Construction and Building Materials 133 (2017) 182–195

Contents lists available at ScienceDirect

Construction and Building Materials

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

Towards a better understanding of wetting regimes at the interface asphalt/aggregate during warm-mix process of asphalt mixtures

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HIGHLIGHTS

- Asphalt surface tension is related to the aromatic and asphaltenes content.
- Asphalt viscosity and asphaltenes content impact equilibrium contact angles.
- Substrate heterogeneities improve wetting quality on mineral surfaces.
- A complete characterization of mineral substrates by SEM is highly recommended.
- The CBOW model is a promising approach to characterize substrate heterogeneity.

ARTICLE INFO

Article history:
Received 4 August 2016
Received in revised form 14 October 2016
Accepted 5 December 2016

Keywords:
Wetting regime
Contact angle
Interface
Asphalt
Substrate
Scanning Electron Microscopy

ABSTRACT

In road applications, the current challenge is to develop more ecological products while maintaining asphalt mixture performance and durability. This sustainable development politics in civil engineering leads to promote techniques saving both energy and raw materials like combining the use of warm mix asphalt concretes (WMA) with the use of reclaimed asphalt pavement (RA). However, one of the current problematic when combining WMA and RA is to assess the quality adhesion of the "asphalt/aggre gate" couple that is a fundamental parameter for the good mixture properties at short time and thereby durability of this composite structure. Indeed, the reduction of the manufacture temperature and the use of an aged binder may also have consequences on adhesion quality between asphalt and aggregates. It is the reason why it is crucial to identify the most impacting factors of wetting phenomena at the interface "asphalt/aggregate". So, in this paper, the role of asphalt characteristics (viscosity, ageing, composition) as well as the one of substrate was investigated.

The substrate heterogeneity degree determines asphalt wetting behavior. For a model glass substrate, only asphalt characteristics have an impact: asphalt viscosity, polarity and saturates content are influent factors. On the mineral heterogeneous substrate, asphalt viscosity and asphaltenes content have an impact on wetting indicators. Substrate heterogeneities also appear as a very important factor which improves considerably wetting quality. The tested mineral substrate has been revealed as a biphasic composite material for which the wetting regime was modeled by a Cassie-Baxter model. A comprehensive approach has been proposed to explain asphalt wetting on heterogeneous substrates in correlation with Scanning Electron Microscopy observations. This Cassie-Baxter model is essential to understand adhesion phenomena on heterogeneous road materials. It seems also to be a promising way to evaluate quality adhesion of "asphalt/RA" mixtures.

1. Introduction

For many years, the objectives of road applications are to produce more ecological road materials to be as efficient and durable as traditional road techniques. In this framework, we take an interest in the development of warm-mix asphalt mixtures incorporating Reclaimed Asphalt Pavement (RAP). Road materials are composed by weight of 95% aggregates and 5% asphalt. Warm-Mix Asphalt (WMA) technologies were introduced to reduce the manufacture temperature by 20–40 °C. Production occurs at a temperature of 30 °C (between 110 and 140 °C) below

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http://dx.doi.org/10.1016/j.conbuildmat.2016.12.015
0950-0618/

temperature used for hot mixing. WMA advantages are numerous such as the reduction of energy consumption and CO_2 emissions [27]. Several studies have shown a decrease in aerosol and polycyclic aromatic hydrocarbons (PAHs) emissions [10,35], a decrease of CO_2 and SO_2 volatile organic compounds (VOC), NO_x as well as of dust [9].

In parallel, the valorization of Reclaimed Asphalt Pavement (RAP) in new manufactured road materials is of great interest thanks to the reduction of non-renewables resources. Indeed, this leads to re-use waste and to less consume natural virgin materials [25,26]. Benefits of using RAP in asphalt mixtures have also been outlined in several studies where significant reductions in greenhouse gas emissions, cost [33] and energy consumption [1] were observed. The use of 50% RAP in asphalt mixtures has even lead to reduce feedstock energy by 40% [1].

The combination of RAP use and WMA technology allows also reducing CO_2 emissions and energy consumption, respectively by 12 and 15% according to Giani [12]. As a consequence, the development of WMA mixtures incorporating RAP represents a double ecological potential.

Nevertheless, a main issue concerns performance and durability of pavement materials. Indeed, adhesion between asphalt and aggregates is a fundamental parameter for the good mixture properties at short time and thereby durability of this composite structure [15,28]. The bond strength must be optimal. However, the reduction of the manufacture temperature and the use of an aged binder may also have consequences on adhesion quality between asphalt and aggregates. Asphalt is a complex hydrocarboned visco-elastic compound which is sensitive to temperature and oxidation phenomena. That leads respectively to a viscosity increase and to an asphalt composition modification [19]. These consequences may also weaken the aggregate/asphalt bond because the binder wouldn't satisfactorily wet the mineral substrate, hence the bond strength couldn't be optimal [17,28]. This weak wetting may have consequences on durability. Indeed, a key factor of asphalt mixture durability is the resistance of the interface asphalt/aggregate to water displacement [13]. Water presence at the "asphalt/aggregate" interface is also considered as the main cause of stripping and road deterioration [3] and results partially from the low adhesion at the asphalt/aggregate interface at short time.

Numerous studies have been performed in order to study asphalt wetting. Asphalt surface tensions have been determined through pendant drop methods [14] or through sessile drops [18]. Mineral surfaces have also been characterized through water contact angles measurements in order to assess surface topography [3]. Concerning the "asphalt/aggregate" interface, previous work has focused to compare surface energy measurements and adhesion work values in order to select the most suitable couples "asphalt/aggregate" [2,4,30]. But the calculated adhesion work includes important bar errors [37]. The asphalt/substrate interface was also studied according a microscopic approach. SEM (Scanning Electron Microscopy) was used to analyze chemically substrate surfaces [16] or to observe the "asphalt/aggregate" interface at the triple point [32]. Few researchers have studied asphalt ageing influence on adhesion at the asphalt/aggregate interface. Only influence of ageing was investigated on asphalt surface tension [31] and no information is available about the aged binder wetting on mineral substrates.

As a consequence, there is a lack of information about understanding of wetting behaviors at the "asphalt/aggregate" interface. The main objective of the paper is to identify parameters impacting wetting regimes in order to better understand adhesion of "asphalt/substrate" couples and predict performance of warm-mix asphalt mixtures incorporating RAP.

2. Materials and methods

2.1. Materials

2.1.1. Asphalt

Six asphalt samples have been tested. Each asphalt sample is codified according to the following nomenclature: XYZ where:

- X corresponds to the asphalt grade (EN 12591): **S** for Soft and **H** for Hard which corresponds respectively to a penetration index of [160–220] × 0.1 mm and of [50–70] × 0.1 mm.
- Y corresponds to the crude oil origin: **1** and **2**
- Z corresponds to the ageing state: **V** for Virgin and **A** for Aged by the RTFOT + PAV procedure [RTFOT (EN 12607) (163 °C, 75 min) + PAV (EN 14769) (2,1 MPa, 20 h, 100 °C)]

Based on this nomenclature, the corresponding codes of study asphalts are: S1V, S1A, S2V, S2A, H2V, H2A.

2.1.2. Substrates

Two model substrates have been tested: polytetrafluorethylene (PTFE) slides (Goodfellow, thickness: 5 mm) and microscopic glass slides (Marienfeld, thickness: 1 mm). A mineral natural substrate was used which comes from mineral volcanic blocks (French quarry supplier). To obtain more details, surfaces were chemically characterized through EDX-SEM analysis. Both glass and mineral slides contain following elements: O, Si, Na, Al, Ca, K, Mg. The mineral slide composition differs by the presence of additional elements as titanium and iron.

PTFE slides were employed for asphalt characterization. They were cleaned using the following procedure: a degreasing solvent was used, then PTFE slides have been rinsed with ethanol and distilled water and dried. Microscopic glass slides were only rinsed with distilled water and dried. For mineral slides, a core drilling was performed on mineral blocks and resulting samples were cut to obtain mineral tablets (diameter: 40 mm, height 8 mm). A polishing procedure has been established according to a previous study [37]. In our study, the first step consisted in grinding slides under water for 5 min using successively three disks (220, 600, 1200 μm). To finish polishing, diamond suspensions (grain size: 9 and 3 μm) were employed with a polishing cloth for 8 min. Slides were finally cleaned using distilled water, dried in oven and stored at ambient air in a box for 14 days prior to measurements. Glass slides were referred as GS and polished mineral slides as PMS. For wetting experiments, several PMS samples were tested (Fig. 1).

2.2. Methods

2.2.1. Asphalt physico-chemical properties

Penetration index (EN 1426) and softening point (EN 1427) are tests dedicated to the measurement of asphalt consistency. Asphalt viscosity was measured with a Malvern Kinexus Pro rheometer (cone-plate geometry) according to the shear rate, from 0.1 s^{-1} to 10 s^{-1}. In this range, Newtonian behavior is established and viscosity is constant. Oxidation indices were determined through FTIR (Fourier Transformed Infrared Spectroscopy) according to the method n° 69 of LPC [20].

Lastly, chemical SARA fractions have been obtained using the SAR-AD method [6,22]. This chemical separation is more refined and allows a best separation of fractions with an additional subdivision of asphaltenes. Fractions were separated through four columns in series. Then, asphaltenes were desorbed from the first column with three different eluents.