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From virgin to recycled bitumen: A microstructural view

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In the present work, soft and hard bitumens recovered from unaged, aged and recycled asphalt concrete (AC) mixtures, which in laboratory tests performed mechanically as well as an AC mixture produced with virgin materials, were investigated regarding rheological, thermal and surface microstructural aspects. For comparison purposes, bitumen containing 50 wt% of virgin bitumen and 50 wt% of bitumen recovered from reclaimed asphalt pavement (RAP) was studied. Some properties of the bitumens remained unchanged throughout the preparation of the AC mixture, aging and recycling: Soft and hard bitumens retained their general rheological properties significantly, and their thermal and surface microstructural properties partially. Soft bitumens presented larger "bee" structures and, therefore, higher surface roughness, while hard bitumens presented smaller "bee" structures and, thus, lower surface roughness. Furthermore, soft bitumens seemed to contain higher crystalline-like content than hard bitumens. For the soft cases, the unaged recovered bitumen did not show the same characteristics (rheological and surface microstructure) as the virgin bitumen. Similarly the recovered recycled bitumen did not show the same characteristics (surface microstructure) as the bitumen prepared from the mixture of virgin bitumen and RAP bitumen. Aging of the AC mixture changed the rheological properties of the soft bitumen by increasing the complex modulus and decreasing the phase angle. Similarly, recycling changed the rheological properties by increasing the complex modulus and decreasing the phase angle. Compositional changes occurred during AC mixture preparation (possibly also aging and recycling) for both soft and hard bitumens. Consequently, more "phases" were observed on the surface microstructure for the recovered bitumens as compared with the virgin bitumens. However, no significant trend was found for the surface microstructure characteristics between the unaged, aged and recycled recovered bitumens. Moreover, the nature of the virgin bitumen influenced the properties of the recycled recovered bitumen, e.g. the glass transition temperature.

1. Introduction

Recycling of asphalt pavements is a clear step towards sustainable pavement construction, being increasingly used worldwide as an alternative for energy and cost savings [1]. However, for achieving high efficiency in the recycling process, the recycled asphalt must display mechanical properties that are similar or improved in comparison with the asphalt prepared with virgin materials. The recycled asphalt can be prepared by mixing reclaimed asphalt pavement (RAP), virgin bitumen, virgin mineral aggregates and rejuvenating agents. In principle, the RAP bitumen needs to be combined with suitable virgin bitumen (or rejuvenating agents) to create recycled bitumen with good adhesive and cohesive properties in order to avoid premature failure of the recycled asphalt pavement [2]. It is still not understood how the virgin bitumen mixes with the RAP bitumen but it is believed that the degree of blending between the RAP bitumen and the virgin bitumen plays a role on the final properties of the recycled asphalt. Therefore, investigations are ongoing in order to understand how and why the virgin bitumen mixes with the RAP bitumen, and which rheological and microstructural changes occur due to blending. Ideally, the link between the microscale and the macroscale properties of asphalt should be established. The degree of blending between virgin and RAP bitumen should depend on different factors such as temperature of mixing, mechanical energy used for mixing and the chemical characteristics of the virgin and RAP bitumens [3–6]. Moreover, the molecular interactions developed in the blend may give rise to particular nano- and microstructures, which may be unique to the degree of blending.

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Atomic force microscopy (AFM) measurements have shown that bitumen presents different structures on its surface upon annealing [7–13]. The microstructural details are related to the chemical composition of bitumen. The combination of dynamic shear rheometer (DSR) and AFM measurements has been used in an attempt to correlate the microstructural properties observed on the surface of annealed bitumen films, and the bulk rheological properties of bitumen prepared from blending virgin bitumen with RAP bitumen. Nahar et al. [14] used AFM for probing the degree of blending between the virgin bitumen and RAP bitumen by measuring the average surface microstructural changes occurring in the "blending zone". It was found that the blended bitumen presented properties, which fell in between those of the two individual bitumens. Also, the rheological properties showed that the complex modulus and the phase angle of the blended bitumen were situated in between those of the two individual bitumens. Similar results were reported by Nazzal et al. [15]. However, in this case the authors have also reported that RAP bitumen resulted in adverse effects on the adhesion properties.

The present work shows an original approach for studying the effects of aging and recycling on bitumen by analyzing the bitumen recovered from unaged, aged and recycled asphalt concrete (AC) mixtures. The bitumens were recovered from AC mixtures, which in laboratory tests performed mechanically as well as AC mixtures produced with virgin materials [16]. The recovered bitumens were studied in terms of rheological, thermal and surface microstructural properties and it was observed that some properties remained after the preparation of the AC mixture, aging and recycling, but other properties changed more or less significantly.

2. Materials and methods

2.1. Materials

The materials chosen for this study were those used in another research project on hot mix recycling funded by the Swiss Federal Roads Office allowing comparison of results at different scales [16–21]. The virgin (V) bitumens are referred to as V160/220, V70/100 and V10/20 and are standard bitumens used in Switzerland. These virgin bitumens were used in the preparation of asphalt concrete (AC) mixtures: V160/220 and V70/100 were used to prepare a Swiss standard surface course and V10/20 in a high modulus base course, with and without reclaimed asphalt pavement (RAP) material [16]. Therein, the recycled AC mixtures were produced by mixing 40% RAP, containing a combination of different pavement layers, with new mineral aggregates and virgin bitumen(s). Artificial aging of the AC mixtures was performed by using the AASHTO R30 method which consists of a short term oven aging (STOA) of the loose AC mixture, representing aging occurring until the mixture is placed in the pavement, and a long term oven aging (LTOA) of the compacted AC mixture representing 5–10 years of in-service aging [22]. For STOA, the mixture was spread to a height of ca. 50 mm on a metal pan and placed in a forced draft oven at 135 °C for 4 h. Thereafter, for LTOA, the compacted specimens were fabricated from the STOA aged mixture and placed in the oven at 85 °C for 5 days.

Unaged (U), aged (A), non-recycled (05 and 30) and recycled (07 and 32) bitumens were recovered from the AC mixtures (Mx) and bitumen BitRAP was recovered from RAP, using the European standard methods where toluene is used for washing the bitumen from the mineral aggregates [23]. V70/100, not mixed with mineral aggregates, was subjected to washing with toluene and to the rotatory evaporation procedure (V70/100_r) in order to study the influence of the recovery procedure on the bitumen properties. A sample with a blend of 2.5 g of V70/100 and 2.5 g of BitRAP (Blend50/50) was prepared by placing it in the oven at 134 °C for 10 min, stirred by hand for 1 min and placed back in the oven for another 10 min, before removing it from the oven and stirring it by hand for another minute. Fig. 1 shows schematically the various bitumens investigated.

Mx05 contained only V70/100, Mx07 contained a combination of V160/220, V70/100 and RAP bitumen (from 40% RAP), Mx30 contained only V10/20 and Mx32 contained both V10/20 and RAP bitumen [16].

2.2. Methods

2.2.1. Sample preparation

Different measurement techniques required different sample preparation methods. For the samples measured with dynamic shear rheometer (DSR) approximately 5 g of bitumen were heated in a ventilated oven at 110 ± 2 °C for 18–20 min and, subsequently, poured into silicone rubber molds of 20 or 8 mm diameter and 2 mm height. The samples were left to cool down at room temperature (~23 °C) and tested 24 h later.

For the differential scanning calorimetry (DSC), approximately 10 mg of bitumen were weighted and spread with a spatula on the bottom of the aluminum DSC pans. Afterwards the aluminum pans were placed in the oven at the annealing temperature of 110 ± 2 °C for 18–20 min for promoting the uniform spreading of the bitumen on the bottom of the pan. Afterwards the samples were removed from the oven, covered and left to cool down at room temperature for 24 h prior to testing, as recommended elsewhere [24,25].

For the atomic force microscopy (AFM) measurements, bitumen films were prepared by removing approximately 10 mg of the bitumen from the supplier's bucket with a spatula at room temperature, avoiding the bitumen at the surface of the bucket that is normally aged, and subsequently spread by buttering action over a ca. 0.8 × 0.8 cm^2 area on a 1 × 1 cm^2 glass slide. The annealing was performed as described before. After removal from the oven, the films were covered to avoid dust deposition and allowed to cool down. This procedure resulted in smooth and glossy bitumen film surfaces. Thereafter, they were conditioned at room temperature for 24 h prior to measurement. The annealing temperature of 110 °C was chosen because it was situated above the melting temperature of the crystalline-like structures present in bitumen.

2.2.2. Dynamic shear rheometer (DSR) measurements

The DSR measurements were performed in a Physica MCR 301 rheometer from Anton Paar. The complex modulus ($|G^*|$) and phase angle (δ) were determined according to the European standard [26] and the equations below:

$$G' = \frac{\sigma_0}{\varepsilon_0}\cos\delta \quad (1)$$

$$G'' = \frac{\sigma_0}{\varepsilon_0}\sin\delta \quad (2)$$

$$\left|G^*\right| = \sqrt{G'^2 + G''^2} \quad (3)$$

$$\frac{G''}{G'} = \tan\delta \quad (4)$$

where σ_0 is the ε_0 are the stress and strain amplitudes, respectively; G' and G'' are the real (elastic) and imaginary (viscous) parts of $|G^*|$.