



Sustainable asphalt mixes manufactured with reclaimed asphalt and modified-lignin-stabilized bitumen emulsions

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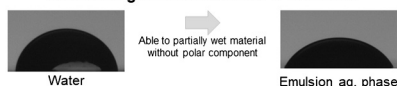
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HIGHLIGHTS

- 100% asphalt reclaimed by lignin-stabilized bitumen emulsion at reduced temperature.
- Cationic lignin in the emulsion aqueous phase promotes hydrophobic surface coating.
- Higher addition of soft bitumen emulsion for lower mixing-compaction temperatures.
- A semi quantitative approach is proposed for estimating emulsion addition.

GRAPHICAL ABSTRACT

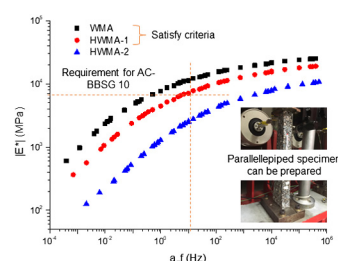
Contact angle of emulsion aqueous phase containing modified lignin emulsifier on PE substrate



Asphalt mixes prepared with 100% RAP and emulsions following reduced temperature procedure



Mechanical performance of asphalt mixes



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ABSTRACT

This work aims to develop asphalt mixes with 100% reclaimed asphalt pavements (RAP) as source of aggregates and bitumen emulsions stabilized by amine-functionalized lignin. To that end, reduced-temperature asphalt technologies between 80 and 130 °C were assessed. Previously, processing parameters of emulsions were evaluated by droplet size distribution and rheology measurements. Furthermore, a study combining Pendant drop and Sessile drop (on a prototype polyethylene surface) tests showed that emulsion aqueous phase containing modified lignin would be able to wet the hydrophobic surface of RAP. Manufactured asphalt mixes were characterized according to French design method for hot mixes, including gyratory compaction, rutting resistance, complex modulus and fatigue resistance tests. The results obtained showed that reduced-temperature asphalt mixes could be successfully prepared to meet French design requirements, which would further minimize impact to environment. However, mix design has required a semi-quantitative approach for estimating 'active' RAP binder content, by balancing the manufacturing temperature and the fresh bitumen required.

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1. Introduction

In the last decade, soaring pressure on the global environmental challenges demands immediate responses, as addressed in UN development agenda [1]. Particularly, issues involving sustainability, environment and economic aspects are mostly discussed. In

transport infrastructure construction sector, an example for such issues is the use of fresh bituminous and aggregate material, which has a high impact, up to 40% of CO_{2eq} (defined as mass of CO₂ emitted in kg over a ton of each layer needed during the lifecycle), if the extraction and production process are considered within the boundaries of analysis [2]. Moreover, conventionally used hot mixes method during road construction may be perceived unsustainable due to its energetically unfavorable process and high CO₂ equivalent emitted to the air [3,4]. In that sense, a reduction

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of virgin material (bitumen and aggregates) and the use of reduced-temperature processes would promote more sustainable road designs, addressing both environmental and economic aspects.

Concerning the raw materials, the use of waste materials can be both economically and environmentally interesting due to the possible reduction of fresh binder, as well as the natural aggregates used [5]. Such example reports a lower down score by 15% in fossil depletion, when reclaimed asphalt pavement (RAP) was employed considering an impact assessment according to two sets of impact categories: midpoint (single issue) and end-point (higher aggregation level) categories, so called ReCiPe method [2]. That analysis is based on the comparison of three-structured layers (surface, binder and base) of which, one is manufactured with all virgin material and the other consist of up to 30% of RAP for the base layer. A reduction of 13–14% in the cumulative energy demand has also been reported for 15% RAP recycled [6]. Besides those benefits, mechanical performance, stiffness modulus and rutting resistance could be improved [7–9]. Obviously, such utmost benefit may be expected for a higher RAP recycling rate. Today, their uses, authorized in Europe according to EN 13808-8 [10], are yet generally limited especially for surface layer [11]. This is related to the difficulty in controlling the characteristics of aged binder and aggregates that potentially affect recomposed asphalt mixes final performance. For that reason, many studies focus on studying the influence of incorporating RAP at higher percentage, even up to 100% recycling, on their mechanical behavior, aiming at speeding up its authorization [12].

Partial blending phenomena, between RAP binder and fresh bitumen, yet require understanding. This widely accepted concept results in altering the performance of blended binder [13,14]. For RAP recycling rate less than 25%, a study reports that no particular change in binder grade is necessary [15]. Another study suggests an increase of degree of partial blending from 70 to 96% when mix compositions have been changed from involving 25% RAP and binder with PG 70–28 specification to 35% RAP and softer PG 50–28 [16]. From diffusion point of view in which diffusion coefficient and viscosity are related by Arrhenius function with temperature, the lower the viscosity of bitumen at a particular temperature (or softer characteristic), the more favorable the diffusion to occur due to high diffusion coefficient [17]. The comprehension of partial blending in HMA seems to drive in a converging direction, yet debatable on the investigation method for the moment. Unfortunately, when RAP is recomposed following a reduced-temperature mixing procedure, partial blending is another challenge. Many works done on recycled warm mixes mostly explain about bitumen content selection [8,18], with the assumption of fully recovered RAP binder and without partial blending. In this work, a semi-quantitative approach, referred to thin-film diffusion phenomenon, has been proposed trying to understand partial blending under reduced-temperature mixing conditions. As a result, the fraction of ‘active’ RAP binder blended with the fresh bitumen could be estimated.

Another interesting material that can potentially be used in road infrastructure is lignin. A study reports that lignin seems to be compatible with bitumen due to the similarity in their aromatic molecular structure [19]. Lignin is a renewable material, derived from wood in the form of lignocellulose. While the cellulose is the main input of pulp plant, lignin mainly ends up burnt in heat regenerator [20]. Due to the aromatic structure, lignin potential as a precursor for advanced materials could be opened through chemical modification. By lignin amination, for instance, cationic emulsifier for bitumen emulsion application could be obtained. Previous study [21] shows that modified lignin may prevent coalescence, as observed by insignificant change in average droplet size and its distribution profile, hence providing good stability.

Despite stability during storage, bitumen emulsion also requires to wet the aggregate surface upon application, by a balance between the negative charge of the fresh aggregate and the positive charge of the cationic surfactant [22]. However, if the bitumen emulsion (and the emulsifier) is designed to coat a 100% RAP instead of virgin aggregates, its physical properties may not be treated equivalently due to the difference in surface characteristics. Thus, further research regarding emulsion/surfactant wettability on a hydrophobic surface is required to assess their interactions with RAP surface.

With the main objective to design asphalt mixes with maximum recycling and minimum impact to environment, this study is divided into three subtopics with the aims of: assessing the ability of emulsifiers derived from lignin to partially wet RAP surface; optimizing processing parameters during the manufacture of bitumen emulsion; and producing reduced-temperature asphalt mixes containing 100% RAP without sacrificing their mechanical performance.

2. Experimental

2.1. Materials

Kraft lignin (KL), tetra-ethylene-pentamine (TEPA) and formaldehyde, were used as reagents for producing cationic modified lignin (MKL). Kraft lignin used has a molecular weight of 10000 g/mol, an ash content of 3.14 wt% and elemental composition (wt%) consisting 61.15 of C, 6.72 of H, 26.92 of O, 2.18 of N and 2.54 of S. As for the reference, *n*-tallow alkyl tri-methylene diamine (referred to as REF), was selected. Penetration and Ring-and-Ball softening temperature tests were performed on bitumen according to EN 1426 and EN 1427 [23,24]. The values obtained were 190 dmm and 39 °C, respectively. Reclaimed asphalt pavement material (RAP) was obtained from a real scale road fatigue simulator under accelerated heavy traffic, was kindly supplied by IFSTTAR (France).

2.2. Preparations

MKL emulsifier reaction procedure was presented elsewhere [21]. Afterwards, concentrated MKL was diluted with acidic water at pH 1 to attain final concentrations between 0.625 and 3.75%. Oil-in-water (O/W) emulsions were prepared by emulsifying 60 wt% bitumen into that aqueous phase, at a selected concentration of 1.5 wt% of MKL emulsifier. For sake of comparison, four emulsification devices were used in this study: A) three batch homogenizers UT T25, UT T50 and a Sill5M; and B) an inline three-stage high-shear homogenizer, referred to as 3STCM. The first three devices are small capacity processing units (200–500 gr). A premix of 145 °C bitumen and 60 °C of aqueous surfactant solution yielded a blend with an approximate temperature of 90 °C, to avoid water evaporation. Short after, emulsification at the maximum speed was done for 4 min. The difference with the in-line homogenizer type is in the capacity and the ability for continuous recirculation of the emulsion. When recirculation line was employed, the maximum capacity is 5 kg, and emulsification time was further evaluated on 3STCM by taking sample through recirculating line starting from 1 to 12 min. For the reference emulsion stabilized by 0.5 wt % REF, surfactant was dissolved in aqueous phase with pH 1 and the rest of procedures were similarly followed.

For RAP binder analysis, sample was extracted using perchloroethylene solvent on automatic asphalt analyser (EN 12697-1) [25] followed by binder recovery process in rotary evaporator (EN 12697-3) [26]. Details on the conditioning for mixing and compaction and the quantity of added water and fresh bitumen are

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