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# Adhesion Evaluation of Asphalt-Aggregate Interface Using Surface Free Energy Method

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**Abstract:** The influence of organic additives (Sasobit and RH) and water on the adhesion of the asphalt-aggregate interface was studied according to the surface free energy theory. Two asphalt binders (SK-70 and SK-90), and two aggregate types (limestone and basalt) were used in this study. The sessile drop method was employed to test surface free energy components of asphalt, organic additives and aggregates. The adhesion models of the asphalt-aggregate interface in dry and wet conditions were established, and the adhesion work was calculated subsequently. The energy ratios were built to evaluate the effect of organic additives and water on the adhesiveness of the asphalt-aggregate interface. The results indicate that the addition of organic additives can enhance the adhesion of the asphalt-aggregate interface in dry conditions, because organic additives reduced the surface free energy of asphalt. However, the organic additives have hydrophobic characteristics and are sensitive to water. As a result, the adhesiveness of the asphalt-aggregate interface of the asphalt containing organic additives in wet conditions sharply decreased due to water damage to asphalt and organic additives. Furthermore, the compatibility of asphalt, aggregate with organic additive was noted and discussed.

**Keywords:** surface free energy; adhesion; asphalt-aggregate interface; organic additive

## 1. Introduction

The compaction temperatures of hot mix asphalt (HMA) are usually above 160 °C, which consumes a large amount of fuel energies and results in the emission of $CO_2$. Warm mix asphalt (WMA) technology has been generalized to the asphalt pavement industry for a few years. The warm mix technologies can reduce the asphalt production temperature by as much as 30 °C. There are two widely used warm mix technologies: adding organic additives and applying water foaming. In the water foaming, due to the lower compaction temperature of WMA, water cannot be completely evaporated out of aggregates. The remaining water can impact the adhesiveness of the asphalt-aggregate interface and lead to moisture damage. Therefore, many researchers start to study problems of water damage in WMA.

Currently, the theories for studying the adhesiveness of the asphalt-aggregate interface include the molecular orientation theory [1], chemical reaction theory [2], surface free energy theory [3,4] and molecular dynamics [5], etc. The surface free energy theory has been applied to research the adhesion of asphalt-aggregate interface. The theory of surface free energy could be used to evaluate water damage and fatigue cracking of HMA [6]. The surface free energies between the asphalt and aggregates were measured using the Wilhelmy plate and absorption methods, respectively, and calculated the adhesiveness of asphalt-aggregate interface with and without water, and it was feasible to use the surface free energy theory to analyze the water damage of HMA [7]. The surface free energy components of asphalt-aggregate interface were analyzed by Wilhelmy plate and adsorption methods, respectively, noting that the surface energy theory could be useful in analyzing water damage in HMA [8]. The surface free energies between the asphalt and aggregates were tested and the adhesion energy ratio was established to predict the adhesion of the asphalt-aggregate interface [9]. The evaluation of the surface energy and moisture susceptibility of various combinations of aggregates and asphalt binders were analyzed [10]. The adhesive properties could be used to estimate adhesiveness of the asphalt-aggregate interface [11]. The asphalt-aggregate interaction for moisture-induced damage mechanisms was studied using surface free energy and predicted moisture-induced damage in HMA [12]. In 2010, the surface free energies of asphalt and aggregate were tested, and the adhesion trends of asphalt-aggregate with and without water were analyzed and calculated [13]. The adhesion models of additive (SAK)-asphalt-aggregate were established and the adhesiveness was used to predict water damage of WMA [14]. The treatment of aggregate surface with hydrated lime narrows down the energy difference under dry and wet conditions, and it helps resist moisture damage [15]. Two different waxes and three kinds of aggregates were used to study the physico-chemical surface characteristics between the aggregates and asphalt. The Dynamic Contact Angle (DCA) and Dynamic Vapour Sorption Devices (DVSD) were used to measure and calculate the components of surface energy. The analysis results indicate that the waxes can adversely affect the adhesion between the aggregates and asphalt [16]. However, there are only a few studies to predict the adhesiveness of organic additive-asphalt-aggregate under the dry and wet conditions according to the surface free energy theory. In the paper, the adhesiveness of asphalt-aggregate and organic additive-asphalt-aggregate systems with and without water is studied based on the surface free energy theory.

## 2. Raw Materials and Methodology

The raw materials in this study include asphalt binder, aggregate, organic additives. Two types of asphalt binders were used: SK-70 and SK-90. The organic additives were Sasobit and RH, and the aggregates were limestone and basalt. The organic additive-modified asphalt was produced by adding 3% Sasobit and RH by weight of the base asphalt in the asphalt matrix. The sessile drop method was conducted to test the surface free energy components of base asphalts, organic additives, organic additive-modified asphalts and aggregates. After that, the adhesion models of asphalt-aggregate and organic additive-asphalt-aggregate in dry and wet conditions were established, and their adhesion works were calculated subsequently according to the surface free energy theory. Energy ratios were established to evaluate the adhesion of asphalt-aggregate and organic additive-asphalt-aggregate interfaces under dry and wet conditions. Finally, the influence of organic additives and water on asphalt-aggregate adhesion was evaluated based on the adhesion works and energy ratios obtained.

## 3. Properties of Raw Materials

### 3.1. Base Asphalt

According to the Standard Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering of China (JTG E20-2011) [17], the properties of base asphalts, SK-70 and SK-90, were measured, and the results are shown in Figure 1. In addition, three replicates for each test was adopted in this paper.

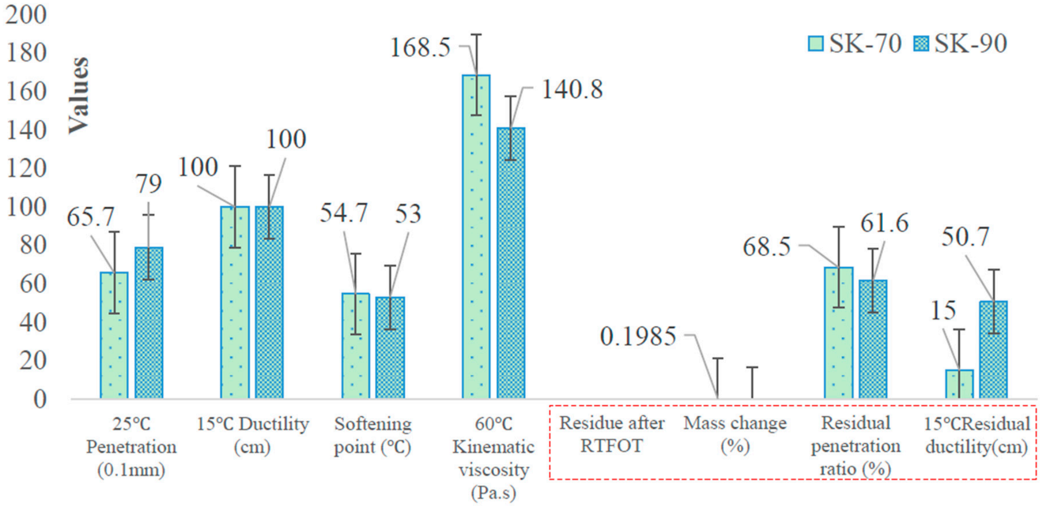

**Figure 1.** Properties of SK-70 and SK-90 asphalt (RTFOT: Rolling Thin Film Oven Test).

The Performance Grade (PG) classifications of SK-70 and SK-90 were evaluated based on the results of the Dynamic Shear Rheometer (DSR) and Bending Beam Rheometer (BBR) tests of the Strategic Highway Research Program (SHRP). The PG of SK-70 and SK-90 are determined to be PG64-24.

### 3.2. Organic Additives

Sasobit is a WMA product of Sasol Wax, located in South Africa (CAS number: 8002-74-2). "RH" is a kind of WMA additive, which is developed by the Research Institute of China Highway Ministry of Transport. The properties of Sasobit and RH were analyzed, and the test results are shown in Figure 2.

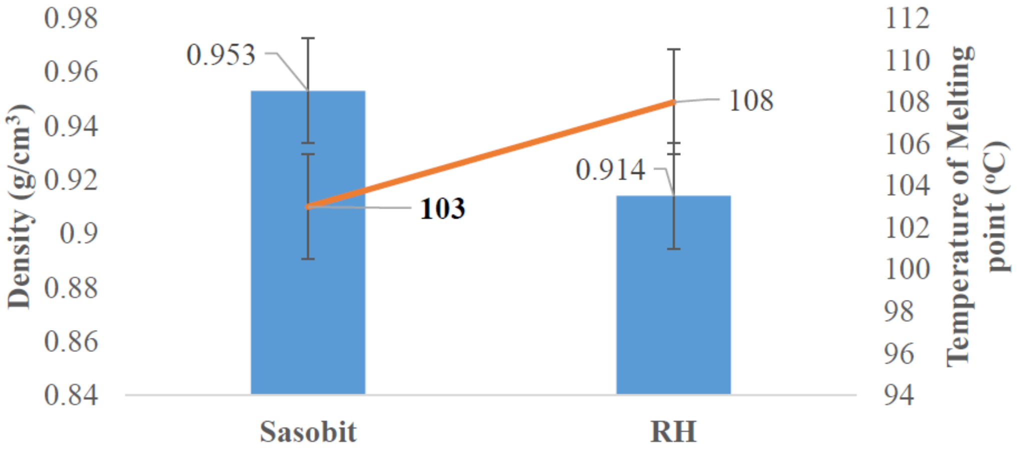

**Figure 2.** Properties of Organic Additives.

### 3.3. Organic Additive-Modified Asphalt

Four modified asphalts are processed by adding 3% Sasobit and RH (by mass of asphalt) into base asphalts, respectively. Sasobit and RH organic waxes can be dissolved easily into base asphalts at a temperature above 100 °C. In this paper, Sasobit and RH were blended into base asphalts at a temperature of 120 °C and stirred manually for 15 min. The properties of organic additive-modified asphalts were tested. The test results are shown in Figure 3.

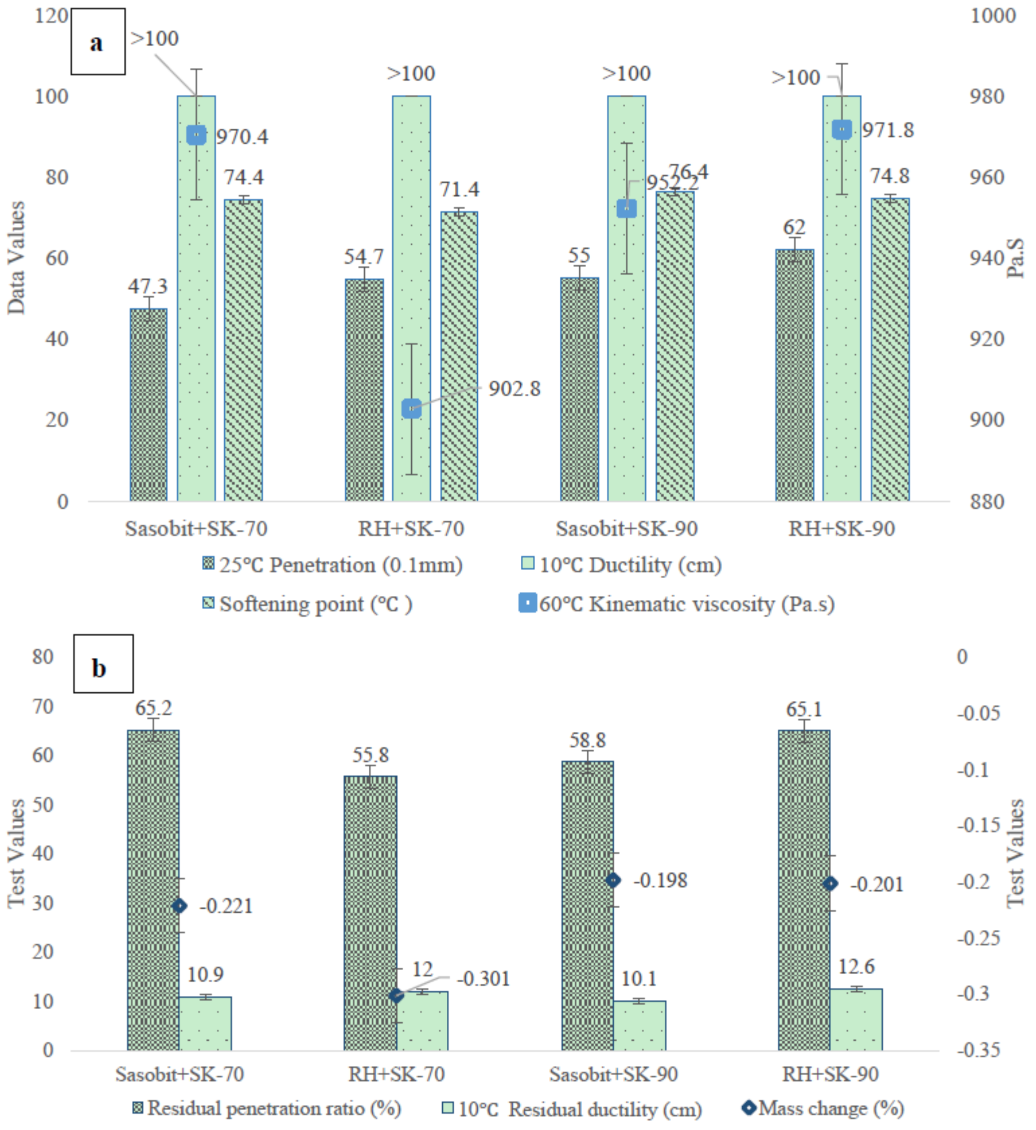

**Figure 3.** Properties of Organic Additive-Modified Asphalts, (**a**) Penetration, Ductility, Softening Point, Kinematic Viscosity of asphalt binders; (**b**) Residual Penetration Ratio, Residual ductility, and Mass Change of asphalt binders.

PG classifications of the four organic additive-modified asphalts are tested according to the DSR and BBR tests of SHRP, and presented in Table 1.

**Table 1.** PG Classifications of Organic Additive-Modified Asphalts.

| Items | SK-70 and SK-90 | Sasobit + SK-70 | RH + SK-70 | Sasobit + SK-90 | RH + SK-90 |
|---|---|---|---|---|---|
| PG | PG 64-24 | PG64-24 | PG64-18 | PG64-24 | PG64-24 |

### 3.4. Aggregates

According to (JTG E42-2005) the Standard Test Methods of Aggregate for Highway Engineering in China [18], the properties of limestone and basalt aggregates are tested. The specific gravities of limestone and basalt aggregates are 2.667 and 2.655, respectively.

### *3.5. Surface Free Energy of Raw Materials*

There are many methods for testing the surface free energy of different materials, including the capillary method, rings method, drop weight method, Wilhelmy plate method, sessile drop method, atomic force microscopy and the nuclear magnetic resonance method. Little and Bhasin [19] measured the surface free energy components of asphalt pavement materials using the sessile drop method. Murat Koc et al. presented a sessile drop device for measuring the surface energy components of both asphalt binders and aggregates [20,21]. In this manuscript, a sessile drop device is employed to measure contact angles on the surface of raw materials. The surface energy components of raw materials are calculated using the measured contact angles.

The test liquids are distilled water, glycerin and formamide. The surface free energy components [22] of all test liquids are known in advance, as shown in Figure 4.

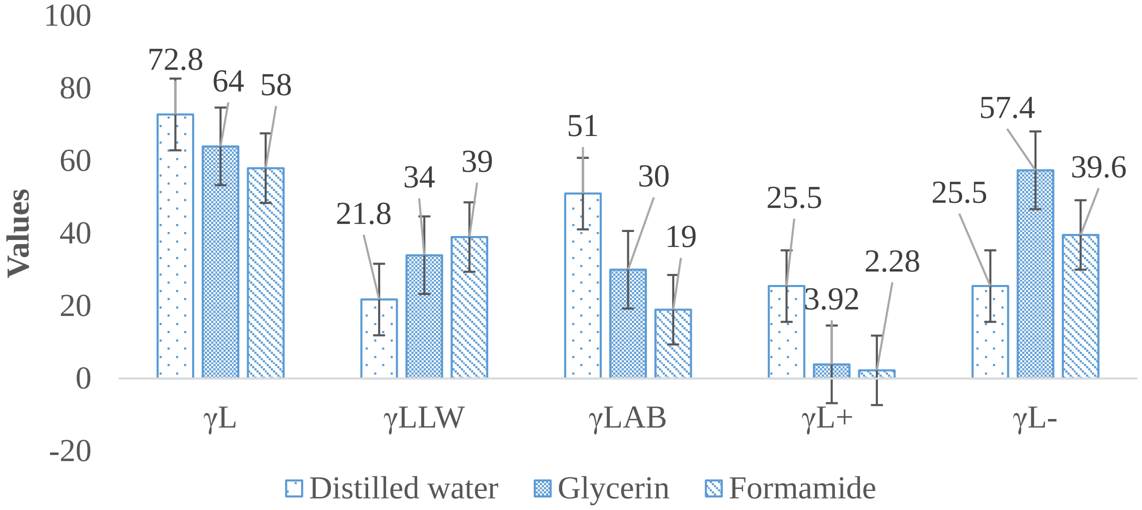

**Figure 4.** The Surface Free Energy Components of Test Liquids (mJ/m$^2$) (Note: γL—the surface free energy of test liquid; γL$^{LW}$—the nonpolar part of the surface free energy of test liquid; γL$^{AB}$—the polar part of the surface free energy of test liquid; γL$^+$—the acidic effect part of the surface free energy of test liquid; γL$^-$—the basic effect part of the surface free energy of test liquid.).

The sessile drop method was conducted to test contact angles between the surface of the raw materials and test liquids. Based on the simplified matrix formula of the Young-Dupre Equation (see Equation (1)) [23,24] the surface free energy components of raw materials are calculated, as shown in Table 2.

$$\begin{bmatrix} \sqrt{\gamma_{L1}^{LW}} & \sqrt{\gamma_{L1}^{+}} & \sqrt{\gamma_{L1}^{-}} \\ \sqrt{\gamma_{L2}^{LW}} & \sqrt{\gamma_{L2}^{+}} & \sqrt{\gamma_{L2}^{-}} \\ \sqrt{\gamma_{L3}^{LW}} & \sqrt{\gamma_{L3}^{+}} & \sqrt{\gamma_{L3}^{-}} \end{bmatrix} \begin{bmatrix} \sqrt{\gamma_a^{LW}} \\ \sqrt{\gamma_a^{-}} \\ \sqrt{\gamma_a^{+}} \end{bmatrix} = \begin{bmatrix} \frac{\gamma_{L1}(1+\cos\theta_1)}{2} \\ \frac{\gamma_{L2}(1+\cos\theta_2)}{2} \\ \frac{\gamma_{L3}(1+\cos\theta_3)}{2} \end{bmatrix} \quad (1)$$

where $\gamma_{L1}$, $\gamma_{L2}$, $\gamma_{L3}$—the surface free energy of distilled water, glycerin, and formamide, respectively; $\gamma_{L1}^{LW}$, $\gamma_{L2}^{LW}$, $\gamma_{L3}^{LW}$—the nonpolar part of surface free energy of distilled water, glycerin, and formamide, respectively; $\gamma_{L1}^{+}$, $\gamma_{L2}^{+}$, $\gamma_{L3}^{+}$—the acidic effect part of surface free energy of distilled water, glycerin, and formamide, respectively; $\gamma_{L1}^{-}$, $\gamma_{L2}^{-}$, $\gamma_{L3}^{-}$—the basic effect part of surface free energy of distilled water, glycerin, and formamide, respectively; $\theta_1$, $\theta_2$, $\theta_3$—the contact angle between raw material and distilled water, glycerin, and formamide, respectively; the rest of the parameters are the same as above.

**Table 2.** Surface free energy components of raw materials (mJ/m$^2$).

| Items | $\gamma_a$ | $\gamma_a{}^{LW}$ | $\gamma_a{}^{AB}$ | $\gamma_a{}^{+}$ | $\gamma_a{}^{-}$ |
|---|---|---|---|---|---|
| SK-70 | 25.570 | 25.541 | 0.028 | 0.000 | 4.416 |
| SK-90 | 21.512 | 21.082 | 0.430 | 0.016 | 2.915 |
| Sasobit | 40.210 | 38.370 | 1.840 | 0.129 | 6.543 |
| RH | 26.94 | 21.63 | 5.32 | 1.63 | 4.34 |
| Sasobit + SK-70 | 17.353 | 14.709 | 2.643 | 2.023 | 0.863 |
| Sasobit + SK-90 | 24.150 | 24.144 | 0.006 | 0.419 | 0.000 |
| RH + SK-70 | 16.75 | 15.10 | 1.65 | 0.21 | 3.22 |
| RH + SK-90 | 13.17 | 9.91 | 3.26 | 1.60 | 1.65 |
| Limestone | 48.351 | 46.427 | 1.924 | 0.108 | 8.603 |
| Basalt | 53.140 | 52.016 | 1.123 | 2.313 | 0.136 |

The method proposed by Fwoke is applied to further verify the validity of the test methods and results [25,26]. Fwoke pointed out that there is a good linear relationship between $\gamma_L \cos\theta$ and $\gamma_L$ of test liquid. If the correlation coefficient is higher than 0.95, it indicates that the test method and results are effective. However, when the correlation coefficient is lower than 0.95, the test method and results are ineffective. In this paper, the correlation coefficients of $\gamma_L \cos\theta$ and $\gamma_L$ of test liquid are all above 0.95, which indicates that using the sessile drop method to test the surface free energy components of raw materials is feasible.

## 4. Adhesion of Asphalt-Aggregate Interface

### 4.1. Adhesion Models of the Asphalt-Aggregate Interface

Adhesion models of the asphalt-aggregate interface in different conditions are established respectively based on surface free energy theory [27–29]. The adhesion model of asphalt-aggregate without water (dry condition) is established, and is as shown in Equation (2).

$$W_{as} = 2(\sqrt{\gamma_a^{LW}\gamma_s^{LW}} + \sqrt{\gamma_a^{+}\gamma_s^{-}} + \sqrt{\gamma_a^{-}\gamma_s^{+}}) \tag{2}$$

where $W_{as}$ denotes the adhesion of the asphalt-aggregate interface, respectively. The adhesion model of the organic additive-asphalt-aggregate system without water (dry condition) is also established as follows:

$$W_{ase} = 2(\gamma_e^{LW} + 2\sqrt{\gamma_e^{+}\gamma_e^{-}}) - 2\sqrt{\gamma_a^{LW}\gamma_e^{LW}} + \sqrt{\gamma_a^{+}\gamma_e^{-}} + \sqrt{\gamma_a^{-}\gamma_e^{+}} - 2(\sqrt{\gamma_s^{LW}\gamma_e^{LW}} + \sqrt{\gamma_s^{+}\gamma_e^{-}} + \sqrt{\gamma_s^{-}\gamma_e^{+}}) \tag{3}$$

where $W_{ase}$ denotes the adhesion of organic additive-asphalt-aggregate without water; the rest of the parameters are the same as above. The adhesion model of asphalt-aggregate with water (wet condition) can be written as follows:

$$\begin{aligned} W_{asw} = \; & -(2\sqrt{\gamma_a^{LW}\gamma_w^{LW}} + 2\sqrt{\gamma_s^{LW}\gamma_w^{LW}} + 2\sqrt{\gamma_w^{+}}(\sqrt{\gamma_a^{-}} + \sqrt{\gamma_s^{-}}) \\ & +2\sqrt{\gamma_w^{-}}(\sqrt{\gamma_a^{+}} + \sqrt{\gamma_s^{+}}) - 2\gamma_w^{LW} - 2\sqrt{\gamma_a^{LW}\gamma_s^{LW}} - 4\sqrt{\gamma_w^{+}\gamma_w^{-}} \\ & -2\sqrt{\gamma_a^{+}\gamma_s^{-}} - 2\sqrt{\gamma_a^{-}\gamma_s^{+}}) \end{aligned} \tag{4}$$

where $W_{asw}$ denotes the adhesion energy of asphalt-aggregate with water; the rest of the parameters are the same as above. The adhesion model of organic additive-asphalt-aggregate with water (wet condition) is built and is expressed as:

$$
\begin{aligned}
W_{asew} = \quad & -(4\sqrt{\gamma_a^{LW}\gamma_e^{LW}} + 4\sqrt{\gamma_s^{LW}\gamma_e^{LW}} - 4\sqrt{\gamma_e^{LW}\gamma_w^{LW}} - 2\sqrt{\gamma_a^{LW}\gamma_s^{LW}} \\
& -2\sqrt{\gamma_a^{LW}\gamma_w^{LW}} - 2\sqrt{\gamma_s^{LW}\gamma_w^{LW}} + 2\sqrt{\gamma_e^{+}}(\sqrt{\gamma_a^{-}} + \sqrt{\gamma_s^{-}}) \\
& +2\sqrt{\gamma_e^{-}}(\sqrt{\gamma_a^{+}} + \sqrt{\gamma_s^{+}}) - 2\sqrt{\gamma_w^{+}}(\sqrt{\gamma_a^{-}} + \sqrt{\gamma_s^{-}} + 2\sqrt{\gamma_e^{-}}) \\
& -2\sqrt{\gamma_w^{-}}(\sqrt{\gamma_a^{+}} + \sqrt{\gamma_s^{+}} + 2\sqrt{\gamma_e^{+}}) + 2\gamma_w^{LW} - 2\gamma_e^{LW} \\
& +4\sqrt{\gamma_w^{+}\gamma_w^{-}} - 4\sqrt{\gamma_e^{+}\gamma_e^{-}} - 2\sqrt{\gamma_a^{+}\gamma_s^{-}} - 2\sqrt{\gamma_a^{-}\gamma_s^{+}} + 2\sqrt{\gamma_a^{+}\gamma_e^{-}} \\
& +2\sqrt{\gamma_a^{-}\gamma_e^{+}} + 2\sqrt{\gamma_s^{+}\gamma_e^{-}} + 2\sqrt{\gamma_s^{+}\gamma_e^{-}})
\end{aligned}
\tag{5}
$$

where $W_{asew}$ denotes the adhesion of organic additive-asphalt-aggregate with water; the rest of the parameters are the same as above.

### 4.2. Energy Ratios

Energy ratios ($EP_1$ and $EP_2$) are built using the adhesive properties of the asphalt-aggregate interface in different conditions. $EP_1$ is used to evaluate the adhesion of the asphalt-aggregate interface affected by water, and is calculated using Equation (6):

$$EP_1 = \frac{W_{as}}{W_{asw}} \; or \; \frac{W_{ase}}{W_{asew}} \tag{6}$$

When the $EP_1$ value is higher than 1, the adhesive property of the asphalt-aggregate interface with water is lower than that of the asphalt-aggregate interface without water. It indicates that water has a negative influence on the adhesion of the asphalt-aggregate interface. When the $EP_1$ value is equal to 1, it predicts that water has no interaction with the asphalt-aggregate interface. When the $EP_1$ value is lower than 1, the adhesion of the asphalt-aggregate interface with water is greater than that of the asphalt-aggregate interface without water. It predicts that water promotes adhesion in the asphalt-aggregate interface. $EP_2$ is used to characterize the adhesion of the asphalt-aggregate interface affected by organic additive, and is calculated using Equation (7):

$$EP_2 = \frac{W_{as}}{W_{ase}} \; or \; \frac{W_{asw}}{W_{asew}} \tag{7}$$

When the $EP_2$ value is greater than 1, the adhesion of the organic additive-asphalt-aggregate is lower than that of the asphalt-aggregate. It means that organic additive has a negative influence on the adhesion of the asphalt-aggregate interface. When the $EP_2$ value equals 1, the organic additive has no influence on the adhesion of the asphalt-aggregate. When the $EP_2$ value is less than 1, the adhesion of the organic additive-asphalt-aggregate with or without water is higher than that of asphalt-aggregate with or without water. It indicates that organic additives improve the adhesion of asphalt-aggregate.

### 4.3. Adhesion of Asphalt-Aggregate

The adhesion energies of asphalt-aggregate interfaces in different conditions are calculated, and the results are shown in Figure 5. In the dry condition, it was observed that the adhesion of the asphalt-aggregate interface of asphalt containing organic additives was higher than that of the base asphalt. This is because the surface free energy of organic additives modified asphalts is lower than that of base asphalt. As a result, the stability of organic additive-asphalt-aggregate is greater than that of asphalt-aggregate. Therefore, the surface free energy of asphalt can be reduced by adding organic additives so that the adhesion of asphalt-aggregate interface in a dry condition can be enhanced. In the wet condition, it was observed that the adhesion of the base asphalt and the asphalt containing organic additives decreased significantly. The reason for this is that water has a higher surface energy that prevents effective bonding between asphalt and aggregate. The asphalt-aggregate adhesive energy of the base asphalt, the Sasobit-modified asphalt, and the RH-modified asphalt, was reduced by 10.8%, 47.9% and 32.9% on average, respectively. This indicates that water has a great influence on the adhesion of asphalt-aggregate. In addition, it was found that water has a greater effect on the asphalt-aggregate adhesion of the asphalt containing organic additives compared to that of the base asphalt. This indicates that after the addition of organic additives, moisture damage can be more severe.

In fact, previous studies have shown that asphalt mixes containing organic additives have a greater moisture susceptibility than the conventional asphalt mix [30]. This is due to the hydrophobic characteristics of organic additives. As a result, the asphalts containing organic additives refuse to form effective bonding between asphalt and the aggregate surface. The comparison between the two organic additives showed that the asphalt containing Sasobit is more susceptible to moisture damage than the asphalt containing RH. The comparison between the two asphalt binders showed that the SK-70 exhibited greater adhesion than the SK-90. This may be because of the higher surface energy of SK-70, as shown in Table 2. In addition, it was found that the SK-70 is more resistant to water damage than the SK-90. The average reduction in adhesion of SK-70-aggregate and SK-90-aggregate interfaces were 33.4% and 27.7%, respectively. The comparison between the two types of aggregates showed that the adhesion of the limestone-asphalt interface was lower than that of the basalt-asphalt interface. This can also be attributed to the higher surface free energy of basalt. However, it was found that the limestone-asphalt interface was more resistant to water damage than the basalt-asphalt interface. The average reductions in adhesion of limestone-asphalt and basalt-asphalt interfaces were 27.0% and 34.1%, respectively.

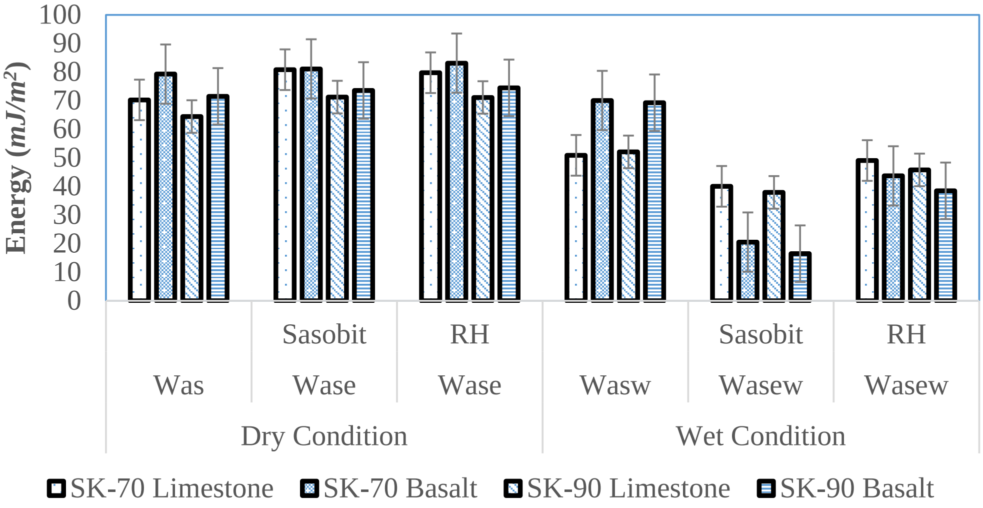

**Figure 5.** Adhesion of the Asphalt-Aggregate Interface in Different Conditions (mJ/m$^2$).

### 4.4. *Adhesion of the Asphalt-Aggregate Interface*

The results of energy ratios ($EP_1$ and $EP_2$) are presented in Figure 6. As mentioned above, the $EP_1$ value is an indication of how water impacts the adhesion of an asphalt-aggregate or organic additive-asphalt-aggregate interface. All the $EP_1$ values were greater than 1, indicating that the adhesive properties of asphalt-aggregate or organic additive-asphalt-aggregate reduce with residual water in the asphalt mixture, and thus the adhesion energy of asphalt-aggregate or organic additive-asphalt-aggregate in a wet condition declines. In addition, the adhesion energy of asphalt-aggregate or organic additive-asphalt-aggregate in a wet condition decreases more rapidly, when the $EP_1$ value becomes higher. It can be found that the $EP_1$ values of organic additive-asphalt-aggregate were always higher than that of asphalt-aggregate. It indicates that the adhesion of organic additive-asphalt-aggregate in the wet condition is far lower than that of asphalt-aggregate with water. This means that the organic additive has significantly negative influences on asphalt-aggregate interface adhesion. Because the two organic additives are also organic waxes, it can form an isolating layer to prevent aggregate from absorbing asphalt when the aggregate interface contains some residual water.

Therefore, the organic additive lowers the adhesiveness of asphalt-aggregate sharply in a wet condition. The $EP_1$ value of asphalt-basalt in a dry condition is less than that of asphalt-limestone, which indicates that the adhesiveness of asphalt-basalt in a dry condition is relatively better. However, the $EP_1$ value of asphalt-basalt in a wet condition is greater than that of asphalt-limestone, indicating that limestone has a positive influence on the adhesiveness of asphalt-aggregate in a wet condition; or limestone is less sensitive to water; or that limestone has a good compatibility with asphalt. The compatibility between asphalt and aggregate should therefore be studied and improved to reach a high adhesion energy of the asphalt-aggregate interface.

The $EP_2$ value indicates that organic additives impacted the adhesiveness of asphalt-aggregate. All the $EP_2$ values of asphalt-aggregate or organic additive-asphalt-aggregate in dry conditions were lower than 1. However, all the $EP_2$ values of asphalt-aggregate or organic additive-asphalt-aggregate in wet conditions were higher than 1. Once there is some residual water in the asphalt mixture, the organic additive has a negative influence on the adhesion of asphalt-aggregate. It can also be concluded that the organic additive is very sensitive to water and has hydrophobic characteristics. The adhesion of organic additive-asphalt-aggregate in wet conditions can be reduced due to the dual action of water and organic additive. For preventing water damage to WMA, the residual water in aggregate should be excluded. The $EP_2$ value of Sasobit-asphalt-aggregate is higher than that of RH-asphalt-aggregate, in either the dry or wet condition, which indicates that Sasobit is more sensitive to water and decreases the adhesiveness of asphalt-aggregate. For ensuring the high adhesion of the asphalt-aggregate interface, the compatibility of asphalt, aggregate and organic additive should be strictly observed.

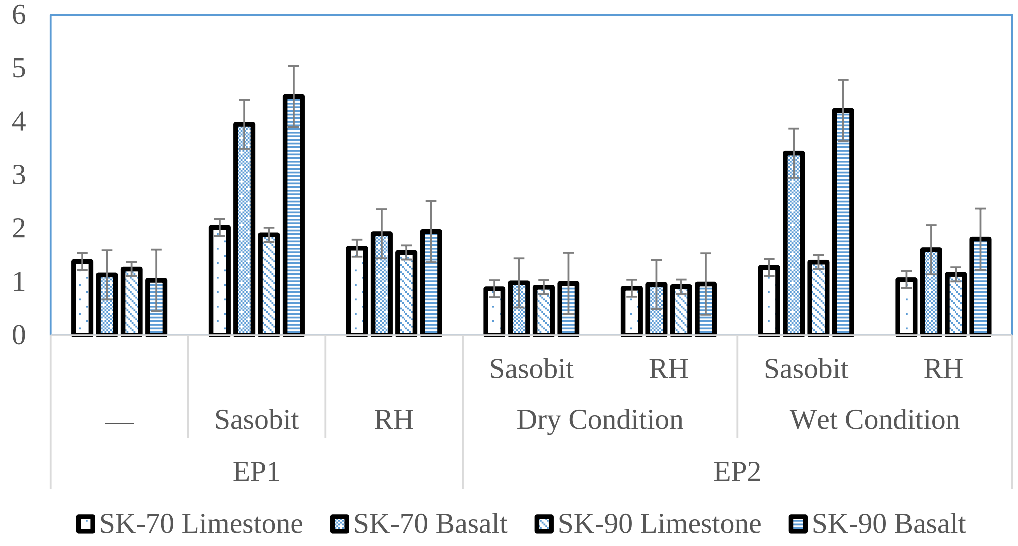

**Figure 6.** The $EP_1$ and $EP_2$ Values of Asphalt-Aggregate Interface in Different Systems.

## 5. Conclusions

The effect of organic additives on the strength of adhesion between the aggregate and asphalt was investigated. The components of the surface free energy of materials were tested and calculated. The surface energy test was used to evaluate the adhesive strength. Based on the test results and analysis, the conclusions can be obtained.

(1) Energy ratio values ($EP_1$ and $EP_2$) can be used to estimate the adhesiveness of asphalt-aggregate affected by water or organic additive. When $EP_1$ and $EP_2$ values increase, the adhesion of the asphalt-aggregate interface influenced by water or organic additive decreases.

(2) Organic additives improve the adhesiveness of asphalt-aggregate interface in dry conditions since the organic additives have hydrophobic characteristics and high surface free energy, although the adhesiveness of the asphalt-aggregate interface in a wet conditions decreases dramatically.

(3) The properties of asphalt and aggregate have some negative or positive impacts on the adhesion of asphalt-aggregate. If the asphalt and aggregate have a good compatibility, the adhesiveness of the asphalt-aggregate interface can be promoted.

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**Author Contributions:** Jie Ji, Hui Yao and Zhi Suo conceived the experiments; Luhou Liu and Peng Zhai performed the experiments; Jie Ji and Hui Yao wrote the paper; Xu Yang and Zhanping You helped revise the paper.

**Conflicts of Interest:** The authors declare no conflict of interest.

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