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Microstructural changes in bitumen at the onset of crack formation

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ABSTRACT

For the first time the initiation and early stages of damage and microcrack growth in bitumen under mechanical loading conditions have been observed on the micrometer length scale by atomic force microscopy (AFM). Bitumen films have been applied on flexible substrates. Tensile loading of the substrate leads to straining of the substrate itself as well as the bitumen film that has been attached to it. The surface microstructure of the bitumen film has been studied with AFM for three loading levels: no loading, moderate (5% strain) and high loading (10% strain) conditions. The initiation and onset of propagation of microcracks have been observed for the moderately loaded specimen. It is found that damage in the shape of crazes and cracks does only occur in one of the two microstructural phases of bitumen, i.e. in the ellipse-shaped domains. Subsequent application of higher loading levels leads to complete embrittlement and fragmentation of the elliptical domains into lamellar structures which tend to align and orient in a direction perpendicular to the direction of loading.

1. Introduction

Asphalt pavements degrade over time under cyclic mechanical loading by traffic passing over it. Next to the distress originating from these mechanical loading cycles, the degradation process of a pavement structure may accelerate due to environmental conditions, such as oxidation and ultraviolet radiation induced hardening of the bituminous binder [1,2]. Thus, structural damage in pavements in the form of cracks originates from mechanical loading of a structure that may have 'weakened' already by the aforementioned environmental causes (time dependent changes of mechanical properties). In an asphalt concrete composite, it is the bituminous binder that can be considered the mechanically weakest link. Within the bituminous binder, damage in the form of cracks is caused by the earlier mentioned combination of mechanical and environmental loads. These cracks may occur inside the bitumen itself (cohesive failure) or at the interface between the bitumen and aggregates (adhesive failure). In this study the focus is on the initiation and propagation of damage inside the bitumen leading to cohesive failure under mechanical loading.

Detailed knowledge about the crack initiation and growth mechanisms in bitumen would add to the understanding and prediction of the material's performance at the macroscopic scale. Numerous research efforts have been undertaken in order to understand the crack growth and fatigue thresholds of asphaltic composites. Experimental data that access the length scale of the pre-macro crack regime would improve the understanding of the bitumen degradation process [3,4], and guide eventually the

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development of material formulations with more beneficial damage characteristics. The first step towards this goal is to develop methods which allow for the experimental observation of surface deformation and fracture processes at the micro to nanometer length scales.

By now it has become an established fact that bitumen is a heterogeneous material at the micrometer scale, i.e. it exhibits a microstructure. Atomic force microscopy (AFM) is the characterization technique of choice for probing this microstructure. From previous AFM studies on bituminous materials several authors have reported the presence of a microstructure characterized by a two or three phase morphology for bitumen [5–10]. The microstructure morphology is usually found to consist of ellipse shaped domains. The isolated domains are found to be dispersed in another phase: the matrix or 'continuous' phase. The elliptical domains are found to possess an oscillating topographical pattern along the long axis, which is referred to as 'wrinkling pattern' [11]. This oscillating pattern gives the domains their typical appearance which is often referred to as 'bee'-structures [8,9,11–13]. Furthermore, the microstructural properties of bitumen are known to depend on the crude source and production process of the bitumen [14,15], i.e. the bitumen grade. The present study aims to observe and better understand the impact of mechanical loading on bitumen at the microstructural level by means of AFM. For various mechanical loading scenarios the manifestation of damage on the microstructural length scale is probed for the first time. The morphology, localization and other characteristics of crazes and early stages of (cohesive) microcracks in bitumen have so far not been reported in literature to any level of detail. Improved knowledge about the initiation and propagation of microcracks can help to 'design' bitumen with microstructure morphologies that are more damage resilient. Control of the bitumen microstructure may be obtained by applying optimized material (thermal) processing conditions or by using additives [14,16]. Finally, the results presented here, can be used to improve cohesive damage models that are utilized in finite element simulations of asphalt concrete's macroscopic mechanical response.

2. Materials and methods

For the research described here a bitumen of penetration grade 70/100 was selected. This type of bitumen is commonly used in the Netherlands as a binder in asphalt pavements. The wax and asphaltene fractions of this bitumen are found to be 3.3% and 9.7% respectively. The bitumen has a penetration of 86 (units of 0.1 mm at 25 °C), a softening point of 48 °C and a mass density of 1.031 g cm^{-3}. The concentration of metal atoms in the bitumen was obtained by instrumental neutron activation analysis, INAA [17]. For this particular bitumen the metal concentrations were found to be 192, 32 and 58 ppm for vanadium, iron and nickel respectively.

A Multimode-V, atomic force microscope from Bruker (Santa Barbara, USA) was used to characterize the microstructure of the 70/100 bitumen at its virgin (mechanically unloaded) state, and to probe the microstructural response after applying various levels of tensile loading. AFM measurements were performed by tapping mode in air, using commercially available silicon cantilevers (RTESPA, Bruker). These cantilevers have a nominal resonance frequency of 330 kHz and a force constant of 40 N/m. A tip scan rate of 1.0 Hz (1 Line/s) was used and the images were recorded at 30 × 30 μm scan size with a pixel resolution of 512 × 512. To probe the bitumen microstructure to more detail, images of 10 × 10 μm scan size were recorded with the same pixel resolution and scan rate.

Tapping mode is one of the dynamic modes of AFM, in which the cantilever is driven at its resonance frequency at a preset oscillation amplitude. During tapping the probe is in a state of intermittent contact with the sample surface, while an electronic feedback loop is used to maintain the preset oscillation amplitude by adjusting the relative position of sample surface to the cantilever. In this way the surface topography is recorded. Another parameter that is measured simultaneously with the topography is the phase shift of the oscillating cantilever, which is related to local mechanical properties of the sample surface. Tapping mode is the method of choice for imaging of bituminous materials because these materials are soft and sticky and intermittent contact prevents the tip sticking to the sample surface. In the case of imaging bituminous surfaces, the use of this mode also reduces the chance of damaging the sample and it minimizes the occurrence of imaging artifacts.

The bitumen specimen was prepared on the non-adhesive side of a piece of commercially available scotch tape (Fig. 1). First, a steel sample puck (12 mm diameter) was attached to the adhesive side of the tape. The sample puck is required to keep the sample firmly attached to the AFM stage. On the non-adhesive side of the tape, opposite to the puck, an amount of 20 mg of bitumen was applied. In order to obtain a thin and flat bitumen film, the steel sample puck was heated for about 30 s on a heater plate at 100 °C. This contact time was sufficient to obtain a smooth and shiny bitumen film on the tape. Next, the specimen was conditioned for an hour inside a convection oven at 100 °C. Subsequently it was cooled in air to room temperature (25 °C) and allowed to equilibrate for 24 h as shown in Fig. 2.

Immediately after this preparation procedure, the microstructure of the bitumen film was recorded at 25 °C. Then the tape was strained from both sides at ambient conditions until it was deformed up to a strain of 5%. This mechanical deformation scenario of the bitumen film will be termed as 'moderate tensile loading' throughout the text. It is obvious that the strain applied to the tape was (partially) transferred to the bitumen film (visual inspection showed that the initial circle shaped bitumen film became attained a more elliptical shape). The strained sample was then allowed to equilibrate at room temperature for 15 min (Fig. 2) and then its microstructure was imaged by AFM. The application of mechanical load introduced an increase in local stickiness. This resulted in occasional delay in probe snap-off during an AFM tapping cycle. An equilibration time of 15 min at 25 °C was found to mitigate this imaging problem.