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# Applying surface free energy method for evaluation of moisture damage in asphalt mixtures containing date seed ash

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## HIGHLIGHTS

- Moisture susceptibility highly depended on bitumen-aggregate adhesion.
- Utilization of DSA has decreased moisture damage of hot mix asphalt mixtures.
- A good correlation exists between analytical and empirical methods to evaluate moisture damage of hot mix asphalt mixtures.
- The work of adhesion between aggregate-bitumen increased with an increase in DSA content.
- Work of adhesion in presence of water is the key factor to determine moisture susceptibility of mixtures.

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## ABSTRACT

Present study deals with applying surface free energy theory in evaluating the possibility of using date seed ash as a bitumen modifier of hot mix asphalt mixtures against moisture damage. For this purpose, pure bitumen as well as ash containing samples was employed to produce mixtures having two different aggregate types (namely, limestone and siliceous). Prediction of surface energy was well supported by the laboratory method (AASHTO T-283) and proves the potential use of ash as a modifier. The achieved results indicated that using date seed ash will improve TSR of limestone and siliceous mixtures (i.e. 12.65% and 20%, respectively).

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## 1. Introduction

Moisture damage of asphalt mixtures is one of the most common damages that occur in asphalt pavements [1]. Reduced strength in asphalt mixes happens mainly as a result of reduced adhesion between aggregate particles and bitumen or decreased cohesion within the bitumen mass of the mixture [2,3]. On the other hand, stresses induced by traffic result in decreased strength in asphalt mixes which finally will lead to damages such as fatigue cracking, rutting, or bitumen bleeding in road pavements [4].

Studies of moisture damage can be performed by applying various methods, such as experimental laboratory testing methods, field tests or analytical methods. Among these, experimental techniques present suitable methods for modeling field conditions. However they do not include studies related to micro-mechanism of moisture damage [5,6]. AASHTO T-283 testing method is widely used as the most accurate experimental method for studying moisture sensitivity of asphalt mixtures. Yet, this method does not examine fundamental features of materials [7]. Surface free energy (SFE) which is explained in detail in this paper is an analytical method by which fundamental features of aggregates and bitumen is studied in twofold systems of bitumen-aggregate and also in presence of water [8].

Surface energy of a material is briefly defined as the amount of work performed to develop a unit area of new surface of a material in vacuum [9]. In the presence of moisture, total free energy of the water-aggregate-bitumen complex is reduced when bitumen is replaced by water at the bitumen-aggregate interface. This is considered a thermodynamically desirable issue resulting in moisture damage [9]. In some researches, it was shown that by decreasing surface energy of bitumen-aggregate interface in presence of water, sensitivity of asphalt mixtures to moisture damage will be increased [9,10]. Bhasin et al. evaluated the effects of additives mixed with bitumen and their impacts on energy components in the aggregate-bitumen contact surface [11].

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There are different methods of measuring surface energy of asphalt mixture components. In some researches, surface free energy of bitumen was measured by using the Wilhelmy plate method and surface free energy components of aggregates using the universal sorption device (USD) [2,9,12]. On the same subject, sessile drop method was used to measure surface energy parameters of bitumen and column wicking method was presented to calculate surface energy components of aggregates [13]. Researches show that sessile drop method also can be used to determine surface tension of different types of materials (i.e. metallic powders and mineral aggregates). For instance, Fuji calculated surface tension of silicon by sessile drop method [14]. Susana utilized sessile drop method to determine surface energy of mineral and metallic powders (i.e. Mg and Fe) [15]. Bachman also applied sessile drop method on sand and silt surfaces to determine surface energy parameters [16,17]. In another research, Kingery evaluated surface tension of silicon, iron and nickel utilizing sessile drop method [18].

Several methods have been adopted to reduce moisture damage in asphalt mixtures. A common method is to add anti-stripping additives such as hydrated lime, fly ash, cement powder and also nano-particles including nano-hydrated lime, nano-fibers, nano-clay etc. to the mixture [19–24]. With this regard, other researchers evaluated the effect of lime on moisture damage of asphalt mixtures and found that moisture susceptibility of mixes were improved [25–27]. In another research, Arabani et al. demonstrated positive effect of Zycosoil (a new nanotechnological material) as an additive to improve moisture susceptibility of warm mix asphalt mixes [28]. Fly ash has been widely used as an additive in asphalt and concrete mixtures and its effects on these have been widely investigated. Fly ash which is a waste material has revealed its amazing properties as one of the most conventional additives used in asphalt pavements. With this regard, it was shown that adding cement and fly ash to asphalt mixes will result in preventing moisture damage in mixes [4]. Other researchers showed that adding fly ash to asphalt mixtures improves mixes strength against stripping [29]. Another type of ash used in pavements is rice husk ash which is one of the byproducts obtained by burning rice's husk. Sargin et al. found that this additive can be used in hot asphalt mixtures [30].

Date is one of the oldest plants in earth [31]. Date trees grow extensively in dry and semi-dry regions such as North Africa, Arabian Peninsula, and Iran [32]. Statistics have shown that in 2013, almost ten million tons of dates were produced worldwide. From these, $1.2 \times 10^6$ tons were produced in Iran [33]. Considering that approximately 10–12% of date weight is its seed, more than $10^6$ tons of date seeds is annually wasted worldwide [32]. Hence, evaluating the feasibility of utilizing date seeds in pavements would be beneficial.

In this study, date seed ash was added to bitumen as a replacement of filler. The main objective of the work was to evaluate moisture susceptibility of HMA mixtures containing date seed ash (DSA). This was performed by assessing moisture susceptibility via laboratory and analysis approaches and the results were discussed.

## 2. The SFE theory

Surface energy is defined as the work which is required for creating a unit area of a new surface of the material in vacuum. A variety of theories have been proposed to estimate surface free energy components [34]. Although these theories are different from each other in terms of inference methods and contracts, all are commonly applied. Some of these definitions define surface energy in one component, some others define it in two components, and yet others in three components. One of the widely used theories for calculating surface energy is Van Oss–Chaudhury–Good theory which has been extensively employed by researchers to assess surface free energy in terms of three components [35]. According to this theory, surface energy based on inter-molecular and inter-surface interactions is divided into three parts, acidic or Lewis acid component ($\gamma^+$), alkali or Lewis base component ($\gamma^-$) and dispersive ($\gamma^{LW}$) component which explains non-polar interactions and also referred to Van der Waals component. The total surface free energy is obtained by combining these three mentioned components as it is seen in Eq. (1):

$$\gamma^{total} = \gamma^{LW} + \gamma^{AB} = \gamma^{LW} + 2\sqrt{\gamma^+\gamma^-} \quad (1)$$

where $\gamma^{total}$ and $\gamma^{AB}$ refer to total surface energy and acid-base component, respectively. Other terms are as described previously.

Van Oss theory is carried out for obtaining surface energy of solids using at least three probe liquids (i.e. the liquid whose surface energy is known) [35]. Fundamental equation of this theory is shown as below:

$$\Delta G_{L,S}^{adhesion} = W_{L,S}^{adhesion} = 2\sqrt{\gamma_L^{LW}\gamma_S^{LW}} + 2\sqrt{\gamma_L^+\gamma_S^-} + 2\sqrt{\gamma_L^-\gamma_S^+} \quad (2)$$

where $\Delta G_{L,S}^{adhesion}$ and $W_{L,S}^{adhesion}$ represent Gibbs free energy of adhesion and work of adhesion between solid and liquid interface, respectively. Indexes $L$ and $S$ stand for liquid and solid parts. Based on the Eq. (2), Van Oss developed the relationship between work of adhesion, surface free energy components and contact angle between liquid and solid interface $\theta$, and suggested the following equation [36]:

$$W_{L,S}^{adhesion} = \gamma_L^{total}(1+\cos\theta) = 2\sqrt{\gamma_L^{LW}\gamma_S^{LW}} + 2\sqrt{\gamma_L^+\gamma_S^-} + 2\sqrt{\gamma_L^-\gamma_S^+} \quad (3)$$

Considering Eq. (3), with substituting bitumen from solid sample (using three probe liquids) and replacing the three angles correspond to probe liquids, a three-equations, three-unknown system would be achieved. With solving the three-equations, three-unknown system, the parameters related to surface energy of bitumen can be calculated [11].

$$\gamma_L^{total}(1+\cos\theta) = 2\left[x_1\sqrt{\gamma_L^D} + x_2\sqrt{\gamma_L^-} + x_3\sqrt{\gamma_L^+}\right] \quad (4)$$

where $x_1, x_2, x_3$ are square roots of the unknown surface free energy components of bitumen.

It is to be stressed that the relations presented so far are utilized for studying the adhesion energy in two-phases of bitumen and aggregate. As the substitution of bitumen with water is thermodynamically favored, bitumen is replaced with water of hydrophilic nature. The amount of energy required to render such possible substitution can be defined as in Eq. (5) below:

$$\Delta G_{ASW}^a = \gamma_{AW} + \gamma_{SW} - \gamma_{AS} \quad (5)$$

In this equation, W, A, and S are the indexes related to water, asphalt, and aggregate particles, respectively.

## 3. Materials

### 3.1. Bitumen

Bitumen used in this research was 60–70 pen bitumen from Isfahan Refinery. Major characteristics of this bitumen are reported in Table 1.

**Table 1**
Basic physical characteristics of bitumen.

| Test | AC 60/70 | Standard |
|---|---|---|
| Penetration (25 °C) | 65 | ASTM D5 [37] |
| Softening point (°C) | 49 | ASTM D36 [38] |
| Specific gravity (g/cm$^3$) | 1.023 | ASTM D70 [39] |
| Ductility (5 cm/min) | 155 | ASTM D113 [40] |
| Flash point (°C) | 325 | ASTM D92 [41] |

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