



Influence of waxes on adhesion properties of bituminous binders



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HIGHLIGHTS

- Loss of adhesion represent one of the main causes of distress in asphalt pavements.
- We investigated the effects of two different waxes on bitumen–aggregate adhesion.
- The affinity was assessed by means of physico-chemical surface characteristics.
- Effects of wax and aggregates acidic/basic type are discussed.
- Independent evidences from rolling bottle tests well support the results.

ARTICLE INFO

Article history:

Received 7 July 2014

Received in revised form 9 October 2014

Accepted 26 November 2014

Available online 27 December 2014

Keywords:

Wax
WMA
Moisture damage
Surface energy
Adhesion
DCA
DVS

ABSTRACT

The benefits at the base of WMA technologies are well established in the literature. They include the reduction in the energy consumption during the production of mixtures, the reduction in the emissions to the atmosphere in the production plant and a safer working environment. Although many studies have demonstrated good performance of this technology overall, some aspects are still unclear.

This paper aims to investigate the effects of two different waxes, mixed with a 70/100 pen bitumen on bitumen–aggregate adhesion. Physico-chemical surface characteristics of three different aggregates were assessed and combined with those of the bitumens by means of the Dynamic Contact Angle and Dynamic Vapour Sorption Devices. These techniques are an alternative to the procedure used to visually assess the bitumen–aggregate affinity, as given in the in EN 12697-11 standard, in which the materials are mixed together and then immersed in water. Using these procedures the materials are not mixed together, but studied separately, and the evaluation of their affinity for one another is analytically expressed by means of the work of adhesion.

Results have shown that waxes adversely affect the affinity of bitumen with aggregates by differing amounts, depending on the wax type and on the acidic/basic type of the aggregate.

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1. Introduction

Asphalt mixture is a complex and heterogeneous material that generally includes aggregates, asphalt binder and air voids. Its overall mechanical response is primarily governed by the asphalt binder and by the stone-on-stone contact between aggregates [5,6].

Loss of adhesion between bitumen and aggregates and loss of cohesion within the mixture, in the presence of water, are referred to as moisture damage and represent one of the main causes of distress in asphalt pavements.

Although not all damage is caused directly by moisture, its presence increases the extent and severity of already existing distresses

like cracking, potholes, and rutting [17]. The presence of moisture results in a degradation of the mechanical properties of the asphalt mixture, i.e. loss of stiffness and mechanical strength, which ultimately leads to the failure of the road structure [8].

Many researchers have recognized that the replacement of bitumen film from the aggregate surface by water, referred to as stripping [11], is linked to interfacial tension relations of these materials.

Hefer et al. indicated a synthesis of theories on bitumen–aggregate adhesion, identifying the thermodynamic theory as a universal approach: these concepts have been used by many researchers for this purpose [10].

Based on the Young–Dupré equation, Van Oss et al. proposed an important relationship between the Gibbs free energy of adhesion and the surface energy of the materials involved [22]. Curtis et al. [3] determined Gibbs free energies from adsorption isotherms.

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Lytton utilized surface energies measured on bitumen and aggregate surfaces to calculate free energies of adhesion and cohesion by applying modern surface energy theories as discussed in the following paragraphs [15]. These concepts have played an important role in discoveries of the rules governing micro-fracture and healing in bitumen–aggregate mixtures and can be used to assess the adhesion characteristics [1,14]. The physico-chemical properties are directly related to the adhesion characteristics of the two materials and are responsible for adhesion or debonding between the materials [16]. Surface free energy (SFE) can therefore be considered to truly represent the physico-chemical surface characteristics of bitumen and aggregates and has been successfully used as a tool for the selection of moisture resistant materials [2].

2. Background

Thermodynamic theory is a feasible way to quantify the adhesion of a liquid on a solid, and hence of bitumen on aggregates. It is based on the concept that an adhesive will adhere to a substrate due to established intermolecular forces at the interface, whose magnitude is related to the surface free energy of the materials involved in the adhesive bond. In order to ensure a strong enough bond, the liquid must show an ability to maintain contact with the solid surface. Dropping a droplet of a liquid on a solid surface, the surface energies of the system will influence the wetting, leading to the formation of a bead of liquid with a determined contact angle. This angle, conventionally measured through a liquid, is the angle where the vapour–liquid interface meets a solid. The properties of interfaces can normally be described as a three-phase boundary. Young (1805) proposed an equation to obtain surface tension from the contact angle, θ formed when a droplet of a liquid is placed on a perfectly smooth, rigid solid. To each interface, forming the triple point, a surface or interfacial free energy or tension can be attributed, which is the work required to increase the interface surface by a unit. Thus, γ_{sv} is the surface free energy of the solid in equilibrium with the saturated vapour of the liquid, γ_{lv} is the surface tension of the liquid in equilibrium with the solid, and γ_{sl} is the solid–liquid interfacial free energy. Young's equation is the result of the triple line in equilibrium (tension in force per unit length) [21].

$$\gamma_{sv} = \gamma_{sl} + \gamma_{lv} \cos \theta \quad (1)$$

The following relationship plays a central role in the study of adhesion [7]:

$$W^a = \gamma_s + \gamma_l - \gamma_{sl} \quad (2)$$

where W^a is the work of adhesion, γ_s is the surface energy of the solid, γ_l is the surface energy of the liquid and γ_{sl} is the interfacial energy between the two materials in contact (liquid and solid for instance). By combining the Dupré Eq. (2) with the Young Eq. (1), the Young–Dupré equation is obtained:

$$W^a = \gamma_{lv} (1 + \cos \theta) \quad (3)$$

The Young–Dupré equation is the starting point for any method that utilizes contact angles to obtain surface free energies by relating the contact angle to the work of adhesion. This is also the way to relate contact angles back to adhesion. According to the acid–base theory, surface energy is comprised of a polar and a non-polar component and is conventionally measured in erg/cm² [22].

$$\gamma^{\text{total}} = \gamma^{\text{LW}} + \gamma^{\text{P}} \quad (4)$$

where the apex 'LW' denotes Lifshitz–van der Waals, whose forces represent the interaction between two symmetrical molecules.

They are composed mostly of London dispersive forces, in addition to Keesom and Debye forces, which are dipole forces that make up van der Waals forces. For these components, the Berthelot geometric mean rule holds (7).

The apex 'P', instead, stands for acid–base. The acid–base component represents polar, or specific, interactions mainly due to hydrogen bonding and are further separated into Lewis acid, γ^+ , and Lewis base γ^- components. These interactions are specific and are only possible between interaction partners with complementary acid–base molecules.

For a one-phase system the work of cohesion is defined as twice the total surface energy of the material:

$$W^c = 2\gamma^{\text{total}} \quad (5)$$

Consider a two-phase system in which the solid is the aggregate, represented by the subscript "A", and the liquid is the binder, represented by the subscript "B". The work of adhesion, from a thermodynamic point of view, can be seen as the opposite of Gibbs free energy ΔG^a , that is the difference between initial and final energy state of the system. The bond strength of binder or aggregate is the combination of these Lifshitz–van der Waals (non-polar) and Lewis acid/base forces and the total bond strength can be given as:

$$\Delta G_{AB}^a = \Delta G^{\text{LW}} + \Delta G^{\text{P}} = -W^a \quad (6)$$

where:

- ΔG_{AB}^a = total bond strength (Gibbs free energy of adhesion).
- ΔG^{LW} = adhesive non-polar bond strength, equal to $2\sqrt{\gamma_B^{\text{LW}}\gamma_A^{\text{LW}}}$
- ΔG^{P} = adhesive polar strength, equivalent to $2\sqrt{\gamma_B^-\gamma_A^+ + \gamma_B^+\gamma_A^-}$.

The adhesive bond strength in dry condition is obtained:

$$\Delta G_{AB}^a = 2\sqrt{\gamma_B^{\text{LW}}\gamma_A^{\text{LW}}} + 2\sqrt{\gamma_B^-\gamma_A^+} + 2\sqrt{\gamma_B^+\gamma_A^-} \quad (7)$$

Consider a three-phase system comprising asphalt binder, aggregate, and water, where these components are represented by 'B', 'A', and 'W', respectively. The following processes occur when water displaces the asphalt binder from the binder–aggregate interface.

Water and aggregates are polar substances while bitumen is a mainly a non-polar material. Thus water has greater attraction for aggregate surfaces than bitumen. This evidence increases the likelihood that water will be in contact with the aggregate, moving the bitumen film. Fig. 1 shows the process in which the water acquires a portion of the surface of aggregate to the detriment of the bitumen.

First, a part of the interface of the aggregate with the binder is eliminated (AB). Based on the definition of interfacial energy, the external work required for this is $-\gamma_{AB}$. Similarly, two new interfaces, between water and binder (BW), and between water and aggregate (AW), are created. The work done for the formation of these two new interfaces is $\gamma_{BW} + \gamma_{AW}$. Therefore, the total work done for water to displace binder from the surface of the aggregate is $\gamma_{BW} + \gamma_{AW} - \gamma_{AB}$. If the displacement process is thermodynamically favourable then it must be associated with an overall reduction in free energy of the system. In other words, the total work done on the system during the displacement process must be less than zero. Usually it is true for almost all asphalt binder–aggregate systems, suggesting that displacement of asphalt binder by water is a thermodynamically favoured phenomenon. In this context, the energy associated with the displacement of binder by water from the bitumen–aggregate interface, or de-bonding, is referred to as the work of de-bonding and is expressed for the adhesive bond strength in wet condition as:

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