

# Towards Green Pavements with Novel Class of SBS Polymers for Enhanced Effectiveness in Bitumen and Pavement Performance

Erik J. Scholten<sup>1+</sup>, Willem Vonk<sup>2</sup>, and Jan Korenstra<sup>3</sup>

**Abstract:** The performance of a base course in a pavement construction can be improved substantially by modification at relatively high level with polymers of the styrene butadiene styrene (SBS) type. As base courses are generally produced with relatively hard binders, a new polymer had to be developed as standard SBS at high concentrations in hard bitumen is likely to lead to poor processing and compatibility. The new polymer design is based on an increased vinyl content of the poly-butadiene mid-block, which has the advantage of being smaller in size at the same molecular weight (lower viscosity hence easier processing) and enhanced reactivity facilitating compatibility. One of the perceived positive contributions of SBS modification is its ability to form reaction products with bitumen molecules (grafting) and a method had to be developed to demonstrate such reactions to take place. The new polymer structure, the development of the method to proof grafting reactions taking place, the testing of the new polymer in base course binders and asphalt mixes, and finally the finite element modeling based on the fundamental material characteristics are presented. These demonstrate that as a result of the significantly improved performance, the overall pavement thickness design can be reduced by as much as 40% thus gaining environmental and economical benefits.

**Key words:** *Fatigue; Finite element modelling; Pavement design; Polymer modified bitumen; SBS.*

## Introduction

Polymer Modified Bitumen (PMB) is now a well established product in the paving industry of which it is recognized and acknowledged that the improved performance warrants the spending of some extra money: the life cycle cost analysis is significantly positive as explained in Engineering Report 215 of the Asphalt Institute [1]. The use of PMBs is rather focused on application in wearing courses and to a much lesser extent in binder courses. Their use in base courses is besides some experimental work, very limited. There is, however, no reason to believe, that modification of the base course binder would not bring an advantage over unmodified base courses. Base courses above all provide the structural basis for an asphalt pavement and their stiffness moduli, and their thickness determine the lifetime of a pavement under certain traffic loads. Thickness and modulus are also the leading parameters in traditional pavement design methods. However, if one were to improve the performance of the base course by modifying the binder such that its resistance to damage ((fatigue) cracking) would be significantly better, the pavement thickness can potentially be reduced. This would allow environmental benefits in the sense of using fewer natural resources, but could even bring economical advantages.

There are a few characteristics of base courses: they are normally bound with relatively hard binders and are intended to prevent bottom up damage into the structure. Modification with polymers

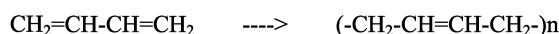
such as styrene butadiene styrene (SBS) renders optimum efficiency by adding at least 6% as the polymer absorbs up to 9 - 10 times its own weight of bituminous components and will at that concentration be the main volume fraction in the modified binder. Extended polymer continuity is the best remedy against crack propagation. However, modification with 6% greater of polymer in hard bitumen creates processing issues and certainly also compatibility issues. It was thus necessary to develop a new polymer structure that would allow such modification. In this paper the basic concept behind the new polymer structure is described and the anticipated mechanism behind the improved compatibility on the basis of grafting reactions is described. A method was developed to demonstrate the grafting to take place more readily with these new structures.

With the new polymer base course mixes were made and tested at the Road and Railroad Research Laboratory of Delft University of Technology. After initial testing a number of mixes were selected for fundamental testing to determine material input parameters for finite element modelling. This was done by the Structural Mechanics section of Delft University of Technology.

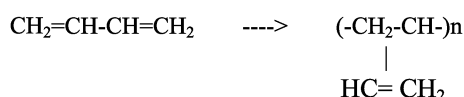
## Polymer Synthesis and Properties

When (1, 3) butadiene (unsaturated bonds at the 1<sup>st</sup> and 3<sup>rd</sup> position) is polymerised by using Butyl Lithium (BuLi) as the initiator, the resulting structure is mainly (1, 4) poly-butadiene and about 8-12% is the so-called (1, 2) poly-butadiene:

(1, 4) addition:



(1, 2) addition (high vinyl content):



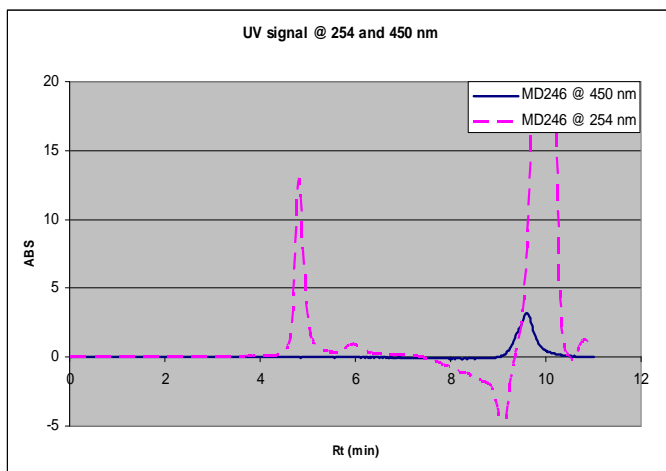
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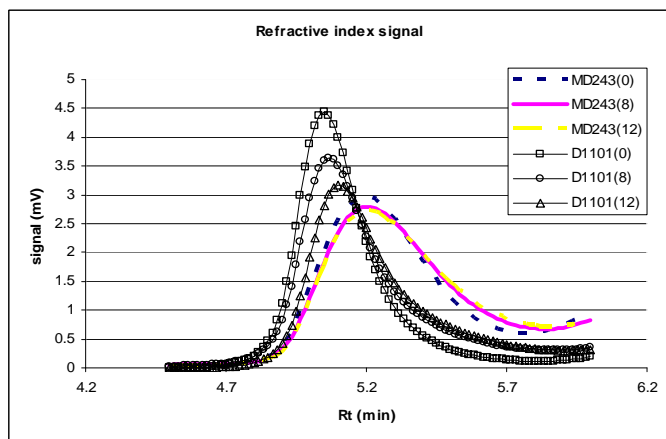
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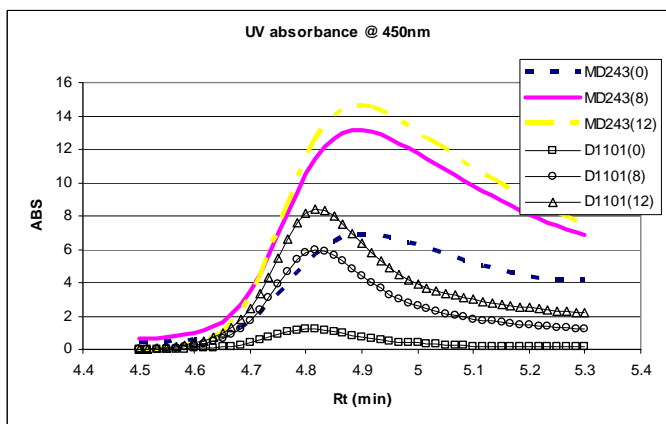
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**Fig. 1.** Absorbance of SBS Polymer after GPC Separation at 254 and 450nm.



**Fig. 2.** Blends with 5% Polymer before and after Treatment at 180°C (Times in Hours in Brackets): RI Detection.



**Fig. 3.** Blends with 5% Polymer before and after Treatment at 180°C (Times in Hours in Brackets): UV Detection.

By the use of special modifiers and processing conditions it is possible to increase the level of (1, 2) poly-butadiene up to 60-70%, which of course has some obvious consequences for the polymer itself. The change of the structure is such that part of the double bonds that are characteristic for Poly-butadiene, are no longer in the backbone of the polymer, but transferred to branches on the

backbone. As can be seen from the above scheme, at the same molecular weight, the polymer will be shorter. This will result in lower viscosities and better compatibility with bitumen. Another feature can also be deduced: if oxidation of the double bond will take place, the backbone will be attacked in case of the (1, 4) addition, while in case of the (1, 2) addition, the double bond in the branch will be attacked and will thus leave the backbone intact.

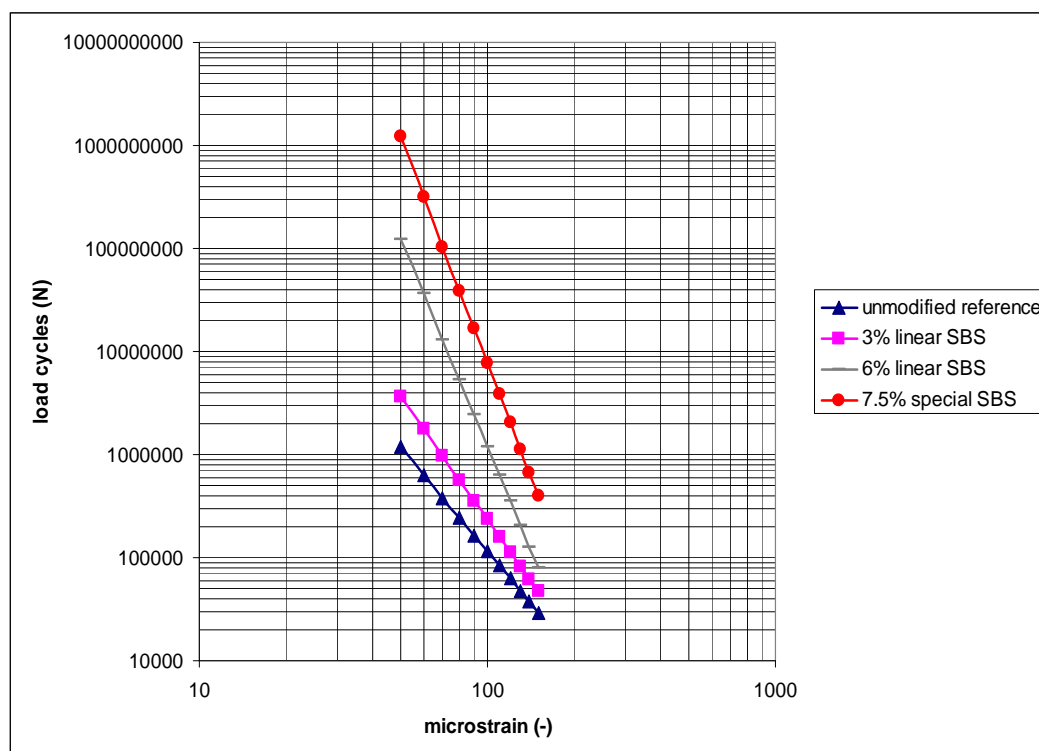
A further difference that was perceived on theoretical grounds was the reactivity of the high vinyl polymers with respect to the reaction with bitumen constituents. Although the polymers are already more compatible on the basis of their reduced size, it has always been postulated that reacting the polymer to bitumen molecules should further improve the compatibility between the bitumen and the polymer. In the next section, the scientific evidence that this is actually taking place will be given.

### Grafting the Polymer on the Bitumen

The thermal stability of bitumen/polymer systems is often studied with the use of Gel Permeation Chromatography (GPC). Using this technique, the binder is dissolved and taken over a column and the molecules are separated on their molecular weight. The polymers used in bitumen modification are much higher in molecular weight than any bitumen component and thus easily distinguished from the bitumen peak. The solvent with these separated molecules is then flowing through a detector, which can be equally done with refractive index (RI) changes or ultraviolet (UV)-detection, which leads to the same graphs, provided that a wavelength is chosen at which the polymer absorbs the radiation. A typical example is given in Fig. 1. The purple line represents the normal detection curve for PMB. When UV radiation of a higher wavelength is used (in this case 450 nm, blue line), the absorption is absent at the position where the polymer passes the detector, while the absorption of the bitumen is reduced and shifted to the left, at which position the higher molecular weight bitumen species (the asphaltenes) will pass the detector.

What the industry looks at in case of PMB ageing, is that particular peak at about 5 mins retention time, which in the case of SBS modification is said to be indicative for the 'breakdown' of the polymer. However, in spite of this phenomenon, which is reflected by a decline and broadening of the peak, the performance remains at high level, both in empirical testing and in more fundamental testing (dynamic mechanical analysis (DMA) for instance). For which as a supposed mechanism the grafting of polymer on bitumen molecules has been provided by Vonk et al. [2]. Using the technique of GPC in combination with UV detection at 450 nm offers an opportunity to demonstrate the validity of this supposition.

Blends of bitumen and 5% of SBS have been subjected to (stirred) storage at 180°C and after various intervals samples were taken and subjected to the GPC and two different detection methods: one with RI and one with UV at 450 nm. In Fig. 2 the standard method is shown with a standard SBS (in this case Kraton D-1101) and a new grade which has considerably higher vinyl content. As can be seen, both show a decline and a broadening of the peak at about 5 mins retention time, although the decline is minimal with the high vinyl grade. Taking the same stream coming from the GPC and now guided



**Fig. 4.** Four Point Bending Fatigue Results (Strain Controlled) at 20°C for Stone Asphalt Concrete (STAC) Mixes with Different Binders.

through the UV detector, one can observe an increase of the absorption 4.8 - 5 mins retention time (Fig. 3). So, while one detection method indicates that polymer seems to be deteriorating during hot storage, the other detection method reveals that (part of) the polymer reacts with the bitumen to form grafts. And these grafts will not only have a compatibilising effect on the PMB, but will also enhance the effectiveness of the modification.

Another conclusion that can readily be drawn is that the rate at which the grafting takes place is considerably higher for the high vinyl grade used, which in the example given is a new grade called MD243. It is clear that also standard SBS grades lead to grafting, but the higher rate obtained with the high vinyl grade allows much better and complete compatibilisation after a simple heat reaction in the storage tank with even the most difficult bitumen in terms of compatibility. Furthermore, this polymer leads to such low viscosities that master-batch pre-blends can be made up to high concentrations (>18%). Another grade, which has been successfully applied for a couple of years now, is Kraton D-1192, which was the first high vinyl grade to be introduced into the market.

### Special Opportunities for High Vinyl Grades

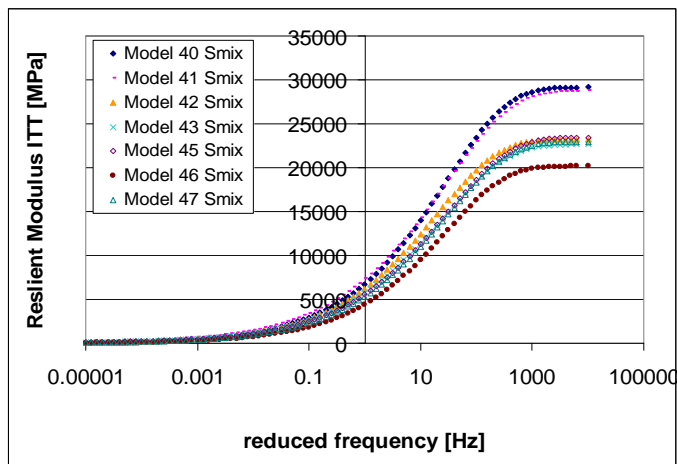
Although the high vinyl polymers can be used in standard applications that require better compatibility, better polymer stability, and low viscosity, the latest developed grades, among which is MD243, open an interesting list of opportunities that were difficult to achieve beforehand. One particular example for that is the use of highly modified base course.

The main task of the base course is to spread the loads coming from the wheels/axles, in order to reduce the deflections, by which the fatigue life can be improved. In other words, in base courses one

is seeking a relatively high stiffness, which, next to the mix design, is achieved by choosing relatively hard bitumen. These asphalt mixes result in a certain fatigue resistance, which then determines how thick the pavement will be designed in order to get a certain minimum lifetime. This means that roads carrying a high traffic load have relatively thick base courses. However, if one were to produce a base course with a binder that has at the same stiffness at much better fatigue resistance, the layer thickness can be reduced, which will save natural resources, and, if the improvement and thus the layer thickness reduction is significant enough, to a cost saving up front.

One well known method of improving asphalt fatigue resistance is the modification with SBS. This polymer has a strong interaction with bitumen and absorbs up to 10 times its own weight/volume of oily bitumen components. So, instead of having bitumen modified with say 4% of SBS, it is better to define the modified binder as one that contains on a volume base about 35 - 40% of extended polymer. At this level of modification it is likely that the extended polymer is not yet continuously present in the binder: it will improve the rutting resistance dramatically and it will also improve the fatigue resistance substantially, but there will be a further, very significant improvement once the extended polymer occupies the continuous phase. In Fig. 4 are the results of a 4 point bending fatigue test (strain controlled, 20°C). It clearly shows the strong improvement in fatigue resistance that is associated with the continuous polymer rich phase as found in the formulations with 6% and 7.5% SBS. The cracks will be hampered by the continuous polymer rich phase.

This situation of high toughness in PMB is achieved at concentrations > 6% SBS and the combination of this high content and the relatively hard base bitumen to be used, creates an issue with compatibility and workability of the asphalt mix. With the newly



**Fig. 5.** Master Curves of the Resilient Modulus (Stiffness) at 20°C for a 11 Tested Mixtures (Linear-Log Scale) Based on Indirect Tension Test Frequency Sweeps.

developed high vinyl grades there is a unique possibility to overcome these problems.

Together with Delft University of Technology (Prof. dr. ir. A.A.A. Molenaar), Kraton Polymers developed a hard binder (~30pen) containing a sufficiently high polymer content with thus a very high toughness. This binder has been selected after testing of a range of different SBS modified binders. In the next paragraphs more information about the test programme will be provided.

## Asphalt Testing Programme

The asphalt mixture tests that have been done are:

- 4 point bending fatigue tests (strain controlled),
- resilient modulus tests, and
- uniaxial monotonic compression and tension tests, of which the results were used to feed the material models implemented in the finite element code CAPA 3D.

Some of the results of the 4 point bending test are in Fig. 4. The 4 point bending test was initially used to compare the fatigue performance of different SBS grades and to make a selection of products to be tested in the next phase.

With the stiffness and fatigue properties known it is possible to use these properties in the design methods based on a linear elastic multi-layer system. The problem with these traditional methods, however, is that they do not give information on how damage actually initiates and progresses in relation to the type of asphalt mixture used, the thickness of the pavement, and the number of load repetitions.

Subsequently more fundamental research was initiated to differentiate between the effects of polymer modification of the binder in a base course mixture.

As a reference a so-called stone asphalt concrete 0/22 (maximum aggregate size 22mm) mixture, with a 40pen binder was used. This reference mixture is called mixture 40 in the paper. In all cases the volumetric composition of the base course was kept constant, with a binder content of 4.6% (w/w) in the mixture and a voids content of the compacted mixture around 5%. Together with the reference mixture six SBS modifications were tested in the mixture testing

program. The mixtures with SBS modified binders are coded as mixture 41, 42, 43, 45, 46, and 47. First of all master curves for the stiffness were determined with the indirect tension test (ITT) to show the influence of the polymer modification on the stiffness. The results of the test at 20°C are given in Fig. 5.

Monotonic uni-axial tension and compression tests were carried out on the seven base course mixtures to provide input for the material modelling.

The monotonic uni-axial tests were done at 5, 20, and 40°C. At each temperature strain rates were varied from 0.001 to 1%/s in tension and from 0.01 to 5%/s in compression to fully recognize the influence of temperature and strain rate on the material properties. The materials show hardening response and softening response. These phases in the material behaviour need to be implemented in calculations to get a good indication of damage development in a real structure.

From these monotonic uni-axial tension and compression tests a number of parameters were calculated like strength, failure energy, E-moduli (tangent and secant), strain at peak, total strain, and Poisson's ratio. Comparison of these properties at different temperatures and strain rates gives a good indication of the characteristics of the materials relative to each other.

In order to compare the mixtures in a pavement structure it is necessary to compare them in a material model that covers the whole range in terms of temperature and strain rate. The material constitutive model, which is used to describe the response of the asphalt mixtures is based on the flow surface proposed originally by Desai [3, 4] and Desai et al. [5] and further developed by Scarpas et al. [6], Erkens [7], Liu [8], and Medani [9] resulting in the asphalt concrete response (ACRe) material model. More information about the model used, the simulation of hardening and softening behavior, and the parameter determination can be found in Molenaar et al. [10].

## Material Modelling and Simulation

### Definitions and Modelled Structures

In order to interpret the finite element modelling (FEM) simulation results, it is necessary to introduce the definition of damage. Damage is defined as the cumulative amount of plastic strain ( $\epsilon^p$ ) in the material. In mathematical terms, damage denoted as  $\xi$  can be defined as:

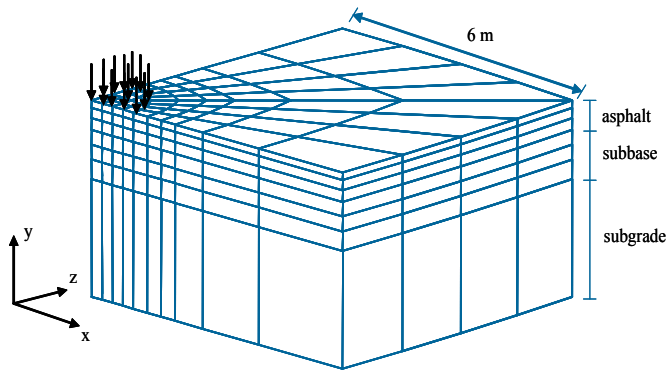
$$\xi = \sqrt{d\varepsilon_{ij}^p d\varepsilon_{ij}^p} = \sqrt{d\varepsilon_{ij}^p \cdot d\varepsilon_{ij}^p + d\varepsilon_{ij}^p \cdot d\varepsilon_{ij}^p + d\varepsilon_{ij}^p \cdot d\varepsilon_{ij}^p + d\varepsilon_{ij}^p \cdot d\varepsilon_{ij}^p + d\varepsilon_{ij}^p \cdot d\varepsilon_{ij}^p} \quad (1)$$

The total damage can be divided into volumetric and deviatoric (shear) components. The volumetric damage can further be subdivided into compressive volumetric deformation and tensile volumetric deformation.

This helps in understanding what kind of damage is developed. Compressive volumetric damage is associated with permanent deformation of the material (rutting). Tensile volumetric damage is

**Table 1.** Dimensions of the Modeled Structure.

Layer Thickness (m)	Young's Modulus (MPa)
Asphalt	Calculated in CAPA
Subbase 0.3	300
Subgrade 15	100



**Fig. 6.** Finite Element Mesh for Stationary Load Analyses.

associated with cracking. Deviatoric damage is the result of tensile-compressive states of stress and can lead to mode II associated cracking.

In this paper the results of the stationary dynamic loading simulation are reported. Also FEM analysis with moving wheel loading has been performed within the framework of this study. Results of the work with moving wheel loading can be found in Scholten et al. [11].

The pavement profile was assumed to consist of three material layers, see Fig. 6. The dimensions of the layers are summarized in Table 1. Both the subgrade and the base layer are assumed to behave linear elastically. The top-layer represents the asphalt layer. The temperature of the asphalt mixture is assumed to be 20°C. The modulus of the asphalt layer depends on the strain rate and is determined in an iterative way.

The dynamic analysis option of finite element code CAPA-3D is utilized to subject the model to a series of successive half-sinusoidal pulses each of 25ms duration and 0.8 MPa stress amplitude. The advantage of using dynamic analysis is that not only strain-rate effects but also mass effects can be considered. As a result of that,

the various phenomena of wave generation, propagation, reflection, etc. that occur in a pavement due to the dynamic nature of traffic loading can also be taken into account.

**FEM Results**

The non-polymer modified reference mixture (mix 40) has been compared with two polymer modified mixtures: mix 41 containing 6% standard SBS and mix 45 which contains 7.5% MD-243. A thicker (0.25 m) non-modified asphalt pavement has been compared with a thinner (0.15 m) modified pavement (containing either of the two SBS formulations 41 and 45).

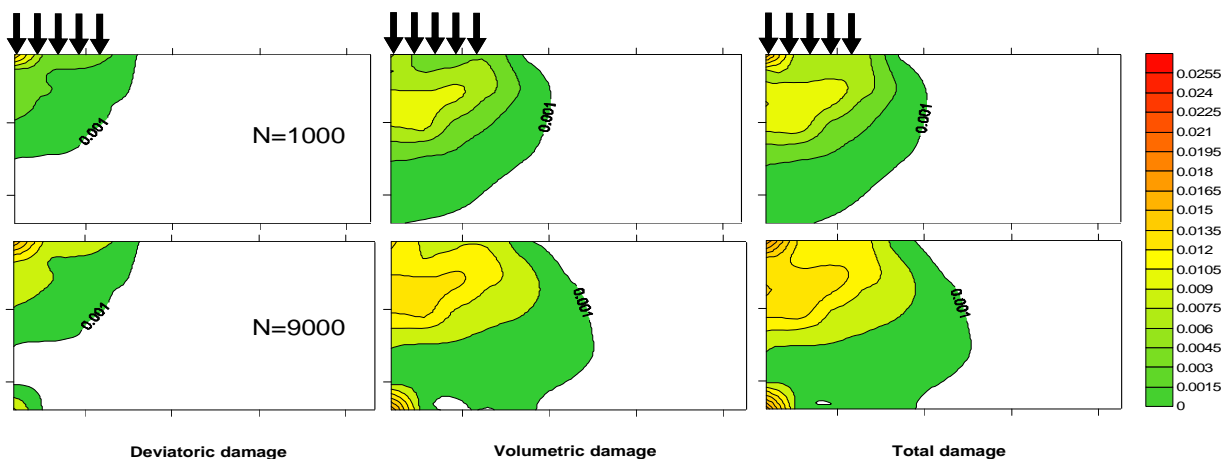
Fig. 7 shows the development and propagation of damage in the unmodified asphalt layer (mix 40) as the number of load cycles increased.

At the top of the asphalt layer of the non-modified pavement damage concentration can be identified in the vicinity of the wheel. The separate projections of volumetric and deviatoric damage show that this damage concentration has mainly a volumetric nature, which indicates the gradual development of permanent deformation (commonly classified as rutting). The damage starts directly under the wheel and propagates rather quickly into the body of the pavement.

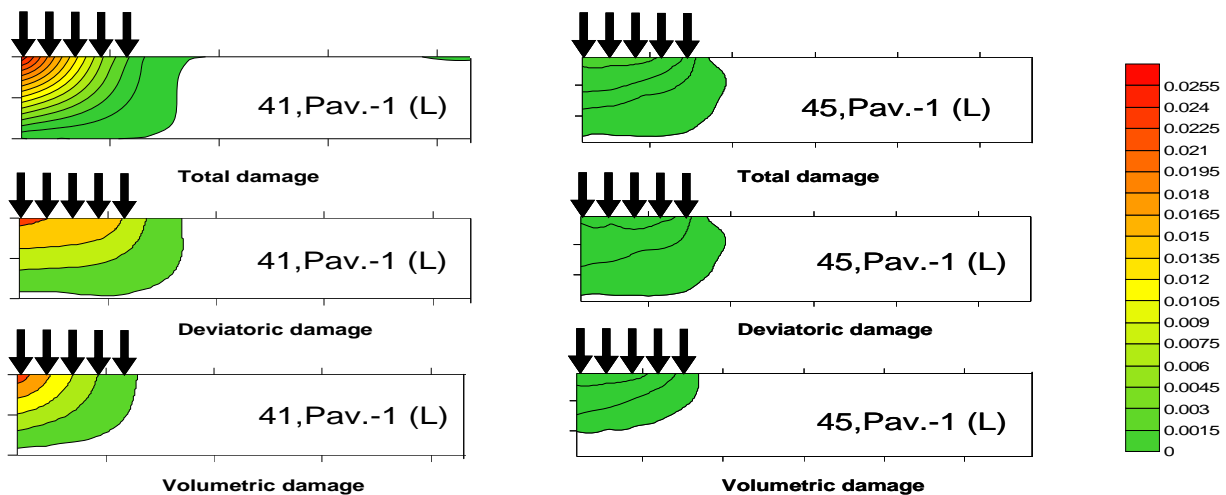
In the non-modified pavement intense tensile damage can be observed developing at the bottom of the layer directly under the wheel, see Fig. 7. This indicates the tendency of initiation and gradual development of tensile cracking at the bottom of the layer.

Fig. 8 shows the damage development for the two SBS modified mixes 41 and 45 in the thinner 0.15 m asphalt layer. The images clearly show that for the thin SBS modified pavements, damage initiates only at the top of the pavement and not at the bottom. At a 40% reduced layer thickness compared to the unmodified reference pavement there is a damage concentration at the top for mix 41 with standard SBS. However, the MD-243 based mix shows only very low levels of damage. This is a spectacular result given the 40% layer thickness reduction that has been applied. The damage occurs mainly in the area under the wheel. Lower in the structure there is hardly any effect from the load.

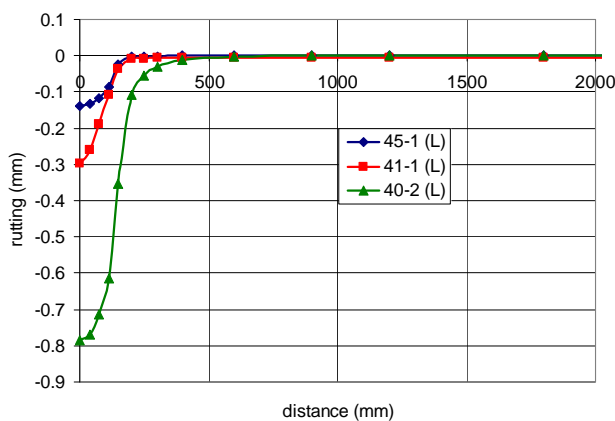
Fig. 9 shows the rutting profile after 9,000 cycles of the non-modified thicker pavement in comparison with the two SBS



**Fig. 7.** Damage Development after 1,000 and 9,000 Wheel Passes in 250mm Pavement Structure with Standard Unmodified Asphalt (Mix 40).



**Fig. 8.** Damage Development in a 150mm Pavement Structure; Mix 41 Contains Standard SBS, Mix 45 Contains 7.5% MD-243, and L for Linear Modelling of Subbase and Subgrade.



**Fig. 9.** Rutting Profile of 250 mm Non-Modified Asphalt Pavement (Mixture 40) and 150 mm Polymer Modified Asphalt Pavement (Mixture 41 with Standard SBS and Mixture 45 with 7.5% MD-243) after 9,000 Load Repetitions.

modified thinner pavements. The figure shows that the accumulated permanent deformation in the pavement with the non-modified mixture 40 is more than 5 times higher (0.8mm versus 0.14 mm) than the pavement with the MD-243. The standard SBS modified mix 41 also shows a big improvement compared to the unmodified mix, but it is outperformed by the mix MD-243 based mix.

As mentioned before, the non-modified asphalt contained a 40pen bitumen, while the best performing modified system was based on a newly developed low molecular weight polymer, MD-243.

It should be noted, however, that a thinner asphalt pavement could result in more damage to the subbase. Therefore the asphalt pavement design always needs to be checked on the subbase criterion.

Even though 9,000 load cycles is only a small part of the lifetime of a pavement, the results already show distinct differences between the mixes. Similar differences were found when applying moving wheel loading as reported in Scholten et al. [11].

As you can see there is a substantial thickness gain achieved and if the costs are calculated, it is demonstrated that the cost saving of this structure, in spite of the use of a full depth modified

construction will be about 25%.

The new polymer MD-243 is one that can be made fully compatible with almost every kind of bitumen. It does not take so much reaction time, but given the pay-back in terms of reduced costs up front, adopting a binder production scheme that would allow the proper binder manufacturing (standard blending equipment can simply be used) would certainly be cost effective. In the above structure the estimated material cost saving would be about 25% for every kilometer of road laid. Besides the cost savings, it would also imply less use of natural resources, which are only likely to go up in prices in the decades and moreover, scarce to get hold of.

### Conclusions

Kraton polymers have introduced a new class of styrenic block co-polymers with the elevated vinyl content that offers significant improvements over standard SBS.

- The molecules are shorter than standard SBS of equivalent mol weight and thus provide better compatibility and lower blend viscosity.
- These polymers react more readily with bitumen components thus accelerating the compatibilisation rate.
- Evidence has been generated that these reactions called grafting of bitumen and polymer molecules using GPC separation technique with UV detection at the unusual wavelength of 450nm.
- The new technology allows modification of hard bitumen normally employed in base courses at levels that a step change in fatigue resistance is obtained (compatibility and workability).
- The advanced mix tests and finite element modelling done at Delft University of Technology showed that the asphalt mix containing a binder with 7.5% MD-243 developed much less damage (accumulated plastic strain) than the unmodified reference or a mix with standard SBS. Also in resistance to permanent deformation the best results were achieved with 7.5% MD-243.
- The high stiffness modulus and the improved fatigue resistance then allow a thickness reduction that will make the material cost

per kilometer some 25% lower (depending on price fluctuations) and will lead to less consumption of natural resources. Thinner, SBS modified high performance base courses are a new solution to construct cost-effective environment-friendly pavements.

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