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Study of remobilization phenomena at reclaimed asphalt binder/virgin binder interphases for recycled asphalt mixtures using novel microscopic methodologies

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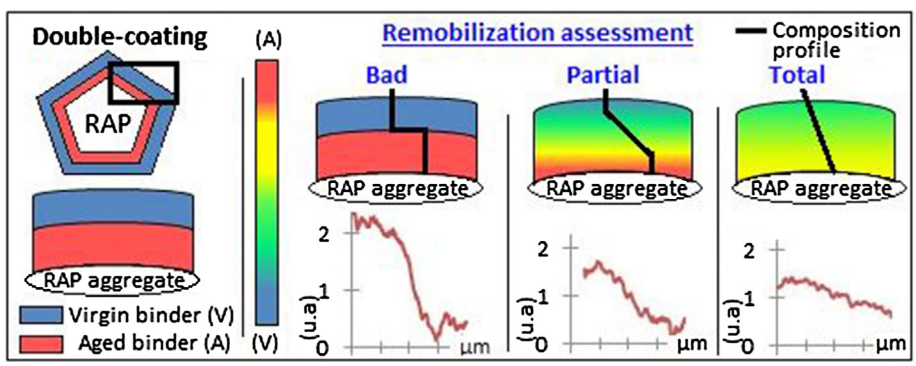
HIGHLIGHTS

- Infrared Imaging ATR is a suitable technique to investigate binder remobilization.
- Carbonyl distribution profile is efficient to monitor the aged binder migration.
- The aged binder viscosity influences significantly the distribution profile slope.
- Higher the aged binder temperature, longer the blending length between binders.
- The rejuvenator addition improves the aged binder remobilization.

GRAPHICAL ABSTRACT



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ABSTRACT

Objectives are to understand remobilization phenomena which occur during the manufacture of recycled asphalt mixtures. Novel X-ray micro fluorescence and infrared microscopy methodologies have been customized to the "aged binder/virgin binder" interphase. Infrared Imaging ATR is the best technique to monitor the aged binder migration coupled to the carbonyl function as a distinguishable marker. Furthermore, the rejuvenator addition and a high aged binder temperature increase the blending zone length between binders. The carbonyl distribution profile slope assesses also the binders blend quality and directly depends on the aged binder viscosity. Another impacting factor could also influence the binder remobilization and could be based on molecular distributions.

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1. Introduction

1.1. Background

In the road industry, the manufacture of Warm Asphalt Mixtures (WMA) incorporating RAP (Reclaimed Asphalt Pavement) represents an important ecological potential because the reference manufacturing temperature is reduced by 40 °C, what leads to significantly reduce gas emissions [5,9,25]. Furthermore, recycling of asphalt mixtures allows decreasing the consumption of non-renewable resources (bitumen, aggregates) because fine layers of aged bitumen around RAP materials are reused.

However, the RAP aged binder has a chemical composition which differs from the virgin one: the asphaltenes content increases with ageing and carbonyl C═O and sulfoxide S═O functions appear in bitumen. These changes lead to a more viscous binder and to a possible heterogeneous mixture between the RAP aged and virgin binder during manufacturing conditions. As a consequence, mechanical properties diverge in comparison with the predicted properties [21]. In fact, mechanical properties of asphalt mixtures incorporating high percentages of RAP depend both on the RAP binder interaction degree with the virgin one and on RAP aged binder properties:

- On one hand, R. Mc Daniel has shown that for a conventional asphalt mixture manufacture, a "partial blending" occurred [16], what has been confirmed by H. V. Nguyen [20]. This observation results from a complete study which has determined different mechanical properties of mixtures elaborated in different conditions: from the "black rock effect" (no RAP binder interaction) to the "total blending" situation (total RAP binder interaction).
- On the other hand, even if the RAP incorporation has led to a better rutting resistance [10,15,18], studies have also outlined dispersed results in moisture resistance [10,27] and fatigue resistance [10,22].

The blend between the RAP and virgin binder is also still misunderstood and it is important to determine the blending degree of recycled asphalt mixtures to guarantee efficient mechanical properties. The blending degree represents the total RAP binder percentage which is mobilized in an asphalt mixture [4]. Several studies have investigated it at the asphalt mixture scale [20,24], at the intergranular space scale [2,8] or by considering a stage method extraction on an asphalt mixture sample [6,11]. Other studies have also focused directly on the binders interphase in order to monitor the aged binder migration during the blend. X-rays micro tomography [7], Environmental Scanning Electronic Microscopy [2] or Atomic Force Microscopy techniques [17] were used with specific markers to monitor the aged binder. The markers have respectively been attributed to iron oxide, titanium oxide or different bee-microstructures. These studies concluded that the mixing time improved the RAP spatial distribution [20]. The manufacturing temperature also impacts the blending degree. Hot manufacturing temperatures (160 °C) lead to a faster RAP binder homogenization kinetics [19] and a thinner RAP binder thickness [2].

However, blending mechanisms remain not completely understood. Indeed, the stage extraction method results [11] have highlighted a double-coating of the RAP aggregate whose the formation has not yet be explained. Furthermore, during the RAP incorporation, the asphalt mix production step may imply a temperature difference between cold incorporated RAP materials and the virgin binder which is never taken into account in the previous studies. Finally, microscopic studies at binder interphase have used an external additive (iron, titanium oxides…) to assess the aged binder remobilization. A question about this methodology would be to verify if the external mineral additive impacts or interferes in the migration kinetics of the aged binder.

1.2. Objectives and experimental approach

Face to this interrogation, the objectives of this paper are to develop novel methodologies based on bitumen internal markers which allow investigating the bituminous interphase by considering a temperature difference between binders. Regarding the asphalt mixture recycling challenge, the second objective is to provide scientific answers about the influence of the RAP conditioning temperature and the rejuvenator use on the remobilization quality.

Remobilization represents ability for the virgin binder to make mobile again and disperse the RAP aged binder layer. For this purpose, several methodologies have been developed to monitor the aged binder migration: X-ray micro fluorescence, infrared microscopy in ATR (Attenuated Total Reflectance) and in Imaging ATR modes. Results are compared and the use of internal or external markers is discussed. Then, the research work is pursued with the most suitable methodology. The influence of the aged binder nature, the rejuvenator addition and the aged binder temperature is evaluated on the remobilization.

2. Materials and techniques

2.1. Pure and additived bitumens

Several binders were tested:

- a Hard Virgin binder, noted H2V, with a 50/70 penetration grade according to EN 12591 standard.
- a Hard Aged binder, noted H2A, obtained after a RTFOT (EN 12607 standard, 163 °C, 75 min) + PAV (EN 14769 standard, 2,1 MPa, 20 h, 100 °C) ageing procedure performed on the H2V binder.
- a Hard Aged binder (H3A), also obtained after a RTFOT + PAV procedure of a virgin binder H3V (35/50) which comes from a second refinery. This binder has been chosen to simulate a typical hardest RAP binder, coming from another origin.

Some additives were incorporated to binders, they correspond to a calcium carbonate filler $CaCO_3$ (referred to as Ca) and a rejuvenator (referred to as REJ) which is based on fatty acid methyl esters compounds. These additives have been chosen because they are directly involved in the formulation of recycled asphalt mixtures and they display a distinguishable infrared response (Fig. 4). For the preparation, an amount of 0.5% (by weight) of a rejuvenator agent has been incorporated to the H2V binder. An amount of 50% (by weight) of calcareous filler has been incorporated to the H3A binder to simulate the presence of high filler proportions in the crushed RAP material. The filler particle size is inferior to 63 μm. Additives were incorporated to respective binders which were first heated at 150 °C. Mixes (H2V + REJ and H3A + 50% Ca) were mechanically stirred with a blade during 2 min at 200 rpm.

Before the interphase investigation, binders were traditionally characterized according to the penetration index (defined by the EN 1426 standard), the softening point (defined by the EN 1427 standard), asphaltenes content (defined by a procedure deriving from the NF T 60–115 standard). Binder viscosities were also measured using a Malvern Kinexus Pro rheometer at three temperatures (120 °C, 140 °C, 150 °C) with a cone-plate geometry and an oscillatory mode, fixed at 0.01 Hz. Results (Table 1) show that the penetration index decreases and the softening point increases