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The influence of boundary conditions on the healing of bitumen

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Damage in pavements is known to reduce over time when the material is left to rest, this phenomenon is identified as healing. It has been shown that healing has a large influence on pavement performance. However, as the healing mechanism is not fully understood, there is currently no accepted method available to assess the healing performance of an asphalt binder. Healing of cracks can be seen as the sum of two processes, cracked surfaces coming into contact (wetting) and strength gain of the contact area (intrinsic healing). This paper aims to increase the understanding of the process of two surfaces coming into contact. Healing of bitumen is assessed using a novel test method, which allows for controlled variation of the stress state during healing. This method consists of bringing two pieces of bitumen together and allowing them to heal under controlled conditions. The extent of healing is then assessed by testing the healed specimens in direct tension. The results, presented in this paper, show that the stress state at assembly and during healing has a significant impact on the extent of healing.

Keywords: healing; bitumen; mechanisms

1. Introduction

Healing refers to asphalt's favourable material property to restore damage autonomously during periods when no load is applied. After a rest period, lost material performance is regained, e.g. strength and stiffness. Both in the laboratory and in practice, asphalt scientist have demonstrated the healing capacity of asphalt (Bazin & Saunier, 1967; Bonnaure, Huibers, & Boonders, 1982; Little, Bhasin, & Darabi, 2015; Maillard, de La Roche, Hammoum, Such, & Piau, 2004; Williams, Little, Lytton, Kim, & Kim, 2001). Consequently, healing is incorporated in many of the guidelines for road design (Van den bergh, 2011).

Almost all research shows that the level of healing increases with longer resting periods and higher healing temperatures; a good overview of healing research can be found in Qiu (2012). Next to this it is also shown that smaller cracks heal more easily compared to larger cracks (Moreno-Navarro, Sol-Sánchez, & Rubio-Gámez, 2015; Qiu, 2012). In the research of Bazin and Saunier (1967) it was demonstrated that a small normal force, perpendicular to the crack surface, was required for good healing. In spite of these insights, there is still limited uniformity between healing measured using different healing test set-ups. For instance, load-controlled healing test normally results in much more healing compared to displacement-controlled healing tests (Lu, Soenen, & Redelius, 2003). Also a four-point bending test will result in less healing compared to a three-point bending test, in a two-point bending test the observed healing is largest (Westera,

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1994). And even though standardised test set-ups are formulated for the fatigue of binders, there is no fundamental explanation why different types of binders display different healing behaviour (Pereira, Micaelo, Quaresma, & Cidade, 2016). As a consequence, it can be concluded that the healing mechanisms in asphalt are not yet unravelled. Due to this lack of insight, it remains debatable which test could best be used to determine the healing characteristics of a material. Consequently, it is not possible to assess the healing performance of an innovative material compared to standard materials unambiguously.

In order to increase the understanding of healing mechanisms, Kim, Little, and Benson (1990) and Little et al. (2015) have proposed the adoption of a healing model from polymer science. This model assumes a five-step healing model as illustrated in Figure 1. The steps are the approach of damaged surfaces (1,2), surfaces wetting each other to create an interface (3), diffusion of molecules over the interface (4) until the material has become homogenous again (5). It is assumed that once the interface is homogeneous again, the original material properties are restored.

Intuitively, this model seems reasonable and one could image that more insight in the separate processes described by the model could improve the understanding of the healing behaviour observed in laboratory tests. However, until now, the focus when applying this model to asphaltic materials has been on the diffusion part. Test set-ups have been formulated to create instant full wetting and study the strength increase as a consequence of diffusion. The creation of contact between surfaces has received less attention. Nevertheless, the work of Qiu (2012) has demonstrated a large impact of boundary conditions on the level of observed healing. As low stresses in a material hardly affect diffusion, these results demonstrate the importance of the surfaces coming into contact on the macroscopic observed healing.

In each test method the amount of stress applied to the material varies, consequently the boundary conditions around micro damage are also expected to be test method specific. If the boundary conditions influence the healing, this could explain large differences obtained in different test methods.

This paper aims to provide an insight on the impact of boundary conditions around the damaged area on the level of healing. In order to investigate this, a newly developed test method is used (Leegwater, Scarpas, & Erkens, 2016). The innovative aspects of this new test method are the execution of the test in direct tension (mode I) and the possibility to control the level of normal force perpendicular to the damage during healing. The healing model presented above is taken as a starting point to interpret test results.

The paper will continue by presenting the healing model in more detail. Next, a description of the used test method and materials is provided. After this, the test results are presented, demonstrating the impact of healing time and stress state during healing. Finally, the test results will be discussed in light of the healing model, in an attempt to assess the importance of boundary conditions.

2. Healing model

2.1. *Conceptual healing model*

As already stated the polymer healing model assumes five stages in crack healing (Wool & O'Connor, 1981). Within these five stages, there are two key processes; firstly, surfaces have to be in contact to transfer loads, this process of two surfaces coming into close contact is referred to as *wetting*. Secondly, surfaces in contact have an ability to transfer loads, which increases over time due to homogenisation of the contact area. This second process, describing the load-bearing capacity of the interfaces in contact, is named *intrinsic healing*. As wetting and intrinsic healing

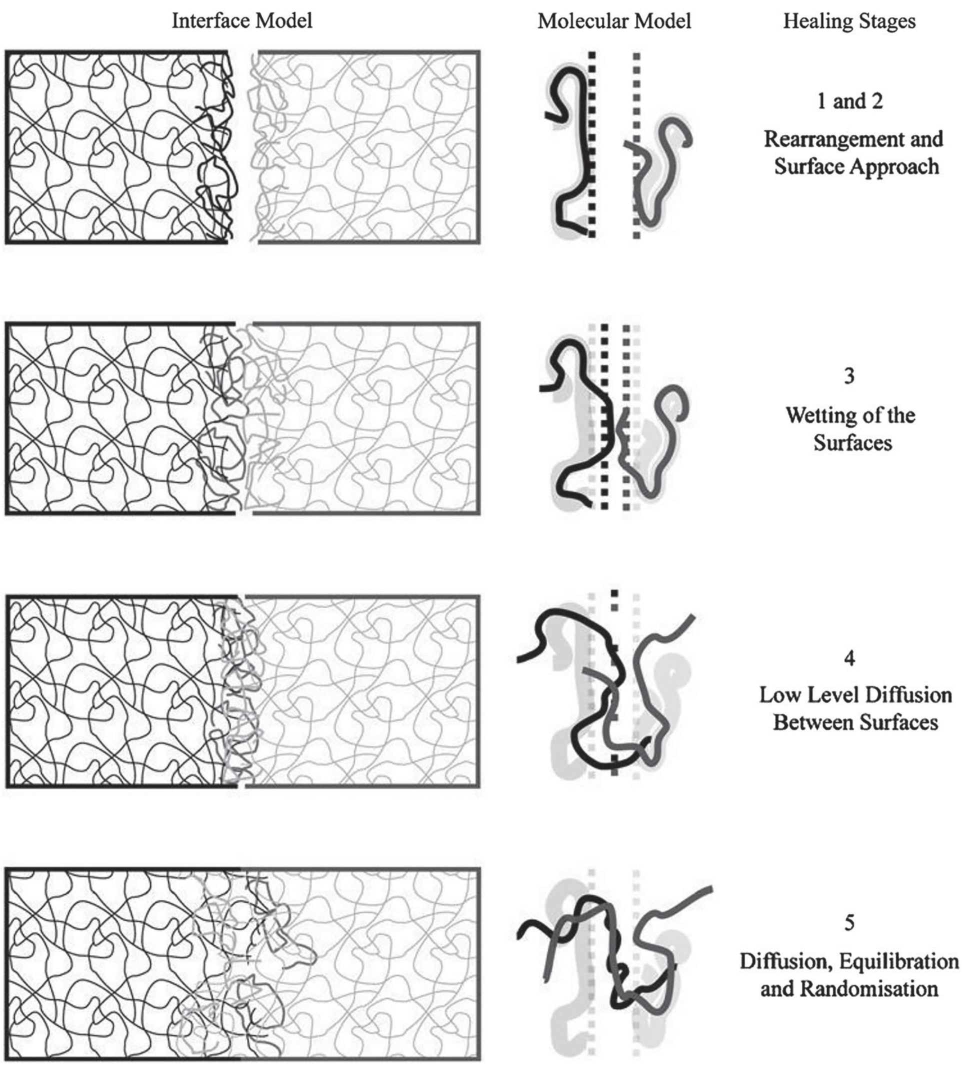

Figure 1. Five steps in the healing process (Wool & O'Connor, 1981; Wu, Meure, & Solomon, 2008).

are physically two different processes they both have their own influencing parameters. Below the influencing parameters for both processes will be discussed in more detail.

In order to understand the process of wetting, we have to understand the factors that play a role in wetting. First of all, the geometry of the surface is of large importance. In case a surface is very rough, the amount of area that comes into close contact when two surfaces are brought together is very limited. This immediately implies that a surface which deforms easily has more potential to gain contact, as initial roughness can change under the influence of loads. In order for two surfaces to come together, there has to be some kind of force that brings them together. In a visco-elastic material there are three mechanisms that can bring the surfaces of internal cracks together; temperature expansion, deformation caused by an external applied force or deformation caused by residual stresses. Next to this it is also possible that the force that brings surfaces together originates from short-range attractions that are a result of surface energies (Schapery, 1989). For soft viscous and visco-elastic materials like bitumen, it has been demonstrated that molecular attractions in the area surrounding a contact area are large enough for the contact area to increase in size over time. Summarising, the ease at which surfaces wet each other is determined by the characteristics of the surfaces (e.g. roughness), the resistance of the material to deformation (visco-elastic properties) and the driving force that brings the surfaces together (external load, thermal expansion, delayed visco-elastic deformations and surface energy).

Intrinsic healing is characterised by the amount of load that can be transferred by two surfaces in contact. The moment two surfaces come in full contact, there will instantly be some level of adhesion as a consequence of surface energy. Over time, molecules will diffuse over the interface and as a consequence of the rearrangement and entanglement, the load-bearing capacity will increase. Given enough time, this process will result in the disappearance of the interface and the reestablishment of the original material properties. Consequently, influencing factors of intrinsic healing are the surface energy of the material and the chemical mobility (self-diffusion rate) of the bitumen.

It should be noted that some of the influencing parameters as given above interact, for instance the size and polarity of molecules will affect the self-diffusion rate, the visco-elastic properties and the surface energy.

2.2. *Mathematical healing model*

The schematics presented in Figure 1 represent the healing of a single crack; here the process of intrinsic healing starts after wetting is completed. When healing is measured on a laboratory scale, not all areas come in contact at the same moment in time. In this case each segment of the surface will be at its own particular stage of healing. Therefore, the macroscopic observed healing is the sum of the total area in contact, multiplied by the time that each segment has been in contact. Mathematically, this can be represented by a convolution integral for the processes of wetting and healing. This formula is already proposed by Wool and O'Connor (1981) and given below

$$R = \int_{\tau=-\infty}^{\tau=t} R_h(t-\tau)\frac{\mathrm{d}\phi(\tau,X)}{\mathrm{d}\tau}\mathrm{d}\tau. \tag{1}$$

In Equation (1) R is the ratio of the healed performance compared to the original performance and ranges from 0 to 1. The formula presents healing as the convolution of a wetting function $\phi(\tau,X)$ and an intrinsic healing function R_h, τ is a running variable on the time axis.

The wetting function is formulated assuming that the contact created between the surfaces has multiple nucleation points that grow in a circular direction over time. This is graphically demonstrated in Figure 2. The number of nucleation points will increase over time and from each nucleation point a contact area will grow. Over time the circular areas will merge until the whole area is in contact. Mathematically such an equation is of the Avrami type. Wool and O'Conner (1981) present the following formula for wetting:

$$\phi(t) = 1 - e^{-kt^m}. \tag{2}$$

The intrinsic healing function is expected to have an initial level R_0 that originates from surface energy. Next to this, the function will increase over time, proportional to the amount of molecules that have diffused over the interface. There are different proposals for the shape of this curve (Bhasin, Little, Bommavaram, & Vasconcelos, 2008; Wool & O'Connor, 1981). A graphical overview of the influence of wetting and intrinsic healing and predicted shape of the healing processes described above is given in Figure 3.

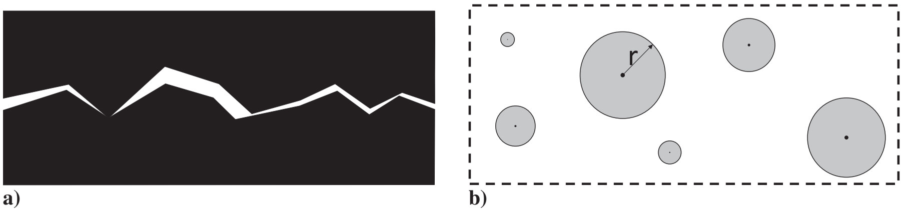

Figure 2. Schematic representation of wetting (a) side view of two surfaces approaching, demonstrating that some points of the surface will come into contact first (nucleation points). (b) Top view of a wetting area at a certain moment in time, the black dots represent the contact nucleation points and grey areas the contact area. The size of the grey area is a result of the area growth over time in the radial direction *r*.

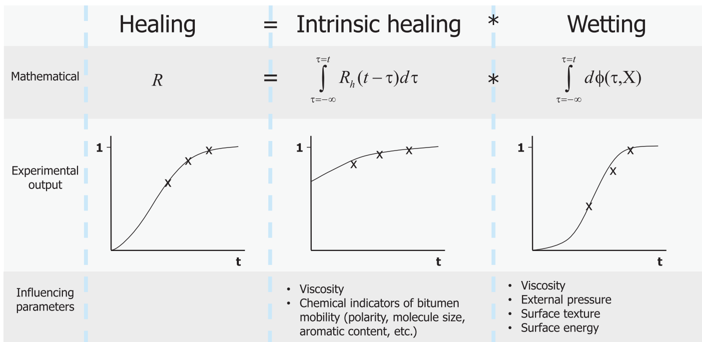

Figure 3. Graphical overview of different aspects of the two healing processes in the healing model.

3. Direct tensile test to asses healing

To quantify the relative importance of the processes that play a role in healing, as described in the previous section, a novel test method has been developed. The damage in asphalt concrete is assumed to be a (micro) discontinuity in the material. The design of the test method is aimed to investigate the most extreme version of a discontinuity; two separate pieces of bitumen. In the test, two pieces of bitumen are brought together and after a period of healing pulled apart again, testing the amount of tensile strength that has built up during the healing period. The global set-up of the test method is shown in Figure 4 and will be described in more detail below.

Bitumen samples with a specific geometry are produced for the test using silicon moulds. The bitumen is poured into a small stainless steel ring with an external diameter of 8 mm; this ring enables handling and fixation during preparation and testing. The size and design of the bitumen samples including the metal ring are shown in the left image of Figure 4.

Two bitumen samples are brought together under controlled conditions and are left to heal for a specific amount of time under a constant temperature, while controlling the boundary conditions during the healing period. The contact area between the two pieces is controlled by a piece of silicon paper with a small hole of Ø 5.5 mm, which is placed in between the two bitumen samples. As can be seen from Figure 4 the samples have a small bulge on top with a diameter of 5 mm, the silicon paper is placed around this bulge. As a result the paper will not hinder the surfaces to come into contact, but will only control the amount of contact area when the bitumen deforms. After healing, the specimens are tested in *direct tension* using a dynamic shear rheometer (DSR) equipped with a temperature chamber and a normal force load cell (Anton Paar, EC Twist 502). In order to obtain a healing ratio, the virgin strength of the material has to be known, therefore

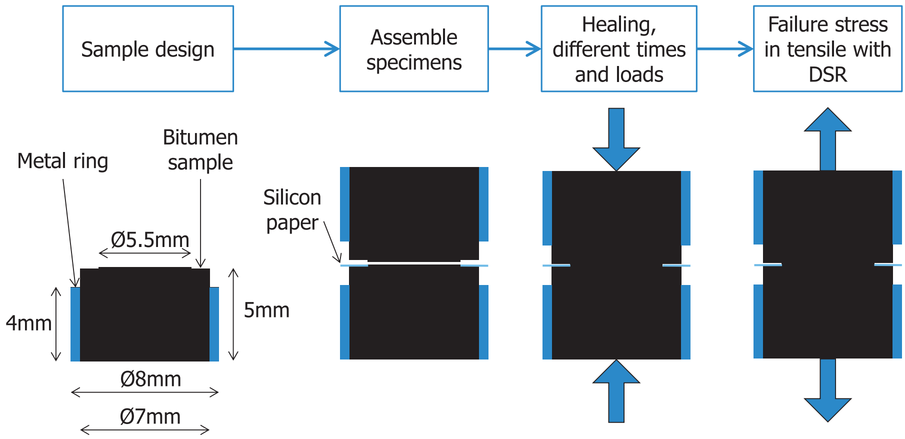

Figure 4. Schematic impression of sample design, preparation and testing.

reference samples are also produced. Due to practical limitations the geometry of the reference samples is slightly different from the assembled samples. The reference samples have the shape of a column with a length 12 mm and a diameter of 6 mm. These differences in geometry are addressed by calculating strength in terms of values per mm^2.

4. Test programme and results

4.1. *Sample preparation*

The bitumen and the mould with the metal rings in place are heated for 1 h at 165°C; this thorough heating is required as the bitumen needs to be very fluid during casting to create specimens with a very smooth surface without any air bubbles. Next, the bitumen is poured in the mould. The bitumen samples are placed upside down in the mould. The samples are left to cool for ½ hour at room temperature, followed by 1 h in the freezer at −24°C. After cooling the excess bitumen is removed from the top of mould using a heated knife, as a result the bitumen exactly fits the ring and the geometry shown in Figure 4 is realised. The sample is placed at −24°C until demoulding pre-conditioning. Demoulding is done under controlled temperature conditions (14°C ± 1°C). After demoulding, the outside of metal rings is cleaned using methylene chloride to improve handling during healing and testing. Test results showed that the stiffness of the samples significantly increased during the first 48 h after preparation, therefore the samples are kept in storage for 48–96 h at (14°C ± 1°C) before assembly and testing.

4.2. *Assembly, healing and loading*

4.2.1. *Two types of healing conditions*

Assembly is performed using two different methods in order to vary the loading conditions during healing. A part of the samples is assembled and left to heal in the storage room, during healing a small weight was used to create a constant normal force perpendicular to the interface. This assembly method is referred to as “Constant load”. A second part of the samples was assembled inside the DSR. These samples were pushed together with a constant displacement speed until a specified load level was reached, from this moment on the displacement is fixed. This assembly method is referred to as “Fixed displacement”. More details on both assembly methods will be presented below.

4.2.2. *Constant load*

Samples that are subjected to a constant load during healing are assembled and stored for healing in a temperature-controlled room at 14°C ± 1°C. In order to ensure alignment during assembly and healing these samples are assembled in small metal storage containers. After assembly a small weight is placed on top of the assembled specimen. The healing time is measured from the moment that the weight is placed until the moment that the weight is removed.

After removing the weight, the sample is placed in a storage room at 0°C ± 2°C for maximum 24 h, until the moment of testing. The time to testing is limited in order to minimise any further healing during storage. At the moment of testing, the assembled specimen is taken out of the storage container and fixed with clamps in the DSR. The sample is kept in the DSR for 15 min at 14°C to allow for temperature equilibrium in the sample and is subsequently tested by displacement-controlled tension, at a speed of 0.5%/s at 14°C ± 0.2°C.

4.2.3. *Fixed displacement*

The samples that are assembled inside the DSR are placed in top and bottom clamp of the DSR. Before assembly the temperature of the DSR is kept at 14°C for 15 min to allow for temperature equilibrium in the samples. The DSR brings the samples together using a displacement speed of 50 μm/s until a pre-defined load level is achieved. From this moment the displacement is fixed, while the specimen stays inside the DSR at 14°C ± 0.2°C. The stress present as a consequence of the assembly method relaxes quickly due to the visco-elastic nature of the material. After a specified healing period has passed, the samples are tested by displacement-controlled tension, at a speed of 0.5%/s at 14°C ± 0.2°C.

4.3. ***Materials and test programme***

4.3.1. *Materials*

The tested samples were produced using two grades of a straight run bitumen from the same crude source, having a pengrade 40/60 and 70/100.

4.3.2. *Test variations*

The samples made with pengrade 40/60 were subject to three different healing regimes; healing under a constant load of 0.001 N/mm^2, healing at a fixed displacement after being assembled at a normal force level of 0.042 N/mm^2 and healing at a fixed displacement after being assembled at a normal force level of 0.210 N/mm^2 (five times higher as the other assembly load). It is important to note the large difference in loading level between the constantly loaded specimens and the specimens that were only loaded at assembly. Using these three different regimes, the specimens are subjected to various healing times starting from 10 s for the specimens healing at a fixed displacement to 1 week (168 h) for the samples healing at a constant load. As the observed increase in healing level is specifically large at short healing times, tested healing times are increasingly large. The samples produced with pengrade 70/100 bitumen were only healed at a constant load of 0.001 N/mm^2. An overview of the test conditions for each material can be found in Table 1. For both binders reference samples are produced and tested after 1 week of storage at 14°C ± 1°C.

5. Test results

The results are presented in the form of a healing ratio over time in Figure 5. Tests are repeated at least two times, the average of the test results is in presented in the graph. The repeatability

Table 1. Overview of the test series.

Bitumen type	Healing conditions	Load level (N/mm^2)	Healing times
40/60	Fixed displacement	0.042	10 s; 1 h; 12 h
40/60	Fixed displacement	0.210	10 s; 1 min; 24 h
40/60	Constant load	0.001	1 h; 1 week
70/100	Constant load	0.001	6 min; 1 h; 24 h; 1 week

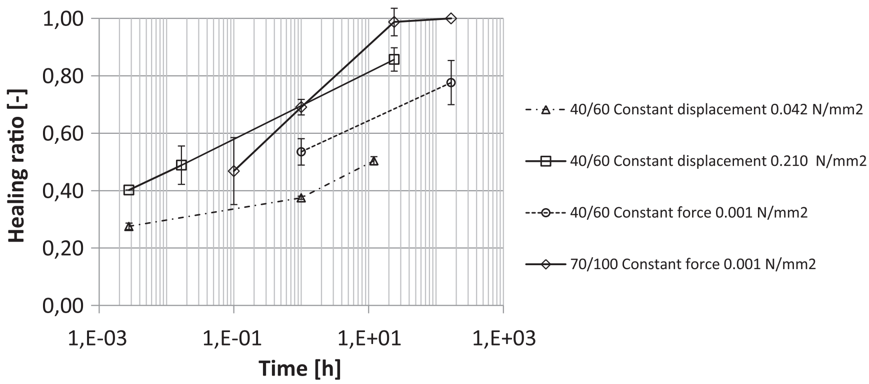

Figure 5. Healing ratios determined by testing tensile strength after healing under different conditions for a variation of healing times.

of the tests can be assessed from the graph by examining the error bars. The test samples that are assembled in the storage room show a reasonable repeatability. The test samples that are assembled inside the DSR show a good repeatability. This can be expected as the latter has less human interference.

The healing ratio is calculated by dividing the maximum tensile strength after a healing period by the strength of a reference sample made of the same bitumen. As the measured healing times span several decades, the log scale is selected for the horizontal time axis.

6. Discussion of results

Figure 5 shows that, the observed healing increases over time, under all applied healing conditions. The speed at which a material heals can be related to the slope of the curves. The softer 70/100 bitumen clearly heals faster compared to the 40/60 bitumen, demonstrating that this test method had discriminating power. Within the selected healing period, specimens made with 70/100 bitumen were even able to achieve full healing, which is not observed for the 40/60 bitumen.

When the two curves for the 40/60 bitumen under fixed displacement are compared, it can be seen that a higher normal force at assembly results in more healing at all time steps. For the shortest healing time of 10 s, the amount of diffusion that has taken place is expected to be very low (Wu et al., 2008), as a consequence all observed healing at this point in time can be attributed to the amount of wetted area and the adhesion of the bitumen. As the adhesion of bitumen is the same for both 40/60 samples, it can be concluded that the amount of wetted area increases with 50% when the assembly load is 5 times larger.

For healing times longer than 10 s the observed healing is the sum of wetting and intrinsic healing. As explained in Section 2.1., even if there is no external load, the contact area is expected to increase, as a consequence of the surface energies attracting areas neighbouring existing contact areas. Next to this, the load-bearing capacity of the area in contact will increase as molecules will diffuse over the interface. It is therefore expected, as also shown by the results, that the healing level will increase for specimens when the displacement is kept constant. It is however striking to observe the impact of a higher initial wetting level, on the long-term healing. After 12 h of healing the samples that were assembled using the higher force have regained 80% of their strength, while the samples assembled with the lower force level only achieved 50% of the initial strength, as such demonstrating the importance of wetting in the healing process.

If the healing levels of the series with fixed displacement are compared to the healing level of the series with constant normal force it can be seen that even a very low normal force level (around 50 times lower compared to the samples assembled inside the DSR) can result in a high level of healing, as long as this small load is applied for a long time.

The importance of the wetting process provides an indication of which material properties influence the healing performance of bitumen. In order to create the full molecular contact over a large area, the material at the surface has to deform very significantly. From this it can be concluded that the deformation capacity of bitumen is a crucial parameter in determining how much area can be wetted in a certain amount of time. The healing capacity of bitumen is consequently expected to increase if a bitumen is able to deform more under the same load. This conclusion matches Dutch empirical design rules, which indicate that bitumen with a higher pengrade is a better healer (Erkens, van Dommelen, van Vliet, & Leegwater, 2014). Future tests are planned to study the healing of different types of binders with the same pengrade, to assess in more detail the correlation between the healing ratio and rheological parameters.

7. Conclusions

The results presented in this paper show an increased healing level with longer healing times, which is in line with previous healing research. Additionally the paper demonstrates the impact of stress state at the damage during healing. Healing proceeds faster if the normal stress increases, showing 50% more wetting if the assembly stress is 5 times higher. This demonstrates the importance of wetting when assessing macroscopic healing. It has been shown that a small load applied for a long time can be as effective to realise healing, as a large load applied for a short time. This also indicates that the ability of a binder to viscously deform is an important parameter when assessing the healing performance of a binder.

One of the practical implications of the presented work is that the stress state in asphalt is very important for the amount of macroscopic healing that can be observed.

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