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Use of Surface Free Energy method to evaluate the moisture susceptibility of sulfur extended asphalts modified with antistripping agents

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HIGHLIGHTS

- Replacing of bitumen with sulfur (Googas) increased moisture susceptibility of asphalt mixtures.
- Addition of antistripping agent (NZ) improved the resistance of SEA against moisture damages.
- SFE test results were compatible with the classic mechanical test results.
- Limestone aggregate led more resistant mixtures against moist condition compared to the granite aggregate.

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ABSTRACT

Aggregate–asphalt binder adhesion bond is a vital factor that affects the resistance of asphalt mixtures against moisture damages. Since, moisture damages are the main defects of sulfur extended asphalts (SEAs), this study aims to improve it by reinforcing the adhesion between asphalt binder and aggregate. An antistripping additive named nanotechnology Zycotherm (NZ) was used to achieve this goal. Surface Free Energy (SFE) method was applied to examine the effectiveness of NZ additive in improving the moisture susceptibility of SEA. The research team utilized two common mechanical tests (indirect tensile strength, and dynamic modulus) to evaluate the validation of SFE method. All samples were constructed with two different aggregates, limestone and granite. In the SFE method, the measured surface energy components of constitutive materials were used to calculate the bond strength between them in dry and wet conditions. The findings showed that adding NZ was a successful technique to compensate the deteriorated adhesion due to using sulfur. Also it was demonstrated that SFE test results were so compatible with the common mechanical tests in predicting moisture damages.

1. Introduction

Considerable growth in bitumen cost during the last four decades has motivated the researchers to look for new ways to reduce the consumption of bitumen. One of the most welcomed alternatives was replacing the bitumen to some extent by other materials such as sulfur. At the beginning, using untreated liquid sulfur to modify HMAs led to some constructional problems like emission of hazardous gases, such as, H_2S. While applying liquid sulfur associated with some constructional problems, it had such benefits as decreasing the production cost and improving the mechanical properties of asphalt mixtures, such as stiffness [1]. So, there was a desire to enjoy the advantageous properties of sulfur, alongside eliminating its implementation problems. Eventually, a kind of dust-free sulfur pellet was developed by treating the raw sulfur with the polymeric base additives. In addition, the new physical form and chemical compound of treated sulfur made it more convenient and safe to be transported from the producer factory to the construction site. Conducted changes in the new version of sulfur product made it appropriate to be used in asphalt mixture as a bitumen extender, while, the previous problems about the emission of annoying odors and hazardous gases such as H_2S, was solved to a high extent [2]. Utilizing the new sulfur pellets reduced the mixing and compacting temperatures to about 135 °C and 90 °C respectively. Because of the changed mixing operation temperature, the new asphalt mixture goes in warm mix asphalt category. Since the mixing temperature of SEA is about 30 °C lower

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than HMAs, replacing this new asphalt mixture with the conventional HMAs leads to a high saving in production expenses, as less fuel is consumed. It is worth mentioning that temperature increment becomes more energy consuming progressively at higher temperatures, so, the 30 °C reduction in the mixing temperature could be so valuable [3]. In spite of the improvements in the mechanical properties of SEAs, there are also some negative consequences too. SEAs are more vulnerable against fatigue phenomenon and wet condition compared to the conventional asphalt mixtures [4]. Extending bitumen with sulfur increases the hardness of asphalt binder, consequently, the cohesion of asphalt binder as well as the adhesion between aggregates and binder decrease [5].There are various methods to improve these weakened properties and reduce moisture sensitivity in asphaltic mixtures. One of the main methods is the addition of antistripping agents (ASAs) which leads to a reinforced bond between asphalt binder and aggregates.

In this study, a new generation of modified sulfur pellets (Googas) and ASA (nanotechnology Zycotherm) are used in the production of warm-mix asphalt (WMA) samples.

1.1. SFE method

The main reason of SEA weakness against moisture conditions is decreased adhesion and cohesion properties of asphalt mixture ingredients. Nowadays, it is still common to use the mechanical tests in dry and wet conditions to evaluate the asphaltic mixture resistance against moisture damages. These kind of mechanical tests provide a general comprehension about the asphalt mixture behavior in moist conditions, in which, all other effective parameters, in addition to adhesion bond parameter, influence the results. Although the classic tests give a comprehensive understanding about the asphalts' behavior, they also have some deficiencies:

- Low compatibility with the real loading and pavement conditions.
- Inability to consider the constitutive material properties alongside with environmental destructive mechanisms.
- Being time consuming.

In order to eliminate these deficiencies, researchers tried to figure out a method capable of characterizing the properties of asphalt mixture's ingredients in wet condition. The new method was supposed to be able to measure the adhesion bond between aggregates and asphalt binder as well as the cohesion of binder, because they are the most determinant parameters in moisture susceptibility of asphalt mixtures. SFE parameters of asphalt binder and aggregate are significant characteristics that could be used to investigate the moisture susceptibility of asphalt mixture. In the SFE method, it is possible to measure the adhesion bond between asphalt binder and aggregates in quantitative form; also this new method helps to determine the tendency of mixture for replacing the asphalt binder with water. This replacement is the mechanism in which stripping occurs in asphalt mixtures. The relative calculations would be done according to the basic thermodynamic laws. The Obtained results provide complete information about the material properties along with the adhesion bond strength between aggregates and binders, therefore, this method would be so helpful to select the appropriate materials and their combinations in mixing design of asphaltic mixtures; so, the selected mixture would be more resistant against moisture damages [6].

1.2. The aggregate surface area (%) in contact with water

Cheng et al. [7] utilized the theory of "nonlinear Viscoelastic behavior" for substances with diffused damage. This theory which was developed by Schapery et al. [8], was used by Cheng et al. to explain the function of loaded asphalt concrete in either controlled stress or controlled strain mode. Cheng et al. applied the cyclic loading mechanism tests which were compatible with the Schapery's "distributed damages theory" criteria.

According to this new method, cyclic loading tests could be used to predict the moisture damages by measuring the aggregate surface area (P%) in contact with water in asphalt mixtures.

As shown in Eq. (1) the wet to dry ratio of asphalt dynamic modulus can be calculated if the adhesion bond values between binder and aggregates are obtained for the asphalt mixture in dry and moist conditions [9].

$$\frac{E^*_{wet}}{E^*_{dry}} = \frac{[\Delta G_{12} \times (1-P) + \Delta G_{123} \times P]}{\Delta G_{12}} \quad (1)$$

In which, ΔG_{12} is the adhesion bond energy between aggregates and asphalt binder; ΔG_{123} is the adhesion bond energy between aggregates and asphalt binder in a moist condition; P is aggregate surface area in contact with water in asphalt mixtures (%); and E^* is complex modulus in dry or wet condition.

If the dynamic modulus parameter is rewritten in the term of ratio between stress and strain $\left(\frac{\sigma}{\varepsilon}\right)$ the following relationship will be obtained when the test is conducted in constant stress mode.

$$\frac{E^*_{wet}}{E^*_{dry}} = \frac{\left(\frac{\sigma}{\varepsilon}\right)_{wet}}{\left(\frac{\sigma}{\varepsilon}\right)_{dry}} = \frac{\varepsilon_{dry}}{\varepsilon_{wet}} = \frac{[\Delta G_{12} \times (1-P) + \Delta G_{123} \times P]}{\Delta G_{12}} \quad (2)$$

In which, ε_{dry} is the applied strain in the asphalt mixture specimen under dry condition, and ε_{wet} is the applied strain in the asphalt mixture specimen under wet condition.

All the variables in this equation are known from SFE and dynamic modulus tests, so the P parameter (the aggregate surface area (%) in contact with water) is the only unknown and could be calculated from this equation.

1.3. The statement and objectives of the present study

In this experimental study, the moisture susceptibility of SEA is evaluated by SFE method and the results are compared with the indirect tensile strength (ITS) and the dynamic modulus (DM) tests for validation. Wet to dry ratios of ITS and DM tests are used to investigate the moisture susceptibility of different samples by mechanical methods, and eventually, these ratios are compared with the SFE test results. Another applied criterion for examining the moisture susceptibility and compatibility of constitutive materials is a combined method (SFE and DM) which determines the aggregate surface area (%) in contact with water per cycle of DM test (P).

The main goals of this research are:

- Evaluating the SEA moisture susceptibility and the effect of using antistripping additive (NZ) on this phenomenon.
- Comparing SFE test results with classic mechanical tests like ITS and DM to determine moisture susceptibility of asphaltic mixtures.
- Utilizing the SFE method to determine the best mix design in the production of studied SEA.

A new generation of modified sulfur mix additive (Googas) and ASA (nanotechnology Zycotherm) are used in the production of warm-mix asphalt (WMA) samples. Tests in dry and wet conditions are performed on different combinations of additives (sulfur and ASA) and aggregates.