Construction and Building Materials 48 (2013) 691–699

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Microscopic analysis of the interaction between crumb rubber and bitumen in asphalt mixtures using the dry process

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HIGHLIGHTS

- Carbon black is transferred to bitumen after a bitumen–rubber interaction in wet and dry processes.
- The lighter components of the bitumen are absorbed by rubber and transferred to carbon black.
- The modified bitumen is stiffer, reducing fatigue cracking.
- Rubber is transformed into a gel preventing the formation of voids around rubber particles of <200 μm.

ARTICLE INFO

Article history:
Received 27 May 2013
Received in revised form 12 July 2013
Accepted 21 July 2013
Available online 24 August 2013

Keywords:
Crumb rubber
Carbon black
Dry process
Fluorescent microscopy
SEM
Rheological characterisation

ABSTRACT

A microscopy study of the modification of bitumen with crumb rubber was carried out to investigate its application in asphalt mixtures using the dry process. The modification involved the transfer of maltenes to the rubber and carbon black to the bitumen, causing the absorption of the lighter-oil fractions of bitumen. The modification increased the stiffness of the bitumen, resulting in an improvement in the binder's resistance to rutting. The changes in shape and porosity developed around the crumb rubber particles by the rubber–bitumen interaction allow the most favourable particle size (<200 μm) to be established for the dry-process in crumb rubber-modified asphalt mixtures.

1. Introduction

Currently, the addition of crumb rubber (CR) to modify bitumens is widely used to solve the environmental problem of scrap tires and to develop high-quality, hot bituminous mixes, especially using the wet process. In the latter, the CR is added to hot bitumen and the mixture is shaken until the bitumen–rubber interaction occurs, the aggregate being added after the interaction. A cheaper alternative is the dry process, where the aggregate and the crumb rubber are hot mixed prior to the addition of the bitumen, allowing a maceration time for the mixture until the bitumen–rubber interaction is completed [1–4].

Nowadays, microscopy descriptions of the rubber–bitumen interaction process are scarce, especially in comparison with the huge volume of work addressing its rheological properties [4–14]. The swelling of crumb rubber in hot bitumen has been described elsewhere, using cold-stage optical microscopy [15], the basket drainage method [5,10,16] or by direct examination of a small number of rubber particles in hot bitumen [17]. However, the transfer of chemical components between rubber and bitumen, and the implications of this in the rheological properties of the mixtures have not been fully elucidated. Rheology has been assessed in terms of storage stability tests (i.e. separation between commercially available crumb rubber and bitumen takes place, the crumb rubber particles sink to the bottom of the storage cylinder) and a spectra showing exchange of elements has been reported [15].

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http://dx.doi.org/10.1016/j.conbuildmat.2013.07.041

Moreover, fluorescent microscopy has been extensively used by other authors to study the components transferred in the swelling of polymers in asphalt bitumen [18,19], but this technique has not been used for crumb rubber modified bitumen because of the sample size limitation.

In these products the absorption of the lighter fractions found in maltenes has been confirmed because they can be detected by fluorescent microscopy. However, this technique has rarely been used to investigate the crumb rubber–bitumen interaction because of the sample size limitation and the products transferred from one component to another are not widely known. This imprecision in our state of knowledge may influence interpretation of the results of rheological tests. Similarly, compositional variations of the modified bitumen, commonly attributed to the absorption of the lighter fractions of the bitumen into the rubber, might not be the best way of proceeding since other compounds present in the crumb rubber particles (e.g., carbon black) could also absorb these light fractions. The variation of the concentration of bitumen into commercially available rubber particle with distance from the centre to the edge of the particle with time is known [20] and this could be used to determine the rheological behaviour of crumb rubber modified bitumen. This needs to be investigated, if possible.

More evident is the absence of microscopy studies for the dry-process in crumb rubber-modified asphalt mixtures (DPCRAM). Such studies would easily allow the rubber particles that have been digested by the bitumen to be distinguished from those that have not, establishing the most suitable particle size for processing high-quality end products. The distinction between digested and undigested rubber particles has not been addressed in the literature but it could be easily investigated if the products of the bitumen–rubber interaction together with the rheological changes undergone by rubber particles with digestion were known and identified. Moreover, microscopy studies in DPCRAM also allow variables that are not usually expected in conventional microscopic studies to be considered, such as the presence of aggregate. This is not a minor issue, since a different distribution of the polymeric phase has been noted in SBS-modified asphalt mixtures (mainly in cracks or voids) compared with simple bitumen-polymer mixtures [21].

This work reports a microscopy study and presents an analytical characterisation of the products resulting from the bitumen–rubber interaction. Once the products have been defined it will be possible to report an improved interpretation of the rheological test results obtained in this work for the dry-process in crumb rubber-modified asphalt mixtures. Using microscopy techniques, an attempt has been made here to estimate the crumb-rubber particle size that does not interact with conventional bitumen (B 50/70) in order to optimise the preparation of the asphalt mixtures using the dry process.

2. Materials and methods

2.1. Materials

2.1.1. DPCRAM

This mixture consisted of inorganic aggregate, crumb rubber and bitumen (Table 1). Samples were obtained from PAS SL laboratory. The design was very similar to that currently used to obtain the usual working formulas of conventional hot asphalt mixtures, excluding analysis of the interaction time. The crumb rubber used was recycled rubber from discarded tyres supplied by RENECAL. Crumb rubber was produced at room temperature by grinding (i.e., the ambient grinding process, therefore commercially available crumb rubber). In order to achieve a significant bitumen–rubber interaction, 0–0.5 mm particle-size crumb rubber was used [3,4]. The composition by weight of the crumb rubber was as follows: rubber (57%), carbon black (33%), ash (9.6%), acetone extract (8%), ZnO (2.2%) and sulphur (1.6%), containing impurities of textile fibres (<0.5%), and FeO (<0.01% for sizes smaller than 0.8 mm). The overall ratio of natural to synthetic rubber was 65/35. Conventional B 50/70 penetration bitumen was used in this study, supplied by the La Rábida refinery (Huelva, Spain). The properties of the bitumen are as follows: penetration (ASTM D5-73, 100 g at 25 °C × 0,1 mm), 66; softening point (Ring and Ball test, ASTM D36-76, EN 1427), 49.8 °C; Fraass brittleness temperature (EN 12593), −13 °C. The nominal chemical composition of the bitumen was as follows: saturates, 10%; aromatics, 50%; resins, 25%; and asphaltenes, 15%. The other properties were in compliance with current regulations. Crushed aggregates of quartzite were supplied by PAS S.L. (Castraz, Salamanca, Spain). The aggregate gradation chart adopted corresponded to the agglomerate called AC16, according to EN standards. The gradation was followed to manufacture all samples, but with the modification of their bitumen content and the percentage of crumb rubber additions. The samples used in this study were obtained from the specimens prepared in [2,3], all of them with 1% of crumb rubber by weight of the aggregate (20% by weight of the neat bitumen) [3]. Higher rubber quantities provided values that were too low in the Marshall stability test and had detrimental effects on the agglomerate: higher air void percentages, compaction difficulties in the field, and rubber segregation [3]. The digestion time or immersion time of rubber particle into bitumen seems to be a critical parameter in the quality of mixtures since the concentration of bitumen into crumb particle with distance from the centre to the edge of the particle varies with time [20]. Therefore, two different samples were selected: a sample without digestion time, i.e. the period of interaction of crumb rubber with bitumen was only a few minutes, and another with a digestion time or immersion time of 2 h that may give a concentration profile similar to the one reported in the literature [20]. The latter yielded the mixtures with the best quality according to [3] leading to the conclusion that it is possible that this quality corresponds to a similar concentration profile as reported in the literature.

Table 1
Composition of the asphalt mixtures.

Samples	Mixture A	Mixture B	Mixture C
Aggregate gradation	AC16	AC16	AC16
Type of bitumen (PROAS)	B 50/70	Styrelf modified 16/60	B 50/70
Bitumen content (wt.%)	5.0	5.0	5.3
Rubber content (wt.%)	–	–	0.9
Mixture temperature (°C)	165	170	170
Compaction temperature (°C)	155	155	155
Digestion time (hours)	–	–	2

2.1.2. Unmodified hot-mix asphalt pavement

This mixture was produced using with the same components as in DPCRAM except for the absence of crumb rubber (Table 1). This sample was obtained for purposes of comparison for both the microscopic and rheological studies.

2.1.3. Asphalt pavement with a bituminous binder modified with polymers (Styrelf 16/60)

This hot asphalt mixture was manufactured with the same type of aggregate used in DPCRAM (Table 1) and it was also used to compare its rheological characteristics relative to those of the DPCRAM and the unmodified asphalt pavement.

2.1.4. Standard samples for microscope study

Given the difficulty of studying thin sections of DPCRAMs under the microscope, thin sections of each component of the DPCRAM were produced that consisted of bitumen, rubber particles, aggregates and epoxy resin, the latter was used to aid in producing solid thin slides. It was also essential to identify the products of the bitumen–rubber interaction, which were studied by means of a hot mixture composed of bitumen and crumb rubber (MCBR, i.e. without aggregate) using thin section that were heated to 170 °C for 2 h, i.e. the same processing conditions as those of the DPCRAM.

2.2. Methods

2.2.1. Microscopy

The study of standard samples, DPCRAM and MCBR, was performed using the following microscopy techniques:

2.2.2.1. Transmitted- and reflected-light microscopy. Components of DPCRAM showed different grades of reflectivity and transmissivity, allowing them to be well distinguished when both techniques are combined. This technique has also been used to determine the size of the rubber particles using a calibrated eyepiece.

2.2.2.2. Fluorescence microscopy. This technique is suitable for studying polymer-modified bitumens and to examine the bitumen–rubber interaction. Unmodified bitumen appears as brown areas under the fluorescence microscope, while polymer-modified bitumen, apart from the brown coloured areas, also appears as