

A Precise Evaluation Method for Adhesion of Asphalt Aggregate

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Abstract: Based on the theory of surfaces and interfaces, this paper puts forward a more precise method to test the adhesive performance of asphalt and aggregate and design relevant experimental procedures. First, the relationship between the adhesive performance and the adhesive bond of asphalt and aggregate is discussed. The Van der Waals interaction plays a dominant role in the analysis of three micro-interaction/Van der Waals interactions, hydrogen bonds, and chemical bonds. A simplified maximum adhesive bond equation is deduced, which is related to the Van der Waals interaction. By analyzing asphalt and aggregate interface bond strength formation mechanisms, the wetting of asphalt on aggregate has a great effect on the adhesive bond based on the wetting theory. The adhesive work, known as an important wetting parameter, is taken as the index to represent the adhesive performance of asphalt and aggregate. The test method with two parameters, the contact angle θ , which is asphalt on aggregate, and the asphalt surface tension γ , is designed. Through tests of four types of asphalt and two types of aggregate, the results show that the adhesive work test method is characterized by clear theory, good feasibility and practicability, and high accuracy, compared with scanning electron microscope (SEM) photograph tests and boiling tests.

Key words: Adhesion; Adhesive work; Angle of contact; Asphalt mixture; Road engineering; Surface tension.

Introduction

The total length of highway increases rapidly in China with the developing transportation infrastructure. Asphalt pavement moisture damage is critical in China; it reduces the level of service and structural stability of asphalt pavement. Distress and corrosion of a significant amount of pavement, largely due to moisture destruction, is an indication of the significance and the severity of the problems, which include premature rutting, raveling, and wear [1]. Research regarding moisture damage has attracted world-wide attention [2, 3]. At present, the water boiling test (ASTM 3625) is the only method in China that is used to measure adhesive performance of asphalt and aggregate. However, the test method has many disadvantages [4]. First, test operation is a problem because the slight boiling condition is difficult to control; second, the test results are measured by eye. This paper, based on the theory of surfaces and interfaces, discusses adhesive work, an important wetting parameter, and the possibility of finding an index to represent the adhesive performance of asphalt and aggregate.

Relationship between Wetting and Adhesion of Asphalt and Aggregate

Studies show that there is good correlation between the adhesive strength of asphalt and aggregate and adhesive performance [5]. As

the adhesive strength increases, the asphalt mixture becomes more resistant to moisture damage [6]. There are three micro-interactions between asphalt and aggregate: the Van der Waals interaction, hydrogen bonds, and chemicals bond; each has different effects. The Van der Waals interaction is dominant among these interactions [7]. Thus, the Van der Waals adhesion interface theory represents the adhesion strength of asphalt and aggregate. Therefore, the maximum adhesion strength [8] is deduced as follows:

$$F_{\max} = \frac{\pi n_a n_s A_{as}}{3^{5/2} l_{as}^3} \cdot S_{as} \quad (1)$$

where n_a is the number of basic particles in a unit volume of asphalt; n_s is the number of basic particles in a unit volume of aggregate; A_{as} is a constant of dispersive action; l_{as} is the distance of asphalt and aggregate; and S_{as} is the adhesive area.

According to Eq. (1) and modern microanalysis techniques, testing these parameters is impossible; thus, it is difficult to calculate adhesive strength directly. However, the equation indirectly shows that the asphalt and aggregate are given and that the adhesive strength is directly proportional to S_{as} and inversely proportional to l_{as}^3 (i.e., the adhesive strength is greater, if l_{as} is smaller and/or S_{as} is larger).

Based on wetting theory, improving the wetting ability of asphalt on aggregate will help asphalt immerge and wet the texture (holes and cracks) on the aggregate surface to observably reduce l_{as} and increase S_{as} , which improves interface adhesion. The strong asphalt and aggregate bond forms at room temperature. Through the above analysis, the wetting performance is taken as an index of adhesive strength because they are directly proportional.

Performance of Asphalt Wetting on Aggregate

According to surface and interface physical chemistry, the adhesive work of a liquid on a solid surface represents the wetting performance.

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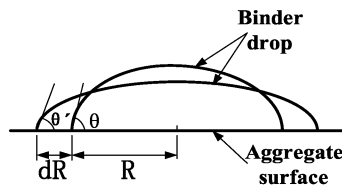


Fig. 1. Cross Section of Interface of Binder and Aggregate.

The adhesion work is used to describe the Helmholtz free energy reduction in which asphalt adheres to aggregate. In other words, the adhesion work is described such that the work is required to separate the asphalt from the aggregate surface. A description [8, 9] is given as:

$$W_{as} = r_a + r_s - r_{as} \quad (2)$$

where W_{as} is the adhesion work; r_a is surface tension of asphalt; r_s is the aggregate surface tension; and r_{as} is the interfacial tension between the asphalt and aggregate.

The adhesion work is a precise measure of wetting performance, but it is not appropriate for describing solid surface tension and interface tension.

Fig. 1 shows the spread process of a binder drop on an aggregate surface. θ is the initial contact angle of the binder on the aggregate, and θ' is the contact angle at equilibrium. R is the initial radius of binder drop, which later increases by dR . The area of asphalt liquid drop increases $\Delta A = 2\pi R dR$; thus, the change of surface energy is:

$$\Delta G^s = 2\pi R dR(\gamma_{as} - \gamma_s) + 2\pi R dR \gamma_a \cos \theta' \quad (3)$$

When the spreading process reaches the equilibrium state,

$$\lim_{dR \rightarrow 0} \frac{\Delta G}{2\pi R dR} = 0 \quad (4)$$

When $dR \rightarrow 0$, θ' is equal to θ because $\Delta\theta \rightarrow 0$. Then Eq. (3) becomes:

$$\gamma_{as} - \gamma_s + \gamma_a \cos \theta = 0 \quad (5)$$

When Eqs. (2) and (5) are combined to eliminate r_{as} and r_s , the adhesion works becomes:

$$W_{as} = \gamma_a(1 + \cos \theta) \quad (6)$$

By Eq. (6), it can be concluded that the adhesion will be calculated by measuring the surface tension and asphalt contact angle with the aggregate. Compared with solid surface tension and

interfacial tension, it is easy to measure surface tension and the contact angle of liquid.

Adhesion Work Test Method

The choice of test temperature is very important. Asphalt viscosity at low temperature is too high to achieve the equilibrium state and measure the contact angel. Therefore, the test temperature should be comparably high, such as the hot mix temperature or close to the hot mix temperature. A temperature of 130°C was used as the test temperature.

Testing asphalt surface tension is simple. The asphalt specimen was kept at the test temperature for half an hour. The surface tension was measured with an automatic digital surface tension device that includes an environment chamber.

Determining the asphalt contact angle of the aggregate test is more complicated. Generating mineral slices is a key process that greatly affects the test results. The big stone is cut into pieces with dimensions of 1.5(W)×3(L)×1cm(H) because the environment chamber in the contact angle device is very small. The plane quality and roughness of all slices must be uniform to determine how these stone pieces affect the contact angles. A series of sandpapers is used to abrade the stone surface to modify the surface from coarse to smooth.

After the mineral slices are completed, all materials (including the asphalt, slices, and glass stick) are kept in a 130°C test oven for at least half an hour. Then the asphalt drop drips on the slice surface using the glass stick. The asphalt drop spreads, and the radius increases. The equilibrium state is achieved when the radius remains constant. The spreading time to the equilibrium state is 60 seconds. After spreading and reaching the equilibrium state, the asphalt contact angel can be measured with a POWEREACHTM JC2000A automatic contact angle device.

Property of Raw Materials

There are four types of binder available for testing: RA is 70# direct distillation asphalt; MA is asphalt with an additional 0.3% (asphalt weight) interface agent [10] added to RA; RPA is 5% (asphalt weight) SEBS (a new type of modified block copolymer, Styrene-butadiene block copolymer (SBS)) modified asphalt (with an RA base asphalt); and MPA is asphalt with an added 0.3% (asphalt weight) interface agent to RPA. Basalt and granite are two aggregates used in the experiment. Tables 1 and 2 summarize the chemical composition and physical properties of these aggregates, while Table 3 summarizes the physical properties of the binders.

Table 1. Chemical Composition of Aggregate (in Weight Percent, %).

Type	SiO ₂	Al ₂ O ₃	MgO	CaO	TiO ₂	K ₂ O	Fe ₂ O ₃	Total
Granite	82.8	7.3	4.7	4.2	-	-	0.8	99.8
Basalt	52.4	18.3	4.0	7.7	1.2	4.0	10.5	98.1

Table 2. Physical Properties of Aggregate.

Type	Soundness	L.A. Abrasion	Crush Value	Impact Value	Water Absorption	Acicular Content	Mud Content
Granite	1.3	18.5	18.3	16.4	0.9	8.9	0.3
Basalt	0.8	9.8	10.6	9.0	0.8	4.7	0.3

Table 3. Physical Properties of Binders.

Type	Penetration/0.1mm (25°C)	Ductility/cm		Separation Test/°C	Elasticity Recovery/%	$T_{R\&B}$ */°C	PI*
		10°C	25°C				
RA	72	15	89	-	14	49	-0.57
RPA	60	41	>120	2	62	59	1.30

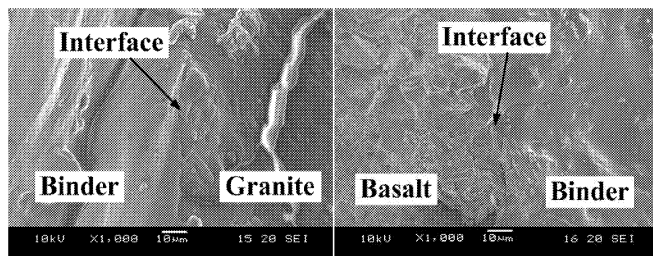
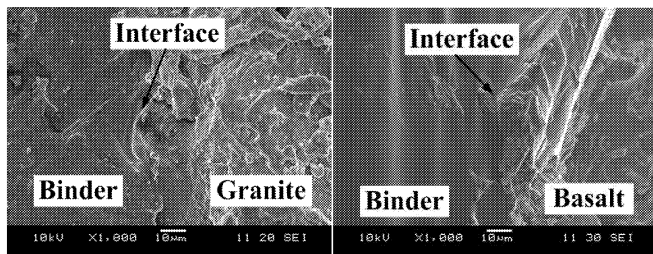
* $T_{R\&B}$: soft point of asphalt tested by “Ring and Ball” method; PI: penetration index.

Table 4. Boiling Test Results.

Aggregate Type	RA	MA	RPA	MPA
Granite	3	5	4	5
Basalt	4	5	5	5

Table 5. Results of Adhesive Work.

Binder	Surface Tension/ $10^{-3}Nm^{-1}$	Contact Angle/°		Adhesive Work/ $10^{-3}Nm^{-1}$	
		Granite	Basalt	Granite	Basalt
RA	32.2	32	25	59.50	61.38
MA	32.4	24	19	62.00	63.03
RPA	36.2	44	37	62.24	65.11
MPA	36.0	35	29	65.49	67.49

**Fig. 2.** Interface Topography of RA on Granite and Basalt.**Fig. 3.** Interface Topography of MA on Granite and Basalt.

Experiment Results and Analyses

The boiling test, a scanning electron microscope (abbreviated to SEM), and an adhesion work test are performed to evaluate their advantages and disadvantages.

Boiling Test Results

Table 4 shows the boiling test results of four binders and two aggregates. The basalt results are better than those of granite.

The adhesive level on granite depends on the binder. There is only about 70% RA asphalt film left on the granite particle surface (level 3); this is the lowest among the different binders; most of the RPA film on granite remains, meanwhile MA and MPA asphalt film remain intact to achieve level 5 after boiling conditioning.

The adhesive level of all binders on basalt gets to level 4 or higher. That of base asphalt RA is level 4, in which a very small amount of asphalt film is peeling. Modified asphalt and/or an interface agent improve adhesion performance such that they can achieve level 5.

Through the above analysis, it can be concluded that adding surface agent to binder is a simple and effective way to improve adhesion. Furthermore, polymer modification benefits adhesive performance but does not affect the level of a binder with an added surface agent.

The boiling experiment is certainly not precise for evaluating very small distinctions between different treatments, such as polymer modification or an interface agent. Thus, the boiling test method is a relatively inaccurate measure of the moisture sensitivity of an asphalt mixture.

Sub-Microscope of Interface of Asphalt and Aggregate

The specimen preparation for the SEM test has three steps. First, aggregate particles are kept in a 100°C oven after they are cleaned with anhydrous ethanol. Second, they are placed in a 130°C hot binder for 15 seconds; then they are removed and placed into icy water. The last step is to cut the particle coated with binder into pieces and to spray gold on them for the SEM experiment.

Fig. 2 shows the interface status of the RA binder on granite and basalt. From the left, the interface is clear enough to determine the accurate position. A small quantity of holes and flaws in granite can be easily observed in the vicinity of the interface, which means that the interface adhesion is discontinuous. Under load or water conditioning, these holes and flaws must cause asphalt pavement damage in the form of cracks or moisture. However, the basalt topography is better.

Fig. 3 shows the interface status of the MA binder on granite and basalt. It is difficult to identify the interface location on both the basalt and granite surface. The MA and aggregate interface is continuous, which indicates that the binder adequately wetted through the aggregate surface.

Figs. 4 and 5 show the RPA and MPA interface topography on granite and basalt. All have continuous interfaces, adequate adhesion, and no holes or flaws, which implies perfect adhesive performance and water resistance. The SEM conclusion matches the boiling test results, but it is still difficult to do further analysis of the boiling test.

Based on the above analysis, SEM photos directly reflect the topography of the asphalt and aggregate interface but only allow qualitative judgment and analysis. The SEM method cannot provide information for the inconspicuous difference or contribute to further research.

Adhesive Work Test

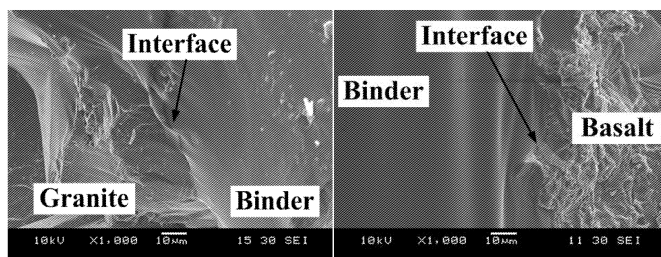


Fig. 4. Interface Topography of RPA on Granite and Basalt.

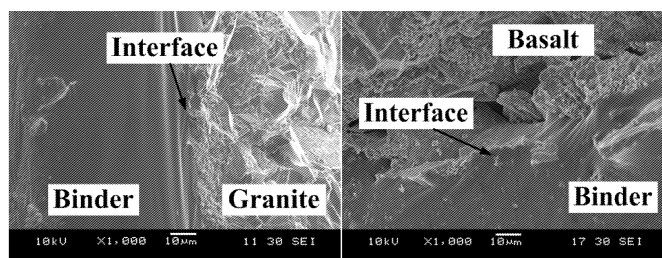


Fig. 5. Interface Topography of MPA on Granite and Basalt.

First, the contact angle is measured with an automatic digital surface tension device at 130°C. The contact angle test photos are shown in Figs. 6 to 9. Through analysis, all contact angles are less than 90 degrees, which means these binders have the ability to wet two kinds of aggregate. However, there is still a significant difference between different contact angles. Given the same binder, the contact angle on basalt is less than that on granite. For granite, the angle of MA is the lowest, the RPA angle is the highest, and the RA and MPA angles are in between. The contact angle of different types of binder on basalt follows a similar pattern to that of granite. The magnitude of the contact angle depends on the binder and aggregate properties. The lower contact angle implies better compatibility between the binder and aggregate.

The binder surface tension is measured by an automatic digital surface tension device at the test temperature, which is given in Table 5.

Adhesive work results, calculated by Eq. (6), are shown in Table 5. The RA surface tension is nearly equal to that of MA, but the adhesive work of the MA binder on both types of aggregate is higher than that of RA, which demonstrates that the interface agent affects adhesion. The compatibility of the binder and aggregate improves, and the contact angles decrease because of the polar components in the agent. Taking the coarse surface (high specific surface area) of the aggregate into account, the effective adhesive work of MA on granite increases by $2.5 \times 10^{-3} \text{ N/m}$.

For 5% SEBS agent content, RPA (modified asphalt) and MPA (modified asphalt containing interface agent) have higher surface tension than RA and MA. The adhesive work of MPA on granite increases $3.25 \times 10^{-3} \text{ N/m}$ more than that of RPA, which has the same result as the boiling test. The adhesive work of MPA on basalt increases to $67.49 \times 10^{-3} \text{ N/m}$, which is $2.38 \times 10^{-3} \text{ N/m}$ more than that of RPA. It is clear that the interface agent improves the adhesive work results, whereas the nuance cannot be distinguished with the boiling test or SEM photo.

The adhesive work of RPA is higher than that of RA, which is why modified asphalt has better moisture resistance than the base asphalt. Therefore, it will benefit adhesive performance by increasing

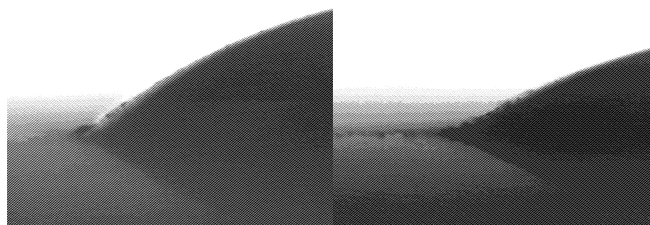


Fig. 6. Contact Angle of RA on Granite (left) and Basalt (right).

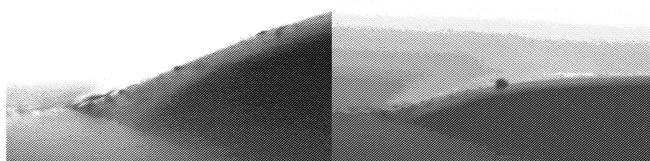


Fig. 7. Contact Angle of MA on Granite (left) and Basalt (right).

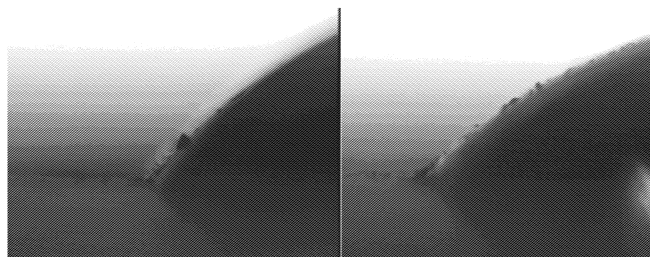


Fig. 8. Contact Angle of RPA on Granite (left) and Basalt (right).

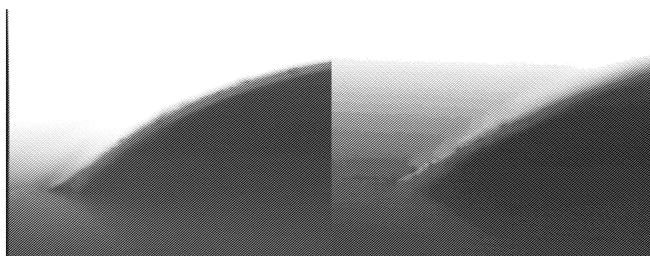


Fig. 9. Contact Angle of MPA on Granite (left) and Basalt (right).

the binder surface tension.

Conclusions

1. The adhesive work is deduced based on the surface and interface physical chemistry, which has clear theoretical meaning. It can quantitatively describe slight differences in adhesive performance that the boiling test cannot distinguish and requires an approach to adhesive phenomena from a higher level.
2. The interface topography between the binder and aggregate viewed with SEM has the advantages of intuitiveness and visibility; however, it is only a qualitative analysis method. The adhesive work experiment is a quantitative analysis with conclusive test results. If necessary, the combination of both methods is an excellent way to evaluate adhesive performance.

3. The operation of the adhesive work experiment is practical and easy. There are no jamming or test condition problems because all data are obtained with a high-precision instrument. Thus, the experimental method is of high value in both applications and theoretical research.

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