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Measurement of water resistance of asphalt based on surface free energy analysis using stripping work between asphalt-aggregate system

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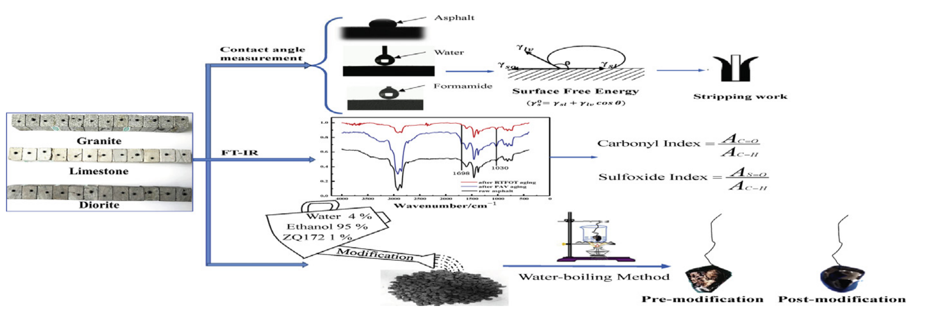
HIGHLIGHTS

- Stripping work of ten asphalts on three aggregates was studied.
- Stripping work was used to evaluate degree of water resistance of asphalt.
- Aging increased Carbonyl index and sulfoxide index enhancing asphalt stripping work.
- Larger ratio of surface free energy component with aging increased stripping work.
- ZQ172 silane as coupling agent enhanced water resistance of asphalt-aggregate.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, different surface energy parameters of three aggregates i.e. granite, diorite and limestone, and ten types of asphalts along-with their aging versions were measured using surface free energy theory by contact angle method. The water resistance of asphalt and asphalt-water aggregate systems was calculated in terms of stripping work. It was found that with extending aging duration, the ratio of the surface free energy component increases, resulting in greater stripping work between asphalt and aggregate. It was confirmed from Fourier Transform Infra-red spectroscopy characterization that with the aging of asphalt, carbonyl and sulfoxide functionalities increased gradually which in turn increased the polar component of asphalt, enhancing its vulnerability towards water, resulting in higher stripping off of asphalt from the aggregate with low water resistance. This effect was greatly minimized by using carbon-carbon double bond rich ZQ172 silane as a coupling agent and modifier of the aggregate, attributed to decreased polarity of the aggregate after modification. The current study based on the results of stripping of asphalt in terms of water resistance, can be of great help in selection of asphalt and suitable modifier for industrial and practical applications.

1. Introduction

Some asphalt pavements with early asphalt layer stripping off can lead to blanching and bleeding [1], which can alternatively cause severe damage to the pavement by interacting with water [2]. The extent of this damage depends on the degree of adhesion

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between asphalt and aggregate interface. A weaker adhesion can lead to serious physical damage of the pavement by water entry into the layers between asphalt and aggregates [3] as water can easily dissolve it via hydrogen bond with the electronegative atom at the limestone interface. This damage to the interface between asphalt and aggregate adhesion leads to stripping off from the aggregate surface, pavement distress and consequently affects traffic safety [4]. Apart from this, water damage to road asphalt greatly shortens the service life and maintenance cycle, causing excessive maintenance and repair costs [5]. Attributed to these factors and impacts of water damage to road materials, it is of great interest to study the mechanism of early water damage of asphalt pavement and proposing effective preventive measures. This can cause significant capital savings, road and traffic safety and extended life of road pavements [6,7].

The methods of asphalt water damage assessment are divided into qualitative and quantitative approaches [8]. But these approaches are indirect in nature and suffer from poor field reproducibility and insufficient theoretical basis [9]. Therefore, it is necessary to explore a theoretically sound method with more adequate knowledge and logic about water damage assessment and better practicability. The latest results of Lytton et al. [10] showed that the surface free energy can be used to characterize the adhesion energy of the mixture, which in turn can help in predicting the stripping extent of asphalt mixture. Due to these merits of surface free energy, greater attentions have been devoted to the characterization of adhesion between asphalt and aggregate nature [11]. In this regard, Hefer et al. [12] used adhesion theory to study the adhesion of asphalt with aggregate and declared surface energy as an important tool for predicting material properties, including determination of water damage to asphalt mixtures. Bhasin et al. [13] studied the adhesion energy of asphalt-aggregate via surface free energy and came up with the idea that energy is released when asphalt is peeled off from the aggregate surface in the presence of water. Furthermore, it was concluded that adhesion strength is highly dependent on the nature of aggregate and asphalt [14]. For example, the effect of polymer modified asphalt on water resistance of asphalt-aggregate system was studied by Jonathan et al. [15] and Iskender et al. [16]. However, up to the best of our knowledge, very little work is available on the link between surface free energy and stripping work of asphalt-aggregate and aging of asphalt-aggregate system. Hence, in this study, surface free energy related to stripping work of three aggregate (granite, diorite and limestone) and then to ten post-aging asphalt types was studied via contact angle method. Asphalt samples (both, before and after aging) were characterized for chemical composition via Fourier Transform Infra-red (FT-IR) spectroscopy.

2. Materials, reagents and instruments

The reagents used in this study were of analytical reagent grade and used without further purification. Formamide was purchased from Jinan Sheng Tong Chemical Co., Ltd. Double distilled water was used throughout the experimental procedures. The tested asphalt were SK70, Esso 70#, 70# in Maoming, Shell 70#, Shell PG76, SK- SBS, Thailand 70#, Esso PG76, Thailand modified, and Maoming SBS asphalt. Their physical characteristics are shown in Table 1. Three types of aggregates (granite, diorite and limestone) were supplied by Fuding Bohai Stone Products Co., Ltd. ZQ172 silane coupling agent was provided by Nanjing Quan Xi Chemical Co., Ltd.

The surface free energy of water and formamide (Table 2) were calculated using Eq. (1) [17].

$$\gamma_{sl} = \gamma_{lv} + \gamma_s^0 - 2\sqrt{\gamma_s^d\gamma_{lv}^d} - 2\sqrt{\gamma_s^p\gamma_{lv}^p} \quad (1)$$

where, "d" and "p" are the dispersion component and polar component of the surface energy, respectively. "γ_s" is solid surface free energy in vacuum; "γ_{sl}", "γ_{lv}" are solid-liquid and liquid-gas interfacial tension at equilibrium, respectively; while "γ_s^0" is the specific surface area of solid in vacuum.

The contact angle measuring instrument (DSA100E) was purchased from Krucss GmbH, Germany. It was equipped with high-resolution charge coupled device (CCD) camera video measurement system and high-performance digital adapter. It processed the recorded data of contact angle with time using SCA20 software.

3. Experimental

3.1. Contact angle measurement

The contact angle of distilled water on different aggregates and its change with time was determined via DSA100E. Before the test, the probe liquid was sucked into the injector and the bubbles were completely eliminated. Then the syringe was loaded into the contact angle tester. The prepared specimen was placed on an adjustable platform and the focal length and position of the specimen were adjusted for best image quality. The process of liquid drop was recorded and the data were processed using SCA20 software, measuring contact angle change with time at a rate of 10 points per second. In order to reduce the influence of surface difference of specimen on the data, three specimens were taken from each group, while each sample was tested with 3–4 points, with a total of 9–12 measurements, from which the average value was calculated. These steps are illustrated in Fig. 1(a & b) and Fig. 2 for water-diorite and asphalt-limestone samples, respectively.

3.2. Measurement and calculation of surface free energy

According to the surface energy theory, every solid surface has an unsaturated force field, which makes the surface to spontaneously attract other materials to reduce its free energy [18]. It is believed that surface free energy theory applied to asphalt-aggregate system can provide detailed information about the relationship between aging of asphalt and the adhesion work, stripping work, as well as the asphalt and aggregate water resistance [19]. When asphalt and aggregate come in contact, the aggregate spontaneously attracts asphalt molecules to reduce the asphalt-aggregate free energy, so as to achieve energy balance of the entire system. This energy exchange enables asphalt to exhibit an adhesion effect to the aggregate [20]. The theory of surface energy can be used to calculate the change in adhesion energy, cohesion energy and Gibbs free energy by measuring contact angle and surface tension which are then used to evaluate the adhesion of asphalt and aggregate [21].Surface tension is composed of a variety of intermolecular force components, including dispersion component γ_l^d and polar component γ_l^p, as related by Eq. (1):

$$\gamma_{sl} = \gamma_{lv} + \gamma_s^0 - 2\sqrt{\gamma_s^d\gamma_{lv}^d} - 2\sqrt{\gamma_s^p\gamma_{lv}^p} \quad (1)$$

According to the "Young's equation" [22]:

$$\gamma_s^0 = \gamma_{sl} + \gamma_{lv}\cos\theta \quad (2)$$

where, "θ" is the contact angle of liquid on solid surface. "γ_{sl}" and "γ_{lv}" are the parameters mentioned in Eq. (1).

By inserting Eq. (2) in Eq. (1), we get:

$$\gamma_{lv}(1+\cos\theta) = 2\sqrt{\gamma_s^d\gamma_{lv}^d} + 2\sqrt{\gamma_s^p\gamma_{lv}^p} \quad (3)$$

γ_{lv}^p and γ_{lv}^d respectively for water and formamide were calculated as shown in Table 2, which were then used to calculate contact angle of liquid on the aggregate surface by changing Eq. (3) to Eq. (4).