The importance of wetting in healing of bitumen

Leegwater, G. A.; Scarpas, A.; Erkens, S. M.J.G.

Publication date
2016

Document Version
Accepted author manuscript

Published in
Functional Pavement Design - Proceedings of the 4th Chinese-European Workshop on Functional Pavement Design, CEW 2016

Citation (APA)
Leegwater, G. A., Scarpas, A., & Erkens, S. M. J. G. (2016). The importance of wetting in healing of bitumen. In S. Erkens, X. Liu, K. Anupam, & T. Yiqiu (Eds.), *Functional Pavement Design - Proceedings of the 4th Chinese-European Workshop on Functional Pavement Design, CEW 2016* (pp. 489-498). CRC Press / Balkema - Taylor & Francis Group.

The importance of wetting in healing of bitumenG.A.

Leegwater, G.A.

Pavement Engineering, Delft University of Technology, TNO, Delft, Netherlands

A. Scarpas

Pavement Engineering, Delft University of Technology, Delft, The Netherlands

S.M.J.G. Erkens

Pavement Engineering, Delft University of Technology, Delft, The Netherlands & Rijkswaterstaat, Utrecht, The Netherlands

ABSTRACT: Asphalt concrete has the advantageous ability to heal autonomously, however the mechanisms behind this are not fully understood. To increase insight in the healing mechanism, the healing model used in polymer science is adopted. It interprets healing as the sum of wetting and intrinsic healing. The presented work introduces a new test set-up, which is designed to investigate the relative contribution of wetting and intrinsic healing by measuring the strength gain when two pieces of binder are brought into contact. Results obtained show that for a soft, pure binder, wetting is the dominant process. This can be concluded from the fact that at least 50% of the observed healing can be attributed to wetting. Wetting is highly dependent on both the load level and the duration of load application. Consequently, it is shown that the level of healing observed in asphalt concrete is very dependent on the boundary conditions.

1 INTRODUCTION

Asphalt concrete has the advantageous ability to heal autonomously. During rest periods damage present in the material is restored to a certain extent. This healing can be observed in the regain of stiffness and/or strength of the material after rest periods.

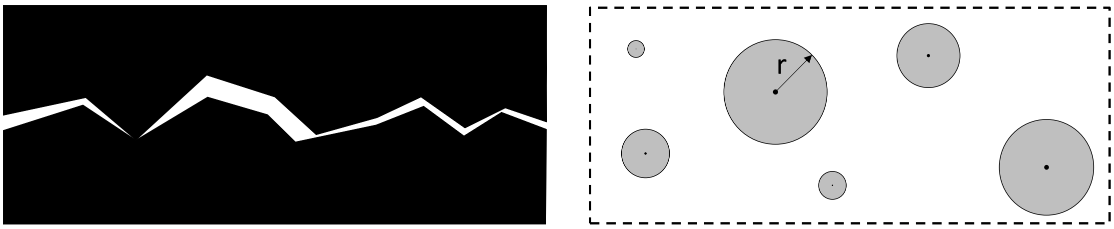
The fact that damage in asphalt concrete can be restored during rest periods has first been reported by Bazin and Saunier (Bazin and Saunier 1967). From this moment on the phenomenon is called healing and it is studied ever since e.g. (Bonnaure, Huibers, and Boonders 1982; Kim, Little, and Benson 1990; Bhasin, Palvadi, and Little 2011). It has been demonstrated that the level of healing increases with longer resting periods and higher healing temperatures. Next to this, it has been shown that healing requires a minimum level of force perpendicular to the damage and small damages show more healing compared to large macroscopic damages (Qiu 2012). However, research shows that the specific test method used has a strong influence of the observed healing. For instance, force controlled tests show more healing compared to displacement controlled tests (Francken 1998). Thorough attempts have been undertaken to explain the observed difference in healing between the different test methods, however no validated explanation has been found up until now. From this lack of insight it can be concluded that the mechanisms behind healing of asphalt are not fully understood, which impedes focused exploitation of this material property. The research presented in this paper attempts to unravel healing behavior in more detail.

There is a variety of approaches that can be chosen when studying healing. Healing is the inverse of damage, consequently the damage that is taken as the starting point of a healing test is an important factor when designing a healing test. The current approach in most pavement design guidelines is such that healing performance is an integral part of the fatigue performance. Therefore a large part of the reported healing research is executed on specimens loaded in fatigue, while introducing rest periods in different ways (Qiu 2012; Van den Bergh 2012). A complicating factor of this approach is that damage development during fatigue tests on asphalt is also not fully understood (Mangiafico et al. 2015). Consequently, if fatigue damage is taken as a starting point for healing tests, uncertainty is introduced by the unknown damage level. So, although measuring healing using fatigue tests has practical relevance, the undefined damage level impedes fundamental understanding of the healing processes.

In this study an attempt is made to measure healing using a very well defined level of damage, more specifically the damage is a physical discontinuity of known shape and size. A test set-up is designed to measure the strength gain when two separate pieces of bitumen are brought into contact under controlled conditions. Bommavaram et al. have already tried this approach in the past (Bommavaram, Bhasin, and Little 2009). In their test set-up pieces of bitumen where brought together in the DSR and healing was measured by testing the gain of shear stiffness versus time. This research has led to valuable insights, however real insight into the mechanisms was not obtained. Complicating factors with their test set-up were the complicated stress state and the lack of information on strength gain.

When interpretation the results obtained with the new designed test method, the model for healing of polymers proposed by (Wool and O' Connor 1981) is used to gain more insight in the healing mechanisms. In this model two processes are discerned, first surfaces have to come into contact, next the areas in contact have a certain capacity to transfer loads. The model has already been adopted by asphalt researchers in the past to try and explain the mechanisms behind healing (Kim, Little, and Benson 1990; Little and Bhasin 2007).

In the model, the first part of the process, where two surfaces come in contact, is referred to as wetting. Real contact between surfaces develops at a very small scale, at this level all material surfaces show a certain level of roughness. As two surfaces approach there will be an increasing number of points between them, the surface area in contact at each of these contact points will grow over time, as shown in Figure 1. As a result, the development of contact area between two surfaces is a three dimensional process. Influencing factors of wetting are; the external load that brings the surfaces together; the geometry (roughness of the surface); the viscoelastic properties of the material and the adhesion of the material (Johnson, Kendall, and Roberts 1971; Kendall 2001). Of these four factors, for bitumen three are influenced by temperature; the geometry, the viscoelastic properties and the adhesion.



a. b.

Figure 1 a. Side view of immerging contact points between two surfaces approaching. b. Top view of increasing contact area; area in contact is shown in grey, nucleating from the center point and growing radially indicated by the arrow (r).

The second process in recognized by the proposed healing model, is the ability of surfaces in contact to carry loads. Bitumen is sticky and will adhere to other material or itself (Kendall 2001). This initial adhesion, that is present from the moment the surfaces touch, is named the instantaneous intrinsic healing. Next to this there is also a time dependent part. In polymer science it has been shown that molecules can bridge over crack interfaces due to molecular motion (de Gennes 1971), this process results in the disappearance to the interface over time. This process is illustrated in Figure 2. Although bitumen molecules are very diverse and distinctly differ from polymers, it is assumed that some kind of reptation or self-diffusion in bitumen will also bridge crack interfaces over time (Kim, Little, and Benson 1990). Influencing factors of intrinsic healing are chemistry of the bitumen molecules (polarity, molecular weight, molecular structure). Consequently it is assumed that intrinsic healing is a material property that is constant if the temperature remains constant.

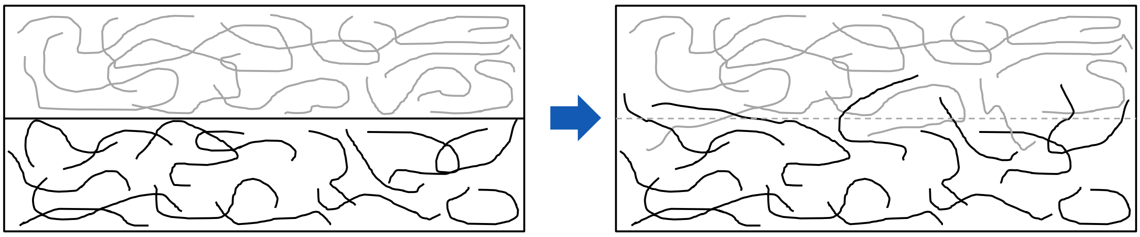


Figure 2. Two pieces of bitumen will adhere to each other as a consequence of the sticky nature of bitumen. Over time, due to molecular motion bitumen molecules will “cross” the interface of two surfaces in contact, resulting in a gradual disappearance of the interface.

The described model implies that damaged specimens can fully heal, if all damaged surfaces are brought into full contact and the contact areas are given enough time to fully homogenize.

In this paper a new test method is presented that investigates the material response when two bitumen surfaces are brought into contact. First, the new test method is presented, after this the results of two test series run with this set-up are presented. Based on the results the relative importance of both the wetting and intrinsic healing are discussed. The paper ends with conclusions and an outlook for further research.

2 TEST METHOD

2.1 *Design of the test*

To quantify the relative importance of the processes that play a role in healing, 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, are pulled apart. The measured tensile strength at failure is used to assess the healing performance. The global set-up of the test method is shown in Figure 3.

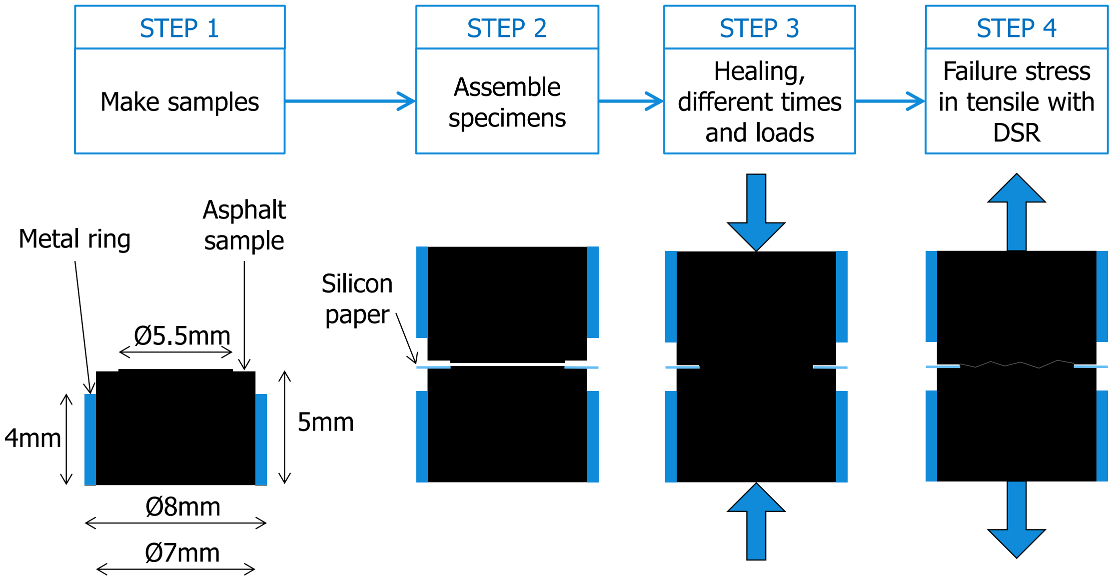


Figure 3. Schematic impression of steps in the test set-up.

Special bitumen samples were designed, introducing a small stainless steel ring around the bitumen, to allow for handling and fixation during preparation and testing. The geometry of a single test sample is shown in Figure 3 on the left. The bitumen sample has a specific shape to control the amount of contact area during healing. Silicon molds were fabricated to create the desired shape, the molds and metal rings are shown in Figure 4. During assembly a round piece of silicon paper with a small hole of Ø5.5mm is introduced in between of the two samples, to fix the contact area during the test.

The assembled specimens are left to heal for a specific time period, while controlling the temperature and the force perpendicular to the contact area. At the end of the healing period, the specimens are tested in direct tension using a DSR equipped with a temperature chamber and a load cell that is able to measure normal force (Anton Paar, EC Twist 502).

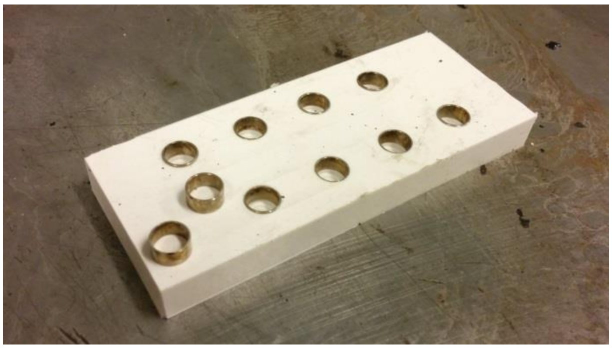


Figure 4. Silicon mold, with stainless steel rings in place during preparation of bitumen specimens.

A parameter that can be used to evaluate the level of healing is the healing ratio, this ratio compares the strength of a healing specimen to the strength of an intact specimen. In order to be able to calculate the healing ratio a reference test has to be executed on an uniform piece of bitumen. As it is difficult to reproduce the exact geometry of the assembled samples, the in Delft more frequently used mortar column geometry was selected for the reference samples. These samples use the same stainless steel rings. The geometry is given in Figure 5.

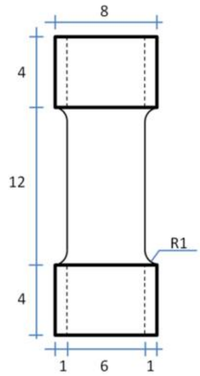


Figure 5. Geometry of the samples used for the reference tests.

2.2 *Materials*

All samples tested were made of a pengrade 70/100 bitumen from Kuwait Petroleum (Q8). Bitumen with a pengrade of 70/100 is relatively soft. This type of bitumen is regularly used for road construction in The Netherlands and consequently also for research purposes. Dutch design guidelines specify that asphalt concrete made with bitumen of pengrade 70/100 has good healing properties.

2.3 *Sample preparation*

The bitumen and the mold with the metal rings in place are both heated for one hour at 165°C, after which the bitumen is poured in the mold. The mold containing the bitumen samples is left to cool for 30 minutes at room temperature and subsequently for 1 hour at -24°C. In the mold the bottom side of the sample is facing up. After cooling the excess bitumen is removed, using a heated knife, making sure that the bitumen exactly fits the ring. The sample is left to cool again at -24°C until demolding. Demolding is done under controlled temperature conditions (14°C +/- 1°C). After demolding the stainless steel rings are cleaned using methylene chloride to improve handling during healing and testing. Tests have shown that the bitumen needs some time to stabilize after production in order to obtain more repeatable results, therefore the samples are stored in the temperature controlled room (14°C +/-1°C) for 48 hours, prior to assembly.

2.4 *Assembly, healing and test conditions*

Two different methods are used for assembly and testing. In the first assembly method, assembled specimens are placed in a temperature controlled storage room during healing. As such, large test series with various healing periods can be run, avoiding long processing times inside the DSR. In the second assembly method, specimens are assembled inside the DSR. This method allows for quick testing after assembly, making it possible to test very short healing times. To make the results of both test methods comparable, assembly and testing are done using the same temperature conditions. A more detailed description of both methods is given below.

In the first method, assembly is done in a temperature controlled room at 14°C +/-1°C. First the samples are taken out of the freezer (-24°C) and left for 1 hour to establish temperature equilibrium. The specimens are then assembled in small metal storage containers that ensure alignment during assembly and healing. The inside of the metal storage containers is covered with silicon paper to prevent sticking of bitumen to the container. First one bitumen sample is placed in the container, then a small piece of silicon paper, with a hole of 5,5 mm in diameter is placed on top, to fix the area in contact (see Figure 6a.). After this a second sample is placed upside down, on top of the first sample, as already shown in Figure 3. Finally, a small weight is placed on top of the assembled specimen (see Figure 6b.). The healing time is measured from the moment the weight is placed.

a. b.

Figure 6. a. A bitumen sample in a storage container, with the silicon paper fixing the contact area showing on top b. An assembled test specimen with a small weight placed on top.

The specimens are taken out of storage after the healing time has elapsed and are stored at -2°C +/- 2°C until the moment of testing, which is maximum 24 hours later. The specimens are loaded in tension while controlling the temperature at 10°C +/-0.2°C. The assembled specimen is taken out of the storage container and fixed in the DSR using clamps (Figure 7). The temperature is kept for 10 minutes at 10°C to allow for temperature equilibrium in the sample. The test is displacement controlled, applying a strain rate of 0.5%/s.

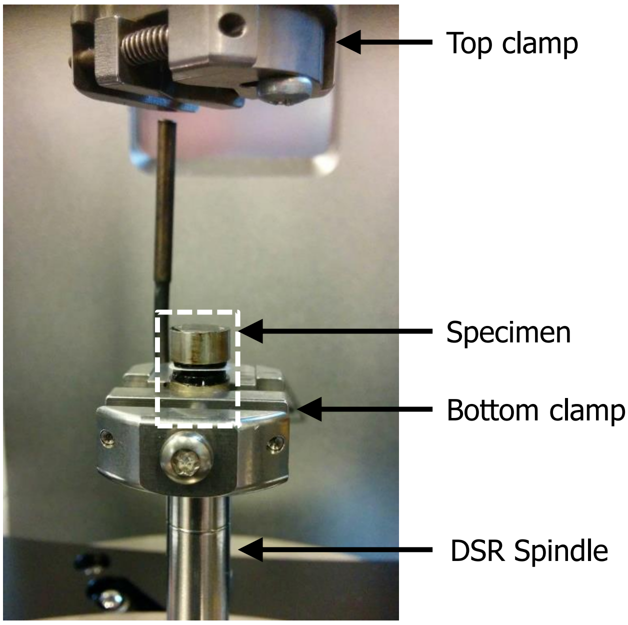


Figure 7. Specimen that has been assembled in the storage room, mounted for testing in the DSR.

In the second assembly method, the DSR is used to bring the two pieces of bitumen together. This means the healing occurs inside the temperature chamber of the DSR. First the samples are taken out of the freezer (-24°C) and stored at -2 °C +/-2°C. One sample is mounted in the bottom clamp of the DSR and one in the top clamp. The clamps with the specimens are placed in the climate chamber of the DSR, and the temperature is brought to 14°C +/-0.2°C. This temperature is maintained for 10 minutes to gain temperature equilibrium in the specimen. This temperature equilibrium time can be short as the samples are very small. Specimens are assembled by moving the two clamps of the DSR towards each other with a speed of 0.01 mm/s, until a certain force level is realized. After reaching the desired contact level, the displacement is kept constant. Due to relaxation the force reduces to 0 N within a time span of seconds, the exact relaxation time depends on the magnitude of the force.

In order to break the specimens, the temperature is changed to 10°C +/-0.2°C. It takes 16 minutes for the temperature to reach 10°C, including 10 minutes equilibrium time. Consequently the specimens are subjected to a healing time of 16 minutes. The tensile test is displacement controlled applying a strain rate of 0.5%/s.

2.5 *Test program*

The test program consists of two parts. The first part investigates the amount of observed healing over time, while maintaining a low constant normal force (0.015N), healing times are; 0.1 hour,1 hour, 24 hours and 1 week. A reference sample is part of the test series. The reference sample is a single column of bitumen, produced and stored under the same conditions. The reference is tested at the age of 1 week. The second part of the test program is designed to investigate the impact of assembly force on the measured healing. Three levels of assembly force were applied to the samples, 0.05N 0.5N and 1.0N. The specimens are loaded until failure just after assembly resulting in a short healing time of 16 minutes.

3 RESULTS

The results of the tests are presented in this paragraph, they will be discussed in the next paragraph.

In Figure 8 the results are presented from tests done on specimens that have been assembled in the temperature room and have been allowed to heal for various periods of time. This graph also presents the strength of the reference samples at the age of one week. The tests on the assembled samples have been run three times and variation in the results is expressed in the graph by error bars showing the standard deviation. Two of three samples tested at 168h fell and broke, consequently this value is based on a single measurement. It can be seen that up until a healing time of 24 hours the tensile strength increases significantly, after this the measured tensile strength does not increase. The observed tensile strength after a healing period of 6 minutes (0.1 hour) is around 50% of the strength observed after 24 hours and 168 hours. It can also be noted that the strength of the reference sample is less compared to the healed specimen.

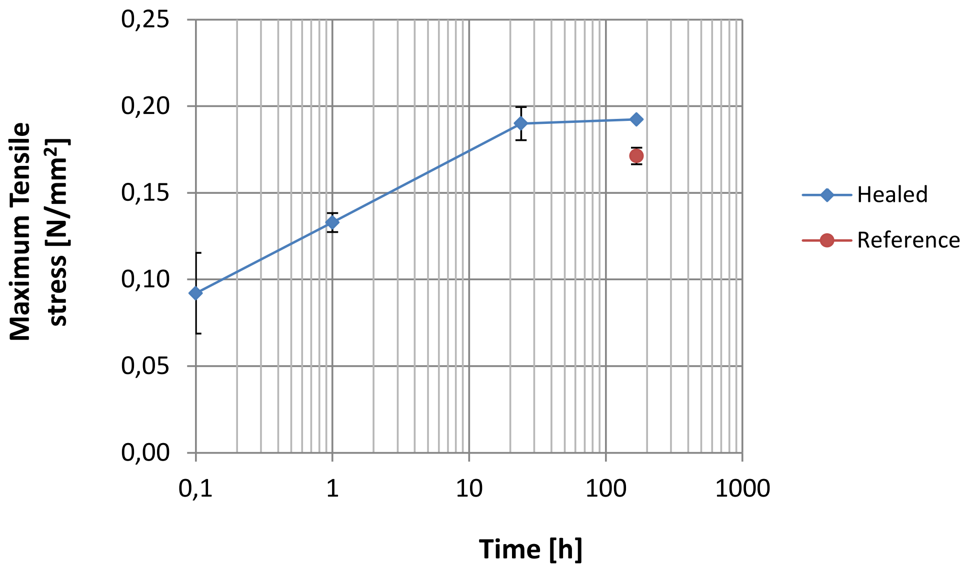


Figure 8. The development of tensile strength versus healing time.

Figure 9 presents the healing strength observed for the samples assembled in the DSR, while varying assembly force. It can be seen in the graph that, as the assembly force increases, the tensile strength of the assembled specimen increases. The tests have been run twice and variation in the results is expressed in the graph. The variation in the test results of assembly at 0.05N and 0.5N is so limited, that the error bars have become invisible. The tensile strength at 1N does show some variation, it is unclear if this is caused by an artefact during the measurement or if this variation is inherent to the measurement.

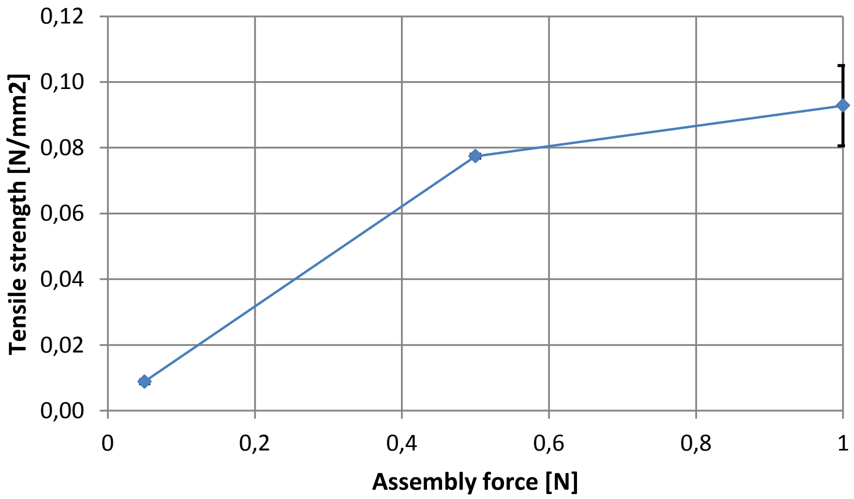


Figure 9. The development of the tensile strength versus the assembly force.

In order to assess the test results from an energy perspective, the amount of work required to assemble and break the specimens is shown in Figure 10. Work is plotted on a logarithmic scale versus the assembly force. Again the observed variation in the results is limited in case when the assembly load is 0.05 N and 0.5N, however for assembly at 1 N the variation is larger.

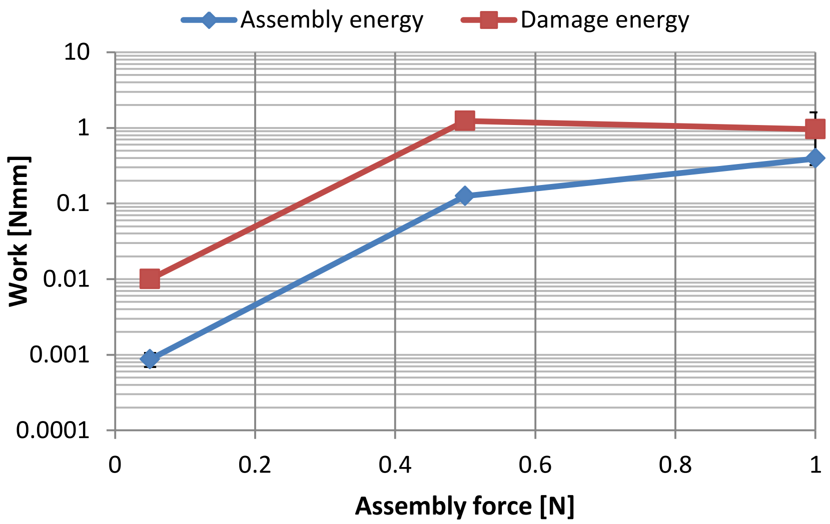


Figure 10. The amount of work required for assembly and breaking, versus the assembly force.

4 DISCUSSION OF RESULTS

4.1 *Speed of healing and the healing ratio*

As expected, Figure 8 shows an increase in tensile strength for longer healing times. However, after 24 hours a plateau value is reached. This plateau value can be interpreted as full healing, as it is in the same order of magnitude as the reference strength. Intuitively 24 hours seems fast for full healing, however one should realize that a 70/100 bitumen is a soft binder and the pure binder is expected to heal much faster compared to asphalt concrete.

The slight difference between the tensile strength at the plateau value compared to the strength, is most likely due to the difference in sample geometry. The reference strength can be used to calculate the healing ratio, giving the strength of a healed specimen relative to the original strength in a factor between 0 and 1. In this case the reference strength is lower than the ultimate strength found in the tests, as a healing ration of more than 1 is not possible a conversion factor is needed. If the strength after 24 hours is assumed to be full healing, this strength should correspond to a healing factor of 1, consequently a conversion factor of 0.9 is needed to calculate the healing factor from the measured tensile strength.

4.2 *The relative contribution of wetting*

A second observation from Figure 8 is the high level of tensile strength just after assembly at 0.1 hour (6 minutes), the healing ratio at this moment is already around 0.5. As explained in the introduction, healing can be seen as the sum of two processes, wetting and intrinsic healing. The intrinsic healing, in its turn consist of two parts, an instantaneous part (adhesion) and a part that develops over time. After only 6 minutes of healing, the contribution of time dependent processes governed by molecular motion are expected to be small.

Consequently, the healing observed after 6 minutes can largely be attributed to a combination of wetting and adhesion. Current results do not provide any quantitative information on the level of wetting after 6 minutes, however full wetting is the natural upper limit. From these two observations, it can be concluded that for the pure binder studied in this research at least 50% of the macroscopic observed healing can be attributed to wetting. Indicating that the realization of contact area is more important for healing, compared to the physical and chemical processes that take place once contact is established. To put this in terms of the presented healing model, for pure soft binders wetting is the dominant process.

4.3 *Impact of load level during assembly*

In the previous paragraph the importance of wetting has been demonstrated. According to the healing model, the process of wetting is influenced by the level of normal force, while the intrinsic healing is not. The impact of the healing conditions on the level of wetting is therefore further investigated by varying the level of normal force during assembly. From Figure 9 it can clearly be seen that, the observed tensile strength is higher when the applied assembly force is higher. Depending on the level of normal force, wetting can be very limited, resulting in hardly any healing, or wetting can be substantial, resulting in a healing ratio of roughly 0.5. This importance of normal force during healing, demonstrates the large influence of boundary conditions during healing of a bituminous specimen.

The importance of normal force has already been mentioned in research in the past (Bazin and Saunier 1967), however it has been seen as a binary condition, “for healing a small normal force is required”. The results presented here indicate that the relation between normal force and healing is proportional: a higher level of normal force will lead to more wetting and consequently to more healing. Logically there is an upper bound, which is the situation of full wetting, after this point additional normal force will not result in additional healing.

If the results of Figure 8 and Figure 9 are compared, it can be concluded that a constant, small load during healing is more effective compared to short, high load during assembly. The application of 0.5N for several seconds produces a similar healing as 0.015N (thirty times lower) for 6 minutes. It can be concluded that not only the load level is important for the amount of wetting that is realized, but also the period of time that this load level is present. This also implies that instant full wetting is physically impossible, however gained insights can be used to formulate an assembly protocol that is able to approach full wetting.

4.4 *Comparison of energy needed for assembly to energy needed for tensile failure*

In order to evaluate the amount of energy needed in making and breaking contact, the amount of work required for assembly and failure is plotted versus the assembly force (Figure 10). The results show that the work required to break the specimens is ten times larger compared to the work done during assembly. The fact that more energy is needed to break the specimens indicates that energy is gained during assembly. This observation matches the principles of surface energy, stating that energy is required to create surfaces and is gained when surfaces disappear.

In this experiment, the energy put into the assembly is most likely transferred into elastic and permanent deformation of the surfaces. These deformations are needed to bring the micro textured surfaces in real contact. The level of intrinsic healing is assumed to be constant in the experiment as a consequence of the conditions (fixed temperature and healing time), this implies that the difference in work required to separate surface is related to the level of wetting. The fact that the energy needed to break the well wetted specimens is 100 times higher, shows the importance of wetting for regaining resistance against fatigue loading.

The ratio of required work of 10 between assembly and break at 0.05 and 0.5 N could be interpreted as follows: the amount of energy that is invested into the creation of contact is proportional to the amount of wetting that is observed. The fact that this relationship is lost a higher load level might be explained by other sources of energy loss. It is likely that, at the highest load level, next to permanent deformation of the surface, energy is also lost in permanent deformation of the whole specimen.

5 CONSLUSIONS, RECOMMENDATIONS AND OUTLOOK

When using the presented healing model, wetting is the dominant process for the pure, soft binder studied in this research. This can be concluded from the fact that at very short healing times already 50% healing is observed.

The presented data also shows that the process of wetting for bitumen is highly dependent on the load perpendicular to the damage. Both the level and the duration of the load application have a large influence on the observed macroscopic healing, showing that a higher load level present for a longer time results in more healing. This implies that the boundary conditions during the rest period, are very important for the healing behavior of asphalt. This conclusion seems trivial, however much work when studying healing has been focused on varying time and temperature, while no attention was paid to the boundary conditions during healing. Only incidentally boundary conditions were varied in a very simplistic manner (vertical or horizontal storage). For future research in healing it is recommended to also address boundary conditions explicitly during healing.

The results presented in this paper are part of a bigger investigation into healing of bitumen and asphalt mortar. The processes that influence wetting are the next focus of the research program. Here the influence of bitumen properties like stiffness and phase angle will be studied and attempts will be undertaken to vary the surface texture of bitumen samples brought into contact.

REFERENCES

Bazin, P, and J Saunier. 1967. "Deformability, fatigue and healing properties of asphalt mixes." In *Intl Conf Struct Design Asphalt Pvmts*.

Bhasin, Amit, Sundeep Palvadi, and Dallas N Little. 2011. 'Influence of aging and temperature on intrinsic healing of asphalt binders', *Transportation Research Record: Journal of the Transportation Research Board*, 2207: 70-78.

Bommavaram, Ramamohan Reddy, Amit Bhasin, and Dallas N Little. 2009. 'Determining Intrinsic Healing Properties of Asphalt Binders: role of the dynamic shear rheometer ', *Transportation Research Record: Journal of the Transportation Research Board*, 2126: 47-54.

Bonnaure, FP, AHJJ Huibers, and A Boonders. 1982. 'A laboratory investigation of the influence of rest periods on the fatigue characteristics of bituminous mixes', *Journal of the Association of Asphalt Paving Technologists*, 51: 104-28.

de Gennes, Pierre-Giles. 1971. 'Reptation of a polymer chain in the presence of fixed obstacles', *The journal of chemical physics*, 55: 572.

Francken, Louis. (ed.)^(eds.). 1998. *RILEM Report 17: Bitumious Binders and Mixes* (London).

Johnson, KL, K Kendall, and AD Roberts. 1971. "Surface energy and the contact of elastic solids." In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, 301-13. The Royal Society.

Kendall, Kevin. 2001. *Molecular adhesion and its applications: the sticky universe* (Springer).

Kim, Y Richard, Dallas N Little, and Fred C Benson. 1990. 'Chemical and mechenical evaluation on healing mechanism of asphalt concrete', *Journal of the Association of Asphalt Paving Technologists*, 59.

Little, Dallas N, and Amit Bhasin. 2007. 'Exploring Mechanism of Healing in Asphalt Mixtures and Quantifying its Impact', *Springer Series in Materials Science*, 100: 205.

Mangiafico, S, C Sauzéat, H Di Benedetto, S Pouget, F Olard, and L Planque. 2015. 'Quantification of biasing effects during fatigue tests on asphalt mixes: non-linearity, self-heating and thixotropy', *Road Materials and Pavement Design*, 16: 73-99.

Qiu, Jian. 2012. 'Self-Healing of Asphalt Mixtures ', Technical University Delft.

Van den Bergh, W. Van de Ven, MFC. 2012. 'The Influence of Ageing on the Fatigue and Healing Properties of Bituminous Mortars', *Procedia-Social and Behavioral Sciences*, 53: 256-65.

Wool, R.P., and K.M. O' Connor. 1981. 'A theory crack healing in polymers', *Journal of Applied Physics*, 52: 5953-63.