamma_j^+}\right)\left(\sqrt{\Gamma_i^-} - \sqrt{\Gamma_j^-}\right) \tag{7}$$

$$\Gamma_j^{AB} = 2\left(\sqrt{\Gamma_j^+ \Gamma_j^-}\right) \tag{8}$$

where:

Γ	= surface free energy component of the binder or aggregate (mJ/m^2);
i,j	= subscript "*i*" for binder (healing or fracture) and "*j*" for aggregate;
h,f	= subscript "*h*" for healing and "*f*" for fracture;
LW	= superscript "*LW*" for Lifshitz-van der Waals (*LW*) component;
AB	= superscript "*AB*" for acid-base (*AB*) component;
+	= superscript "+" for Lewis acid component of surface interaction;
−	= superscript "−" for Lewis base component of surface interaction;
Γ*ij*	= interfacial surface energy between binder and aggregate due to "*LW*" or "*AB*" (superscripts) components (mJ/m^2);
ΔG	= total surface free energy due to "*h*" or "*f*" (subscripts) for "*LW*" and/or "*AB*" (superscripts) components (mJ/m^2).

Equations (1) through (8) quantify the bond strength within the binder mastic and the binder-aggregate adhesion. The lower the value of ΔG_h, the greater the potential to self-heal and the higher the value of ΔG_f, the greater the resistance to fracture for AC [5].

3. DETERMINATION OF THESURFACE ENRGY OF BITUMEN AND AGGRAGATES

Different methods exist for the indirect measurement of the surface energy of aggregates and bitumen. For surfaces with a lower surface energy, like bitumen, in this research program the Wilhelmy Plate method is used. The high energy surface of aggregates is accurately measured using the Universal Sorption method, which is based on the adsorption isotherm of the aggregates and the used probe liquids [11].

3.1 Wilhelmy plate method (WP method)

In the Wilhelmy Plate method a thin slide of a specimen is lowered into a probe liquid (Figure 1). The thin slide is hanging under a microbalance. The microbalance measures the change in force of the slide when immersing in the probe liquid. With this change in force the contact angle between the slide and the probe liquid is indirectly measured. An indirect contact angle of a specimen immersing in a probe liquid is called the advancing contact angle. An indirect contact angle of a slide receding from a probe liquid is called the receding contact angle. For the bitumen slides both indirect contact angles are measured. From the indirectly measured contact angles, the surface energy of a specimen can be calculated using a relationship between the contact angle and the surface energy of the specimen.

Each bitumen type is tested with five different probe liquids: distilled water, diiodomethane, glycerol, ethylene glycol and formamide. This results in an over-determined system of equations with five equations and three unknowns. Using the least squares method for this over-determined system of equations results in surface energy components of the bitumen samples.

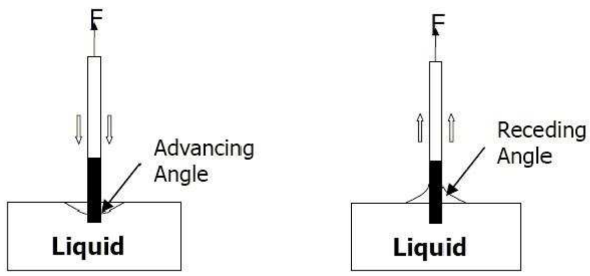

Figure 1: Principle of the Wilhelmy Plate method for a slide coated with bitumen [12].

3.2 Universal Sorption Device (USD)

Surface energy measurements conducted by means of the USD are based on the fact that molecules in the atmosphere are adsorbed and desorbed on a surface under influence of pressure, temperature and surface energy of the adsorbent. At the start of the test a vacuum is created in the testing chamber surrounding the aggregates. The pressure is increased by adding one liquid vapour in the testing chamber at constant temperature. Now molecules from the vapour are adsorbed on the surface of the aggregates. At some moment in time the number of molecules adsorbing onto and desorbing from the aggregates are equal resulting in an equilibrium (Figure 2).

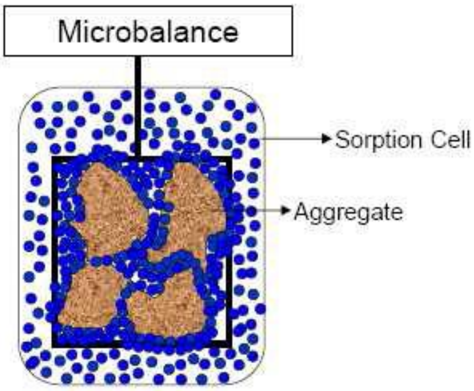

Figure 2: Principle of USD method [13].

At this moment the mass of the aggregates including the adsorbed vapour is measured. Then the pressure is increased again by adding the probe liquid vapour in the testing chamber at constant temperature. Now the aggregates including the adsorbed molecules are weighed again. An adsorption isotherm is produced by repeating the previous procedure for about 8 to 10 pressure levels. An adsorption isotherm shows the relationship between the equilibrium mass of the vapour adsorbed on the solid surface and partial vapour pressure of the probe vapour.

4. EXPERIMENTAL PROGRAM

The experimental program covers six bitumen types and ten aggregates. Both bitumen and aggregates are used in daily practice:

- Both standard penetration bitumen and polymer modified binders are tested. The types of bitumen are encrypted with the code B1 to B6;
- 10 different kinds of aggregates are tested. All these aggregates are used in asphalt mixtures in the Netherlands. Some of these aggregates are found in the Netherlands, but most of the aggregates are imported from abroad. Also some waste and recycled aggregates are within the test program.
  The aggregates are encrypted with the code A1 to A10.

After performing the tests with the WP-method and the USD, the surface energy components of the bitumen and the aggregates are computed from the output parameter from the tests, contact angles for the bitumen samples and spreading pressures for the aggregates. Finally the surface energy results are translated to the input parameters of the CMSE approach, which can be calculated with only the surface energy experiments conducted.

5. TEST RESULTS

5.1 Surface energy bitumen

The surface energy components are computed for the measured advancing and receding contact angle. The computed surface energy components of the bitumen samples for the advancing contact angles are given in table 1. and for the receding contact angles in table 2.

Table 1: Surface energy components of the bitumen samples using the Wilhelmy Plate Test, from advancing contact angles in (mJ/m²).

Bitumen	γ_s^{TOT}	γ_s^{LW}	Stdev.	γ_s^{+}	Stdev.	γ_s^{-}	Stdev.
B1	19.05	17.9	0.6	0.2	0.1	1.9	0.3
B2	29.57	25.7	0.8	0.1	0.1	26.6	1.8
B3	16.40	13.1	0.5	0.6	0.1	4.8	0.7
B4	29.93	29.9	0.5	0.0	0.0	0.1	0.1
B5	18.37	17.0	0.4	0.3	0.1	1.6	0.4
B6	21.97	22.0	0.8	0.0	0.0	2.5	0.3

Table 2: Surface energy components of the bitumen samples using the Wilhelmy Plate Test, from receding contact angles in (mJ/m²).

Bitumen	γ_s^{TOT}	γ_s^{LW}	Stdev.	γ_s^{+}	Stdev.	γ_s^{-}	Stdev.
B1	40.73	38.2	0.7	0.1	0.0	15.5	0.9
B2	45.57	39.1	0.8	0.6	0.1	16.5	1.5
B3	43.16	38.2	0.8	0.3	0.1	23.3	1.3
B4	47.46	37.6	0.9	1.3	0.2	19.3	1.1
B5	39.94	34.9	1.3	0.3	0.1	22.2	1.0
B6	39.63	39.6	0.9	0.0	0.0	12.2	0.8

Bitumen are generally expected to show a non-polar behaviour with advancing contact angles. This means with a very high non-polar component (γ_s^{LW}) and with very small polar components (γ_s^{+}, γ_s^{-}). This behaviour is very well illustrated by all bitumen except bitumen B2. Bitumen B2 has a remarkable low advancing contact angle with water. This results in quite a large polar component as shown in table 1. The Lewis base component of 26.6 mJ/m^2 is even larger than the Lewis base component of water (25.5 mJ/m^2). Within the bitumen there is a large deviation between the non-polar components. For instance the non-polar component of B3 is more than halve that of bitumen B4. A higher non-polar component tends to have better resistance against water in asphalt mixtures. Surface energy calculated from receding contact angles for bitumen are normally larger than the surface energy calculated from advancing contact angles. This can also be seen in these measurement results. Only the Lewis base component of bitumen B2 decreases if calculated from receding contact angles.

5.2 Aggregate surface energy

The results of the experiments of the USD are measured spreading pressures for the three probe liquids with the aggregate samples. The fundamental work of adhesion between a probe liquid and the surface of an aggregate can be written as:

$$W_a = \Pi_e + \gamma_l^{tot}(1 + cos\theta) \quad (9)$$

where:

W_a = the fundamental work of adhesion;
Π_e = the spreading pressure at saturation vapour pressure of the probe liquid;
γ_i^{tot} = the total surface energy of the probe liquid;
θ = contact angle between probe liquid and aggregate surface.

In adsorption tests the contact angle between probe liquid and aggregate surface, θ, is per definition going to zero, resulting in.

$$W_a = \Pi_e + 2\gamma_l^{tot} \tag{10}$$

The van Oss theory relates the surface energy of the probe liquid and the solid surface with the fundamental work of adhesion [7], according:

$$W_a = 2\left(\sqrt{\gamma_l^{LW} \cdot \gamma_s^{LW}} + \sqrt{\gamma_l^- \cdot \gamma_s^+} + \sqrt{\gamma_l^+ \cdot \gamma_s^-}\right) \tag{11}$$

where:
W_a = work of adhesion;
γ_1^{LW} = free energy of Lifshitz-van der Waals forces of bitumen;
γ_s^{LW} = free energy of Lifshitz-van der Waals forces of aggregate;
γ_1^- = contribution of Lewis base of bitumen;
γ_s^+ = contribution of Lewis acid of aggregate;
γ_1^+ = contribution of Lewis acid of bitumen;
γ_s^- = contribution of Lewis base of aggregate.

Now a similar system of equations as with the contact angles of the Wilhelmy Plate can be established. However, now relations between spreading pressures and surface energy components of the aggregates are considered using the probe liquids distilled water, n-hexane and methylpropylketone. Three equations with three unknows surface energy components of the aggregates occur, which can be solved easily. In table 3 the surface energy components are given.

Table 3: Surface energy components of the different stone samples in (mJ/m²) and SSA in (m^2/g).

Aggregate	γ_s^{TOT}	γ_s^{LW}	γ_s^+	γ_s^-	SSA
A1	201.72	49.03	7.27	801.84	0.94
A2	119.93	49.12	1.08	1165.47	0.49
A3	50.00	43.83	0.01	1529.47	0.40
A4	89.29	47.97	1.04	408.87	0.76
A5	105.68	45.64	0.68	1332.82	0.34
A6	182.06	54.88	3.00	1348.21	0.95
A7	458.14	46.69	53.61	789.46	1.49
A8	111.41	40.20	1.18	1078.78	0.50
A9	149.62	49.13	2.10	1204.49	0.92
A10	173.18	52.66	1.76	2061.17	0.22

As can be seen the non-polar surface energy component (γ_s^{LW}) does not vary much over the different aggregates. However the polar components (γ_s^+, γ_s^-) show quite some deviation. Very remarkable is the very high Lewis acid component (γ_s^+) of aggregate A7. In general, natural aggregates tend to show a mono-polar behaviour with a large Lewis base component (γ_s^-) and an almost non-existing Lewis acid component (γ_s^+). This behaviour is very clear by the other aggregates. Interesting to see is also the very low Lewis base component of aggregate A4, which is 5 times smaller than the same component of aggregate A10.

5.3 Input parameters of the CMSE approach

The components of equations (5) to (8) can be calculated with the measured surface energy components of the bitumen and aggregate combinations. The non-polar input parameter for the healing, crack initiation, and propagation is calculated with equation (5), which uses the non-polar surface energy components of the bitumen and the aggregate. The polar input parameter of the bitumen can be calculated by multiplying the Lewis acid and Lewis base component of the bitumen according equation (6). The polar input parameter of the interacting bitumen and aggregate can be calculated with equation (7). The required surface energy components are the Lewis acid and Lewis base of the bitumen and the Lewis base component of the aggregate. The polar input parameters of the aggregates are calculated in the same way as the polar input parameter of the bitumen, but now using equations (1) and (2). The results of all the bitumen aggregates combinations are given in the tables 4 and 5.

Table 4: Non-polar part of the surface energies due to healing of all the bitumen-aggregate combinations in (mJ/m²).

ΔG_h^{LW}	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1	59.28	59.34	56.05	58.64	57.20	62.72	57.85	53.68	59.34	61.44
B2	70.99	71.06	67.12	70.22	68.49	75.11	69.28	64.29	71.07	73.58
B3	50.67	50.71	47.90	50.11	48.88	53.60	49.44	45.88	50.72	52.51
B4	76.62	76.69	72.44	75.78	73.92	81.06	74.77	69.38	76.70	79.41
B5	57.80	57.86	54.65	57.17	55.77	61.15	56.41	52.34	57.86	59.91
B6	65.62	65.68	62.04	64.90	63.30	69.42	64.03	59.42	65.68	68.00

Table 5: Polar part of the surface energies due to healing of all the bitumen-aggregate combinations in (mJ/m²)

ΔG_h^{AB}	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1	30.72	30.94	32.41	19.45	32.30	34.98	43.29	30.01	32.54	41.01
B2	49.08	36.35	30.28	25.73	35.92	45.45	96.62	35.86	41.01	47.81
B3	54.67	56.25	59.63	35.10	58.90	63.19	74.53	54.50	58.90	74.58
B4	1.68	0.65	0.05	0.64	0.51	1.08	4.57	0.68	0.90	0.83
B5	36.94	38.98	41.90	24.11	40.97	43.48	48.31	37.72	40.62	51.71
B6	8.77	3.63	0.71	3.43	2.98	5.84	23.26	3.76	4.92	4.67

The total surface energy due to healing is the sum of the non-polar and the polar part according to equation (3). The total surface energies due to healing of all the bitumen and aggregate combinations are given in table 6. In the CMSE fatigue analysis approach the surface energies calculated from advancing contact angles are taken for the due to healing.

Table 6: Surface energies due to healing of all the bitumen and aggregate combinations in (mJ/m²).

ΔG_h	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1	90.01	90.28	88.46	78.09	89.50	97.70	101.14	83.69	91.88	102.45
B2	120.08	107.41	97.40	95.95	104.41	120.56	165.90	100.15	112.08	121.38
B3	105.34	106.97	107.53	85.21	107.79	116.80	123.98	100.37	109.62	127.09
B4	78.30	77.34	72.49	76.42	74.43	82.14	79.34	70.06	77.60	80.23
B5	94.74	96.84	96.55	81.29	96.74	104.63	104.72	90.06	98.48	111.62
B6	74.38	69.31	62.74	68.33	66.29	75.26	87.29	63.18	70.61	72.67

Finally the surface energy due to fracture or dewetting can be calculated using equation (4). The value of the surface energies due to fracture or dewetting of all the bitumen and aggregate combinations are given in table 7. In the CMSE fatigue analysis approach the surface energies due to fracture or dewetting are calculated from the receding contact angles of the bitumen.

Table 7: Surface energies due to fracture or dewetting of all the bitumen and aggregate combinations in (mJ/m²).

ΔG_f	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1	125.84	116.56	107.42	106.56	113.24	128.61	160.06	107.84	120.16	129.08
B2	154.59	150.49	145.83	127.14	149.35	165.23	189.68	140.45	154.73	173.90
B3	141.66	131.69	122.79	116.23	128.91	145.98	184.04	122.55	136.24	149.11
B4	173.28	171.90	169.97	139.41	172.24	188.70	211.34	161.20	176.79	202.84
B5	138.70	129.46	121.30	113.27	127.01	143.51	180.01	120.61	133.94	147.31
B6	106.98	95.48	83.95	94.33	90.79	105.36	137.16	87.40	98.35	100.63

With these non-polar and the polar part of the total surface energies due to healing are calculated. However at this moment the healing indices cannot be calculated yet, because the parameters from the mechanical tests are not determined yet.

6. WORK OF ADHESION AND RESISTANCE TO STRIPPING

In this last paragraph two other applications of surface energy are described. The fundamental work of adhesion is calculated together with the resistance against stripping of all bitumen-aggregate combinations.

6.1 Work of adhesion

When the surface energy components of the solid, e.g. a stone, and the liquid, e.g.bitumen, are measured and known, the fundamental work of adhesion can be calculated. This fundamental work of adhesion is the energy required to separate the bitumen molecules from the stone surface. The fundamental work of adhesion can be interpreted as the least amount of energy that is necessary to split the bitumen from the stone surface. With equation (11) the fundamental work of adhesion is calculated for all the bitumen and aggregate combination. The results are given in table 8.

Table 8: Fundamental work of adhesion between the bitumen-aggregate combinations in (mJ/m²).

	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1[a]	90.01	90.28	88.46	78.09	89.50	97.70	110.17	83.69	91.88	102.45
B2[a]	120.08	107.41	97.40	95.95	104.41	120.56	165.90	100.15	112.08	121.38
B3[a]	105.34	106.97	107.53	85.21	107.79	116.80	123.98	100.37	109.62	127.09
B4[a]	78.30	77.34	72.49	76.42	74.43	82.14	79.34	70.06	77.60	80.23
B5[a]	94.74	96.84	96.55	81.29	96.74	104.63	104.72	90.06	98.48	111.62
B6[a]	74.38	69.31	62.74	68.33	66.29	75.26	87.29	63.18	70.61	72.67
B1[r]	125.84	116.56	107.42	106.56	113.24	128.61	160.06	107.84	120.16	129.08
B2[r]	154.59	150.49	145.83	127.14	149.35	165.23	189.68	140.45	154.73	173.90
B3[r]	141.66	131.69	122.79	116.23	128.91	145.98	184.04	122.55	136.24	149.11
B4[r]	173.28	171.90	169.97	139.41	172.24	188.70	211.34	161.20	176.79	202.84
B5[r]	138.70	129.46	121.30	113.27	127.01	143.51	180.01	120.61	133.94	147.31
B6[r]	106.98	95.48	83.95	94.33	90.79	105.36	137.16	87.40	98.35	100.63

[a] means surface energy of bitumen from advancing contact angles. [r] means from receding contact angles. Green means the best 25%, yellow the next 25%, orange the next 25% and red the lowest scoring 25%.

From the table it becomes clear that the calculated fundamental adhesion of aggregate A7 with almost all bitumen scores very high. This can especially be explained by the very large Lewis acid component (γ_s^+) measured for aggregate A7. This component is multiplied by the large Lewis base components from the bitumen. According to these calculations aggregate A4 has the lowest scores of all aggregates. Bitumen B2 seems to be a good bitumen with respect to fundamental adhesion. Bitumen B6 has the lowest scores for all bitumen. If the surface energy of bitumen B4 is calculated from advancing contact angles, bitumen B4 has one of the lowest scores for the fundamental work of adhesion. However if the surface energy from receding contact angles is used, bitumen B4 has the best scores with all the aggregates.

6.2 Resistance to stripping

Stripping occurs when the interfacial energy between bitumen and aggregate is lower than the interfacial energy between water and aggregate. The correlation between the surface properties of materials and their tendency to strip in the presence of water is relatively well established in the literature. The three quantities based on the surface energies of asphalt binders and aggregate that are related to the moisture sensitivity of an asphalt mixture are [11]:

- work of adhesion between bitumen and aggregate W_{AB}
- work of debonding or reduction in free energy of the system when water displaces the bitumen from a binder-aggregate interface W_{ABW},
- work of cohesion of bitumen or mastic, W_{BB}.

The work of adhesion of the bitumen-aggregate system under the influence of water can be calculated using the following equation:

$$W_{ABW}^{wet} = \gamma_{AW} + \gamma_{BW} - \gamma_{AB} \tag{12}$$

where:

W = work of debonding or reduction in free energy of the system when water displaces the bitumen from a binder-aggregate interface;

γ_{AW} = the interfacial energy between aggregate and water;

γ_{BW} = the interfacial energy between bitumen and water;

γ_{AB} = the interfacial energy between aggregate and bitumen.

The two energy terms W_{AB} and W_{ABW} were combined into a single parameter ER_1 that is directly proportional to the moisture resistance of the asphalt mixture as:

$$ER_1 = \left|\frac{W_{AB}}{W_{ABW}^{wet}}\right| \tag{13}$$

where:

W_{AB} = work of adhesion between bitumen and aggregate;

W_{ABW} = work of debonding or reduction in free energy of the system when water displaces the bitumen from a binder-aggregate interface.

A higher ratio of ER_1 shows a higher resistance against stripping, because in that case the work of adhesion between bitumen and aggregate is relatively larger. The parameter ER_1 is calculated for all the bitumen and aggregate combinations. The results are shown in table 9.

Table 9: Stripping parameter ER_1 for all bitumen-aggregate combinations.

	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
$B1^a$	0.447	0.371	0.314	0.687	0.340	0.359	0.473	0.363	0.366	0.293
$B2^a$	0.554	0.396	0.449	0.678	0.355	0.408	0.776	0.385	0.405	0.322
$B3^a$	0.549	0.461	0.561	0.758	0.430	0.451	0.574	0.456	0.458	0.384
$B4^a$	0.374	0.306	0.346	0.682	0.271	0.289	0.316	0.291	0.296	0.218
$B5^a$	0.485	0.411	0.494	0.742	0.379	0.396	0.458	0.403	0.404	0.329
$B6^a$	0.340	0.261	0.287	0.544	0.230	0.253	0.351	0.249	0.257	0.190
$B1^r$	0.604	0.448	0.515	0.830	0.401	0.452	0.737	0.432	0.451	0.352
$B2^r$	0.829	0.647	0.782	1.110	0.590	0.649	0.976	0.627	0.649	0.530
$B3^r$	0.696	0.515	0.603	0.896	0.464	0.525	0.901	0.498	0.522	0.417
$B4^r$	1.000	0.793	0.981	1.294	0.730	0.797	1.186	0.773	0.797	0.665
$B5^r$	0.683	0.509	0.597	0.878	0.460	0.518	0.880	0.494	0.516	0.414
$B6^r$	0.485	0.348	0.380	0.704	0.304	0.350	0.588	0.332	0.350	0.259

To account for the influence of the wetting properties of the bitumen in the sensitivity to stripping a new parameter was developed, ER_2. This parameter is the same as parameter ER_1 only the cohesive bond is deducted from the work of adhesion between the asphalt binder and aggregate:

$$ER_2 = \left|\frac{W_{AB}-W_{BB}}{W_{ABW}^{wet}}\right| \tag{14}$$

where
W_{BB} = cohesive bond energy of the bitumen.

Again a higher ratio of the parameter shows a higher resistance against stripping, because in that case the work of adhesion between bitumen and aggregate is relatively larger. The parameter ER_2 is calculated for all the bitumen and aggregate combinations in this project. The results are shown in table 10.

Table 10: Stripping parameter ER_2 for all bitumen-aggregate combinations.

	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
$B1^a$	0.258	0.215	0.179	0.352	0.195	0.219	0.309	0.198	0.214	0.184
$B2^a$	0.281	0.178	0.176	0.260	0.154	0.208	0.499	0.158	0.191	0.165
$B3^a$	0.378	0.319	0.390	0.466	0.299	0.324	0.422	0.307	0.321	0.285
$B4^a$	0.088	0.069	0.060	0.148	0.053	0.078	0.077	0.042	0.068	0.055
$B5^a$	0.297	0.255	0.306	0.407	0.235	0.257	0.298	0.239	0.253	0.220
$B6^a$	0.139	0.095	0.086	0.194	0.077	0.105	0.174	0.076	0.097	0.075
$B1^r$	0.213	0.135	0.124	0.195	0.112	0.166	0.362	0.106	0.145	0.130
$B2^r$	0.340	0.255	0.293	0.314	0.230	0.291	0.507	0.220	0.267	0.252
$B3^r$	0.272	0.177	0.179	0.231	0.153	0.214	0.479	0.147	0.191	0.176
$B4^r$	0.452	0.355	0.433	0.413	0.328	0.396	0.654	0.318	0.369	0.354
$B5^r$	0.290	0.195	0.204	0.259	0.171	0.230	0.489	0.167	0.208	0.190
$B6^r$	0.126	0.059	0.021	0.113	0.039	0.087	0.248	0.031	0.068	0.055

In the tables 9 and 10 it is shown that particularly aggregates A4 and A7 have very high values for the parameters ER_1 and ER_2. This means that these aggregates are expected to have the largest resistance against stripping, although this depends on the bitumen as well. For instance bitumen B6 has clearly one of the lowest scores even with aggregates A4 and A7. Remarkable again is that bitumen B4 has the lowest scores combined with all the different aggregates if the surface energy is calculated from advancing contact angles, but has the highest scores if the surfaces energy is calculated from receding contact angles. Among the tested bitumen, bitumen B3 seems to have the best overall scores in the combination with all the aggregates.

The moisture sensitivity of asphalt mixtures is also inversely related to the overall microtexture of the aggregate surfaces, which is approximately proportional to its specific surface area (SSA). In order to accommodate the influence of surface roughness at a micro level, two additional parameters were considered:

$$ER_1 \cdot SSA = \left|\frac{W_{AB}}{W_{ABW}^{wet}}\right| \cdot SSA \tag{15}$$

$$ER_2 \cdot SSA = \left|\frac{W_{AB}-W_{BB}}{W_{ABW}^{wet}}\right| \cdot SSA \tag{16}$$

where:
SSA = specific surface area of aggregate.

Higher ratios of these parameters show higher resistance against stripping. These stripping parameters are calculated for all the bitumen and aggregate combinations in this project. The results are shown in tables 11 and 12.

Table 11: Stripping parameter $ER_1 \cdot SSA$ for all bitumen-aggregate combinations.

	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1[a]	0.420	0.183	0.124	0.523	0.115	0.339	0.705	0.180	0.337	0.064
B2[a]	0.520	0.194	0.178	0.517	0.120	0.387	1.157	0.191	0.373	0.071
B3[a]	0.516	0.226	0.222	0.577	0.146	0.426	0.856	0.226	0.421	0.085
B4[a]	0.351	0.150	0.137	0.520	0.092	0.273	0.470	0.144	0.272	0.048
B5[a]	0.455	0.202	0.195	0.566	0.128	0.374	0.684	0.200	0.372	0.072
B6[a]	0.319	0.128	0.113	0.415	0.078	0.240	0.523	0.124	0.237	0.042
B1[r]	0.567	0.220	0.204	0.633	0.136	0.428	1.099	0.214	0.416	0.078
B2[r]	0.779	0.318	0.309	0.846	0.200	0.614	1.456	0.311	0.598	0.117
B3[r]	0.654	0.253	0.239	0.683	0.157	0.497	1.344	0.247	0.480	0.092
B4[r]	0.939	0.390	0.388	0.986	0.247	0.754	1.769	0.383	0.734	0.147
B5[r]	0.642	0.250	0.236	0.669	0.156	0.490	1.311	0.244	0.475	0.091
B6[r]	0.455	0.171	0.151	0.537	0.103	0.331	0.876	0.164	0.322	0.057

Table 12: Stripping parameter $ER_2 \cdot SSA$ for all bitumen-aggregate combinations.

	A1	A2	A3	A4	A5	A6	A7	A8	A9	A10
B1[a]	0.242	0.106	0.071	0.268	0.066	0.207	0.461	0.098	0.197	0.040
B2[a]	0.264	0.087	0.070	0.198	0.052	0.197	0.744	0.078	0.176	0.036
B3[a]	0.355	0.157	0.154	0.355	0.101	0.307	0.629	0.152	0.295	0.063
B4[a]	0.083	0.034	0.024	0.113	0.018	0.074	0.115	0.021	0.062	0.012
B5[a]	0.279	0.125	0.121	0.310	0.080	0.243	0.444	0.118	0.233	0.049
B6[a]	0.131	0.047	0.034	0.148	0.026	0.100	0.260	0.038	0.089	0.017
B1[r]	0.200	0.066	0.049	0.149	0.038	0.157	0.539	0.052	0.134	0.029
B2[r]	0.320	0.125	0.116	0.240	0.078	0.275	0.756	0.109	0.246	0.056
B3[r]	0.255	0.087	0.071	0.176	0.052	0.203	0.714	0.073	0.176	0.039
B4[r]	0.425	0.175	0.171	0.315	0.111	0.375	0.974	0.157	0.340	0.078
B5[r]	0.272	0.096	0.081	0.197	0.058	0.217	0.729	0.083	0.192	0.042
B6[r]	0.118	0.029	0.008	0.086	0.013	0.082	0.370	0.015	0.063	0.012

Remarkable to see is that the importance of the bitumen becomes much less when stripping parameters $ER_1 \cdot SSA$ and $ER_2 \cdot SSA$ are used instead of stripping parameters ER_1 and ER_2. Table 11 and 12 show that according to the parameters $ER_1 \cdot SSA$ and $ER_2 \cdot SSA$ aggregates A7, A4 and A1 have the best anti-stripping potential. A10 and A5 have the lowest scores for these stripping parameters. The bitumen type seems not much of an importance. These aggregates A10 and A5 are expected to have the lowest resistance against stripping. This is mainly because these aggregates have the lowest SSA compared to the other tested aggregates (for A10: 0.22 m^2/g and for A5: 0.34 m^2/g).

7. CONCLUSIONS AND RECOMMENDATIONS

Based on the results of this research project, the following conclusions and recommendations can be formulated:

1. The CMSE fatigue analyses approach shows a way to explain the resistance against continuous loading of asphalt mixtures from a fundamental starting point. The model takes into account healing and anisotropic effects and bridges the gap between molecular theory and material behaviour;
2. The different bitumen show practically non-polar behaviour with advancing contact angles. This is in accordance with bitumen in general. Only bitumen B2 has a distinct surface energy, with a very high Lewis base component larger than the Lewis base component of water;
3. Almost all aggregates show practically mono-polar behaviour. This is in accordance with aggregates generally found in literature. However the deviation in the size of the Lewis base component is quite significant. The base component for A4 is about 5 times smaller than the base component of aggregate A10. Only aggregate A7 has not particularly a mono-polar behaviour. From the fundamental adhesion and stripping parameters calculations it is clear this has a very large effect;
4. Aggregates A7 and A4 have the highest scores for the stripping parameters and therefore according to the theory are expected to have the largest resistance against stripping. Aggregate A10 scores low for all stripping parameters. Aggregate A5 scores lower for stripping parameters $ER_1 \cdot SSA$ and $ER_2 \cdot SSA$. This is mainly caused by the additional importance of the specific surface area. Aggregate A5 has the lowest SSA after aggregate A10. Bitumen B3 has high scores for stripping parameters ER_1 and ER_2 and bitumen B6 has overall one of the lowest scores for stripping

parameters ER_1 and ER_2. Bitumen B4 is expected to have an overall good resistance against stripping, if calculations are made from receding contact angles.

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