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Chemically modified bitumens with enhanced rheology and adhesion properties to siliceous aggregates

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HIGHLIGHTS

- Evaluation of potential moisture damage in two chemically modified asphalt mixtures.
- Bitumen modifications by thiourea and an isocyanate/castor oil prepolymer.
- Both chemical paving binders with improved end-performance.
- Moisture-induced damage highly influenced by the binder-aggregate adhesion mechanism.
- Polyurethane modification decreases sensitivity of the asphalt mixture to water.

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ABSTRACT

Moisture damage is one of the major premature failures that worsens the performance and shortens service life of pavements. This research assesses the effect that two chemical modifiers (thiourea and an isocyanate-functionalized castor oil prepolymer) exerts on the bitumen rheology and on the resistance to potential moisture damage of asphalt mixtures based on siliceous aggregates. Both thiourea and the isocyanate-based prepolymer improve the viscous and viscoelastic behaviours of bitumen at high in-service temperatures, particularly the isocyanate-based reactive modifier. Likewise, the visual inspection of the degree of bitumen coating on loose mixtures exposed to water in a “rolling bottle” test, and further surface free energy measurements, showed that the bitumen modification by the isocyanate-based prepolymer seems to reduce moisture-induced damage on siliceous aggregates. Conversely, thiourea-based modification increases sensitivity of the bitumen–aggregate mixture to water. Different adhesion mechanisms, based on chemical or physical bonds with the aggregate surface, have been proposed for each modifier.

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1. Introduction

Even the best designed and constructed road pavements deteriorate over time under the combined effects of traffic loading and weathering. The most common distresses are: rutting, or permanent deformation at high temperatures [1]; thermal cracking, or thermal fracture due to the lack of flexibility at low temperatures [2]; and fatigue cracking, due to the repetitive application of loads [3]. As a consequence, the bitumen performance has been improved by the addition of widely used virgin polymers (SBS, SBR, EVA polymers) [4,5] or reactive agents (like polyphosphoric

acid [6]). Other no-common additives employed in the bitumen modification are waste polymer [7,8] (plastic from agriculture, crumb tyre rubber, etc.), sulphur [9] or polyurethanes [10–12].

In addition to them, moisture sensitivity of an asphalt mixture, generally called stripping potential, is among the most important distresses of asphaltic pavements [13]. Thus, the penetration of moisture through asphalt mixtures increases the extent and severity of the previously commented common distresses. Many variables affect the amount of moisture damage which occurs in an asphalt mixture. Some of them are related to the materials forming hot mix asphalt (HMA) such as aggregate (physical characteristics, composition, dust and clay coatings) and bitumen (chemical composition, grade, hardness, crude source and refining process). Others are related to mixture design and construction (air void

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level, film thickness, permeability, and drainage) or environmental factors (temperature, pavement age, freeze–thaw cycles and presence of ions in the water) [14].

On the other hand, the conventional method of identifying moisture sensitive asphalt mixes during the mixture design process is by comparing results from mechanical tests on dry and moisture-conditioned asphalt mixtures. However, these tests are not always reliable and do not provide any information regarding the causes for good or poor performance of a particular mixture [15]. To that end, the surface energy theory can explain adhesion and cohesion bonds according to both aggregate and binders physico-chemical properties [13,16,17]. Based on surface energy measurements, Cheng et al. [18] pointed out that the affinity of the aggregates for water is far greater than that for bitumen. As a result, water is able to replace (strip) the bitumen from the aggregate surface, with the rate of replacement dependent on the aggregate-bitumen bond strength in the presence of water. Consequently, the thermodynamic energy potential that drives the stripping phenomenon depends on the surface properties of the bitumen, the aggregate and water [17]. In order to prevent asphalt from experiencing moisture damage, both the surface properties of aggregates and binders can be changed. Regarding the latter, different bitumen modification routes by polymers (such as styrene-butadiene-styrene (SBS) or styrene-butadiene-rubber (SBR) [14,17]) and amine-based additives [19,20] have been widely performed. On the other hand, some experiences using epoxy resins [21] or organic-based calcium compounds [22] have been also studied in the past.

However, it is generally recognised that there is a lack of experimental data concerning the moisture damage of modified asphalt mixtures [23], which are obtained by mixing the aggregate with a modified binder. With this aim, this paper reports the effect that bitumen modification by two chemical modifying agents (thiourea and an isocyanate-functionalized castor oil prepolymer) exerts on the binder rheology and on the resistance to potential moisture damage of asphalt mixtures containing siliceous aggregates, which are well-known for their poor adhesion features. To that end, two different evaluation methods of moisture sensitivity were conducted: (i) visual assessment of the degree of bitumen coverage on loose bitumen–aggregate mixtures (the so-called “rolling bottle” test); and (ii) non-mechanical tests on mixture constitutive parts, by measurements of the surface free energy components of asphalts parts separately and assessment of the strength of adhesion bonds between binder and aggregate. In addition, the rheological characterisation of the modified binders was also conducted through viscous flow curves, at 60 °C, and temperature sweep measurements.

2. Experimental section

2.1. Materials

Bitumen with penetration grade 70/100 was used as base material for the bitumen modification. The results of penetration grade (EN 1426:2007), softening temperature (EN 1427:2007) and “SARAs” fractions [24] are shown in Table 1 in terms of average values and their calculated standard deviations.

Two different chemical modifiers are proposed. The first one, thiourea, is a non-polymeric substance, whilst the second one is an isocyanate-functionalized-castor-oil-reactive prepolymer. Detailed information on these modifiers is given below:

- (1) Thiourea has been previously used as an additive to improve coating quality and to inhibit corrosion in several applications. It was supplied by Sigma Aldrich, has a molecular weight of 76.12 g/mol and its melting point lies within the interval 175–179 °C.
- (2) Castor oil supplied by Guinama S.L. (Spain), with hydroxyl index of 125 mg KOH/g (measured according to standard ASTM D1957), was functionalized with isocyanate groups, by its reaction with a polymeric 4,4'-diphenylmethane diisocyanate (with an isocyanate groups, –NCO,

Table 1

Penetration value, Ring & Ball softening temperature, “SARAs” fraction and colloidal index value for the neat bitumen used.

	Values ^a
Penetration (dmm)	82 ± 3
R&B softening point (°C)	58.0 ± 1.5
Saturates (wt.%)	6.2 ± 0.8
Aromatics (wt.%)	56.3 ± 2.1
Resins (wt.%)	23.4 ± 1.2
Asphaltenes (wt.%)	14.1 ± 1.1
Colloidal index (C.I.)	0.26

^a Mean values ± SD (standard deviation, $p = 0.05$).

content of 31 wt.%) provided by T.H. TECNIC, S.L (Spain). This reaction was carried out at 60 °C, for 48 h, under agitation and with N₂ blanketing. In order to ensure an excess of non-reacted –NCO groups in the reaction product, an equivalent ratio –NCO/–OH of 8:1 was selected. This ratio was found optimal in a previous work [12]. The resultant reactive prepolymer had an average molecular weight of 2640 g/mol, and is expected to bring about “chemical” modification of bitumen, via reaction of –NCO groups and bitumen compounds having “active” hydrogen (i.e. –OH, >NH, etc.) [10–12,25,26].

In order to establish a comparative analysis with a non-reactive additive, a commercial thermoplastic elastomer styrene-butadiene-styrene (SBS) “Kraton D-1101” (provided by Shell Chemical Company U.K.) has also been studied. This triblock copolymer is able to modify through physical dispersion in bitumen. It has a molecular weight of $1.5 \cdot 10^5$ g/mol and a styrene content of 31 wt.%.

Finally, acidic siliceous aggregate provided by Mountsorrell Quarry, located in Leicestershire, U.K., has been used in this research. They are composed largely of silicon and oxygen, with the addition of ions such as aluminium, magnesium, iron and calcium. Due to the acidic nature of bitumen, this type of siliceous aggregate is well known for its poor field moisture damage performance [27,28]. Before being used, the aggregates were washed with deionized water and then dried in an oven at 105 ± 5 °C overnight.

2.2. Preparation of modified binders

Blends of bitumen with the different chemical modifiers were processed in an IKA RW-20 stirring device (Germany) equipped with a four-blade turbine. A vessel (60 mm diameter; 140 mm height) containing bitumen was heated up to the processing temperature. Once this was reached, the modifier was added and the blend was stirred under the conditions described in Table 2. For the non-polymeric modifier, 9 wt.% thiourea was processed at 180 °C (above its melting point) for 1 h. These samples will be referred to as “Th-binder”. As for the polymeric additive, the processing of their corresponding modified binders consisted in firstly mixing bitumen and 2 wt.% MDI-CO for 1 h at 90 °C and, subsequently, adding 2 wt.% water for 45 min at the same processing temperature. This additive is a liquid with very low viscosity at the processing temperature of 90 °C. For this reason, a mechanical agitation at 1200 rpm for 1 h is enough to properly disperse it into the bituminous matrix. The resultant modified binders will be referred to as “MDI-binder”.

In addition, a bituminous reference sample with 3 wt.% of SBS (“SBS-binder”, hereinafter) was prepared with a Silverson LSM (Silverson Machines Ltd., U.K.) homogenizer under the conditions detailed in Table 2, as this rubber requires more severe conditions to obtain a proper homogeneous dispersion.

3. Testing procedures

3.1. Tests and measurements on modified binders

Different rheological tests were conducted on neat and modified bituminous samples using a controlled-stress rheometer Physica MCR-301 (Anton Paar, Austria): viscous flow measurements, at 60 °C; and temperature sweep tests in oscillatory shear, between 30 and 100 °C, using a continuous heating ramp of 1 °C/min, a frequency of 10 rad/s and deformation of 1% strain (within LVE interval). Plate-and-plate geometry (25 mm diameter; 1 mm gap) was always used. In order to ensure accurate results, at least two replicates were conducted for every sample.

FTIR spectra were obtained with a Digilab FTS3500ARX (Varian, USA) apparatus. Solutions of neat bitumen and “MDI-binder” were prepared by dissolving 0.4 g of each in 25 ml of toluene. This solvent was chosen because of its lack of absorbance in the spectral

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