

Evaluation of Interfacial Adhesion Property between Cement Asphalt Emulsion Mastic and Aggregate

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Abstract: Characteristics of interface between cement asphalt emulsion mastic and aggregate are different from those between cement and aggregate or those between asphalt and aggregate. An interfacial splitting test was put forward to evaluate different cement asphalt emulsion mastics aggregates interfacial adhesion. Scanning Electron Microscope (SEM), Energy Dispersive X-ray Analysis (EDAX) and Electron Probe Micro-analysis (EPMA), were adopted to analyze the microstructures and the compositions. Results indicate that interfacial splitting strength tends to increase with the increase of the ratio of mineral filler to asphalt emulsion in mass (MF/AE) and reaches a maximum when MF/AE is 1.1, and then decreases. A significant improvement in the interfacial adhesion is achieved by increasing cement or mineral filler fineness and by adoption of higher strength cement, but overly fine mineral filler is disadvantageous. Limestone aggregate is easier to improve the interfacial splitting strength than granite aggregate.

Key words: *Cement asphalt emulsion mastics; Aggregate; Interfacial adhesion property, Interfacial splitting test.*

Introduction

Asphalt emulsion mixtures have logistical advantages over hot asphalt mixtures, in that they can be stockpiled or transported over long distances without special precaution. Ordinary asphalt mixtures need to be heated to construct, which can pollute the environment and consume the energy; and the mixtures can become soft in high temperature or brittle in low temperature. Energy savings can also be realized through the use of asphalt emulsion mixtures. For these reasons, suitable asphalt emulsion mixtures are in great demand but at present products are not available for use in all situations. However, the use of asphalt emulsion is largely restricted to various types of surface treatment (such as slurry surfacing and surface dressing) and bond coat [1]. Recently, efforts have been directed to the use of emulsion in mixtures used for patching with the cement addition [2-4]. Ordinary Portland cement can produce hydrated calcium silicate (C-S-H) gel after hydration, which has high strength after hardening and has a helpful effect on performance improvement of asphalt emulsion mixtures [5-7].

In asphalt mixtures, interfacial property between asphalt and aggregate has a direct effect on performance of the mixtures. However, in China, the Water Boiling Test (ASTM D3652) is the only method which is used to measure interfacial adhesion between asphalt and aggregate. The boiling condition is difficult to control and the result generated by operator's eyes is inaccurate. As to asphalt emulsion, its adhesion to aggregate is only tested by a mixing test. The adhesion result is evaluated by distribution degree of asphalt emulsion on aggregate surface, which is subjective. Therefore, some researchers put forward another method to evaluate the interfacial bond. For example, by calculating the work of

adhesion between asphalt and aggregate, Saad [8] calculated that between water and asphalt or aggregate. Xiao [9] measured asphalt aggregate contact angle to study asphalt and aggregate interface bond strength. However, the asphalt in these tests is liquid and the method is invalid to the cement asphalt emulsion mastic. In addition, binding functions of cement and asphalt emulsion in cement asphalt emulsion mixtures are different from those in cement concrete or asphalt mixtures [10, 11]. Especially, interfacial adhesion between aggregate and cement asphalt emulsion is also different from that between aggregate and cement or that between aggregate and asphalt [12-14].

This study was done in order to solve the problems and to provide an improved insight into how to quantify the interfacial adhesion ability between aggregate and bonded mastic. Different mastics and aggregates were adopted and the interfacial adhesion was evaluated with an interfacial splitting test. The change rules of the interfacial splitting strength were studied in the paper. The optimal MF/AE and fillers were obtained. Some micro apparatuses, such as SEM, EDAX, and EPMA, were adopted to study the microstructures and to analyze the compositions of the mixtures.

Simplified Interfacial Adhesion Theory

The interaction at the organic phase and inorganic phase interface has a direct effect on interfacial adhesion. An emphasis lies on the strength to separate the two phases on the ideal contacting condition to determine the interfacial adhesion ability between aggregate and cement asphalt emulsion mastic. Cherry [15, 16] put forward the strength lying in the half infinite parts of the same material, which is simplified that

$$F_i = \frac{2\pi m^2}{a^3} \left(\frac{A}{12} - \frac{B}{90a^6} \right) \quad (1)$$

If the materials are different at the interface,

$$F_i = \frac{2\pi m_1 n_2}{a^3} \left(\frac{A_{12}}{12} - \frac{B_{12}}{90a^6} \right) \quad (2)$$

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where F_t is acting force between material molecular; n_1 , n_2 is material molecular density respectively; a is material molecular distance; A , B (or A_{12} , B_{12}) are constants respectively.

When the material molecular lies in a balance distance, then $F_t=0$, so

$$\frac{A_{12}}{12} = \frac{B_{12}}{90r_{12}^6} \quad (3)$$

where r_{12} is material molecular distance; and

$$F_t = \frac{2\pi n_1 n_2 A_{12}}{12a^3} \left[1 - \left(\frac{r_{12}}{a} \right)^6 \right] \quad (4)$$

If separating the two phases, a force being bigger than $(F_t)_{\max}$ should be exerted at the interface, and

$$(F_t)_{\max} = \frac{2\pi n_1 n_2 A_{12}}{12} \cdot \frac{2}{3^{2/3} r_{12}^3} \quad (5)$$

Good [17] put forward the interfacial energy formula,

$$\gamma = \frac{\pi n^2 A}{32r_0^2} \quad (6)$$

where r_0 is balance distance between material molecular; and at the interface,

$$\gamma_{12} = \frac{\pi n_1 n_2 A_{12}}{32r_{12}^2} \quad (7)$$

where γ_{12} is interfacial tension; so

$$(F_t)_{\max} = 2.06 \frac{\gamma_{12}}{r_{12}} \quad (8)$$

The equation indicates that interfacial tension and interfacial distance have a great influence on the interfacial adhesion. The higher the interfacial tension, the stronger the interfacial adhesion. The longer the interfacial distance, the weaker the interfacial adhesion. So the key factors enhancing the adhesion strength between the mastics and aggregate are to increase the interfacial tension and to decrease the interfacial distance. Xiao's study can also confirm the importance of interfacial tension on interfacial adhesion [9]. In addition, decreasing the aggregate surface deficiency is also important because there exists stress concentration phenomenon on loading, which results in the higher interfacial stress than the main stress and the destruction of the interfacial adhesion ahead.

Table 3. Properties of Aggregates.

Properties	Limestone	Granite	Limestone Filler		
			Limestone Filler	Limestone Filler 30	Limestone Filler 60
Density /(g·cm ⁻³)	2.804	2.852	2.871	2.878	2.894
Resistance Fragmentation /%	14.3	9.8	—	—	—
Specific Surface Area /(m ² ·g ⁻¹)	0.367	0.358	0.365	0.898	0.947
Hydrophilic Coefficient	0.82	0.87	0.83	0.80	0.88
Zeta Potential /mV	-4.653	-20.060	-4.650	—	—

Table 1. Properties of Asphalt Emulsion Evaporation Residue.

Properties	Results
Content of the Residue (in mass) /%	68
Penetration (25°C, 100g, 5s) /0.1mm	84
Softening Point (t _{R&B}) /°C	46
Ductility (15°C) /cm	45

Table 2. Properties of Ordinary Portland Cement.

Physical Properties	Cement A			Cement B
	Cement A	Cement A30	Cement A60	
Density /(g·cm ⁻³)	2.977	2.979	2.983	2.981
Specific Surface Area /(m ² ·g ⁻¹)	0.396	0.734	0.755	0.406
Residue on 45µm sieve /%	20.12	12.19	10.46	17.97
28 Curing-day Compression Strength /MPa	41.8	52.1	55.7	52.6
Hydrophilic Coefficient	0.57	0.52	0.51	0.54
Zeta Potential /mV	-2.484	-1.320	5.414	3.426

Raw Materials and Experiment Setups

Raw Materials

A new type of slow setting cationic asphalt emulsion was used and its evaporation residue properties were shown in Table 1. Two types of cement, cement A and cement B, were used and their physical and chemical properties were given in Table 2. The aggregates used in this study were crushed limestone and granite. The mineral fillers were limestone filler. Their properties are given in Table 3. The limestone filler and the cement were milled for 30 minutes and 60 minutes, respectively, with a planetary ball mill. They were named limestone filler 30, limestone filler 60 and cement 30, cement 60 for a convenient description in the paper.

Interfacial Splitting Test

An interfacial splitting test was designed to evaluate interfacial adhesion. The adhesion ability was measured by the interfacial splitting strength, which is shown in Fig. 1. A steel model with 40 mm×30 mm×40 mm sizes was used. The maximum size is near to 4.75 mm used in bonding test between asphalt emulsion and aggregate in China [18]. Before being tested, the aggregate with

40mm×30mm×20mm sizes was incised and the surface was polished firstly. The aggregate was put into the steel model and the cement asphalt emulsion mastics with different proportions were mixed in a pot. The mastics were poured into the steel model, vibrated and planed. Finally, the specimens were placed at 20°C-25°C temperature for 12 hours and were cured with the models at (20±5)°C temperature and 90% relative humidity for 28 days. During the curing period, specimens were always covered with plastic film.

Two 2-mm steel bars in diameter were used at the interface upper and the below. The pressure was imposed on them until the interface cracks with 25mm·min⁻¹ speed. The load was recorded and the interfacial splitting strength was calculated with the formula

$$f_{is} = \frac{2F}{\pi A} \quad (9)$$

where f_{is} is interfacial splitting strength, MPa; F is load, N; A is interfacial area, mm².

Splitting strength of five specimens was tested and the average value was calculated. The result over the 15% average value was eliminated and the average value of the remnants was calculated, which was the splitting strength at the interface.

Results and Discussion

Effect of MF/AE

Mastics contain cement A (cement A, cement A30 or cement A60), mineral filler (limestone filler, limestone filler 30 or limestone filler 60), and asphalt emulsion. The cement used in mastics* is cement B and the other components are the same as the mastics. MF/AE is 0.9, 1.0, 1.1, 1.2 and 1.3. The results of interfacial splitting strength between different mastics and aggregates are shown in Fig. 2.

Fig. 2 represents that interfacial splitting strength tends to increase with the increase of MF/AE and reaches a maximum when MF/AE is 1.1; and then decreases. The reason is that if MF/AE is 0.9, there exists much asphalt emulsion and free asphalt, as shown in Fig. 3 a), which can wrap cement particles and impede cement hydration. With the increase of MF/AE, cement and mineral filler dosage begin to increase. Accordingly, water released from asphalt emulsion demulsification is much absorbed and there exists more cement hydrates and asphalt touching aggregate surface. Therefore, when MF/AE = 1.1, the mastic structure is dense and there is much fibrous cement hydrates, C-S-H, as shown in Fig. 3 b), which can enhance the interfacial splitting strength. When MF/AE is beyond 1.1, specific surface areas tend to increase. The mastic displays sticky and forms a wall between the mastic and the aggregate, which can deteriorate interfacial structure and decrease interfacial splitting strength. Therefore, MF/AE has a direct effect on the interfacial adhesion, which is consistent with Zhang's study result [19].

Effect of Fillers' Fineness

The results of the interfacial splitting strength with different fillers' fineness and aggregates are given in Fig. 4.

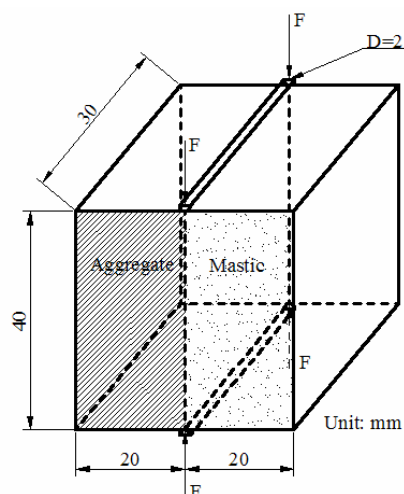


Fig. 1. Chart of the Interfacial Splitting Test.

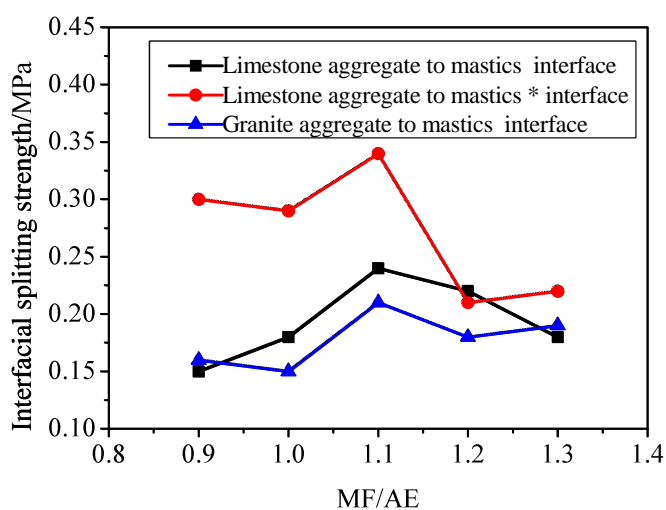


Fig. 2. Chart of Interfacial Splitting Strength with Different MF/AE and Aggregates.

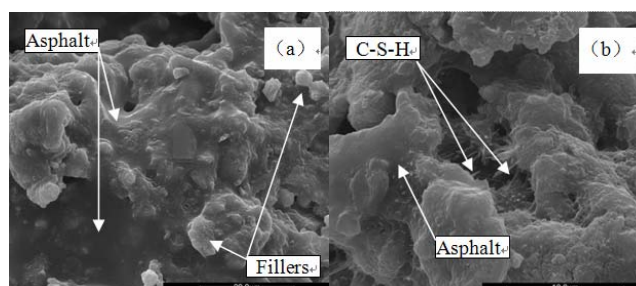


Fig. 3. SEM Pictures of Different Mastics: a) MF/AE=0.9; b) MF/AE=1.1.

Fig. 4 a) exhibits that cement fineness is helpful to enhance the interfacial splitting strength and the interfacial splitting strength between limestone aggregate and the mastic with cement A60 is the highest. The reason lies in that specific surface area and Zeta potential of cement illustrated in Table 2 increase, and hydrophilic coefficient decreases after being milled for 30 min or 60 min. It shows that finer cement can hydrate quickly. This can enhance 28 curing-day compression strength and produce more hydrates when

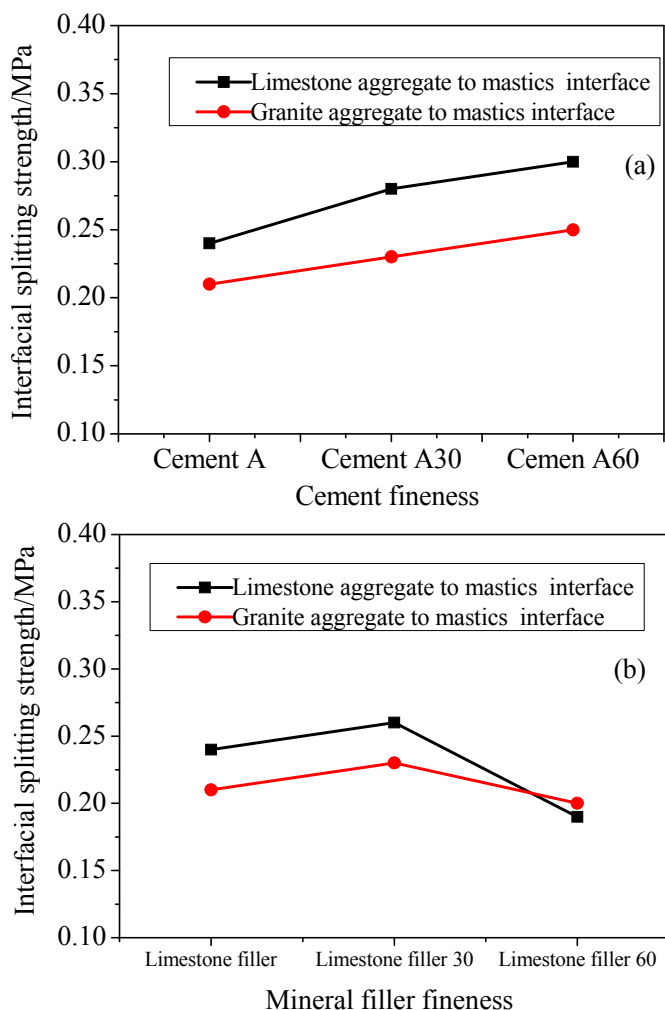


Fig. 4. Chart of Interfacial Splitting Strength with Different Fillers' Fineness and Aggregates: a) Cement; b) Mineral Filler.

the cement contacts with water. The hydrates can decrease the interfacial distance r_{12} .

Fig. 4 b) indicates that finer mineral filler can enhance the interfacial splitting strength. It is attributed that finer mineral filler can easily absorb asphalt emulsion. This makes the asphalt emulsion release much water and provide more water for the cement hydration. So the interfacial tension γ_{12} is enhanced. However, overly fine mineral filler tends to decrease the interfacial splitting strength. The reason is that the hydrophilic coefficient and specific surface area of the overly fine mineral filler increase to 0.880 and 0.947 $m^2 \cdot g^{-1}$, as shown in Table 3, which make the mastic dry and loose.

Effect of Aggregate Lithology

The results of the interfacial splitting strength with different mastics and lithologic aggregates are illustrated in Fig. 5.

Fig. 5 suggests that aggregate lithology also has a remarkable influence on the interfacial splitting strength. Limestone is easier to improve the strength than granite. In contrast to limestone, granite surface is denser and its Zeta potential is -20.060 mV, which is not helpful to combine with anionic asphalt binder. In addition, the

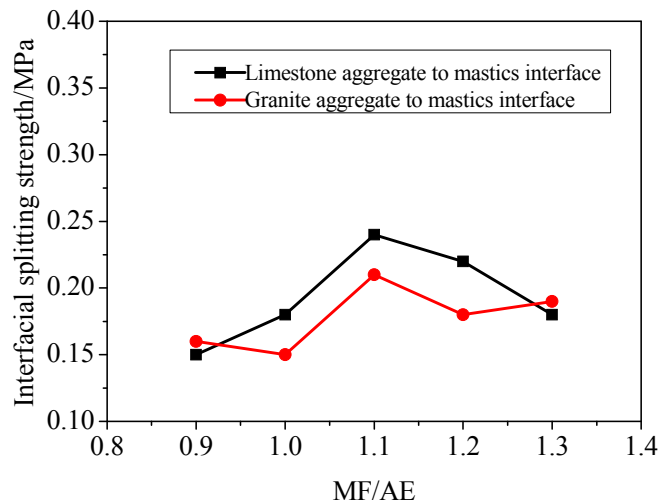


Fig. 5. Chart of Interfacial Splitting Strength with Different Mastics and Lithologic Aggregates.

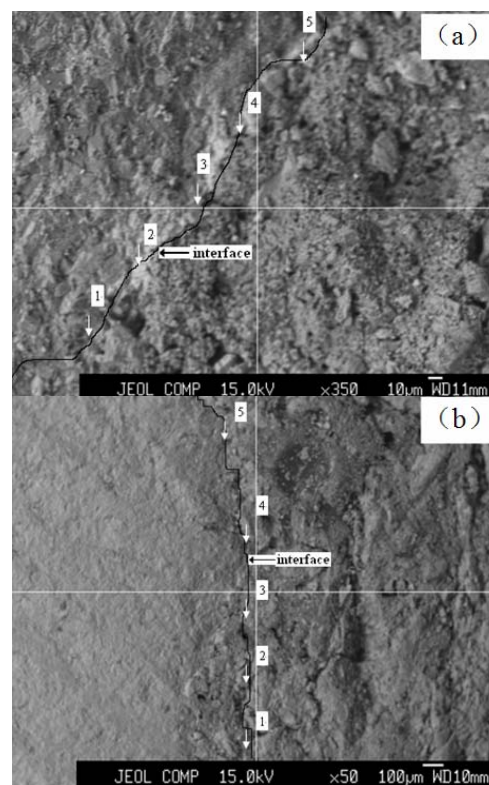


Fig. 6. EPMA Pictures of the Interface with Different Aggregates: a)Limestone; b)Granite.

cement hydrate amounts are also different. The mass percentage of Si, Ca, SiO₂, and CaO was determined by the EPMA at five testing points, as shown in Fig. 6, of the interface between aggregates and mastics. It indicates that that mass percentage at the limestone aggregate interface is 10.26%, 28.24%, 10.57% and 31.52% respectively, while that between granite aggregate interface is 25.67%, 11.20%, 33.51%, and 11.67%. It shows that mass percentage of Ca element at different points in mastics with limestone filler is higher than that in mastics with granite filler, However, mass percentage of Si element is lower. The results reveal that the presence of alkali surrounding is favorable to the existence

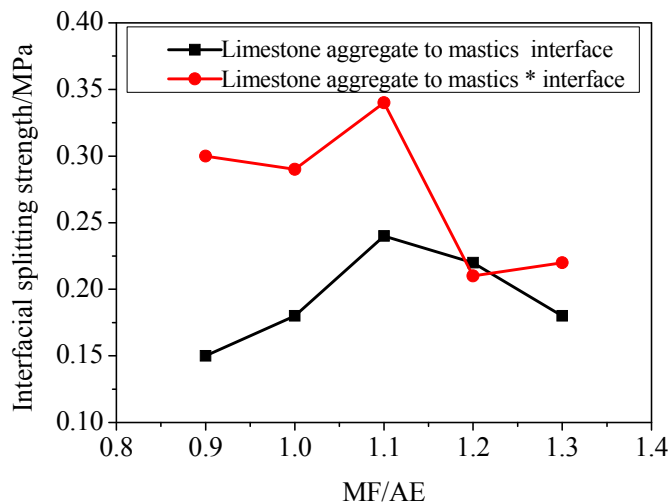


Fig. 7. Chart of Interfacial Splitting Strength with Different Cement Kinds and Limestone Aggregate.

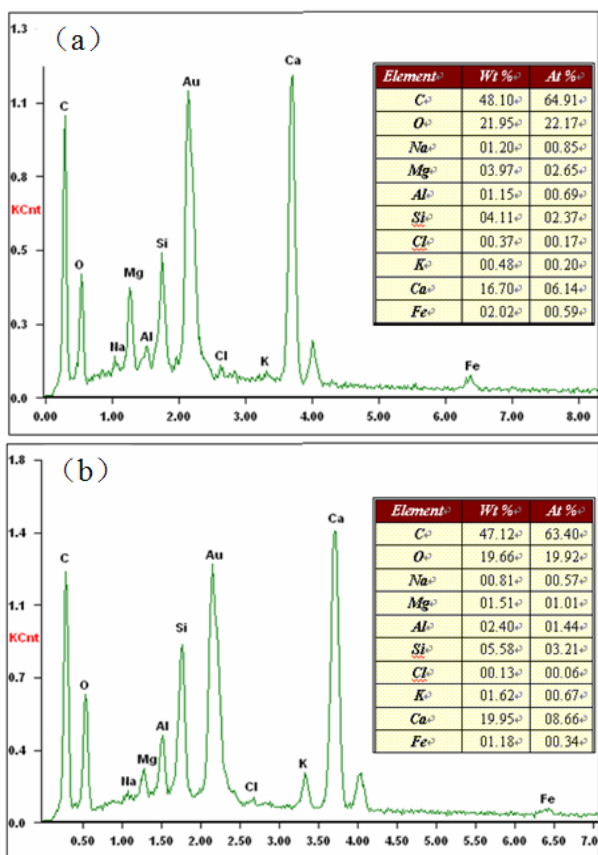


Fig. 8. Spectrums of Different Mastics: a) with Cement A; b) with Cement B.

of cement hydration productions [20], such as C-S-H, which can enhance Ca element proportion. Thus interfacial splitting strength is higher. However, it should be considered that there is much Ca element in limestone, while much Si element is in granite. Therefore, it can not show the function of cement hydrates in the strength improvement.

Effect of Cement Kinds

The results of the interfacial splitting strength with different cement and limestone aggregate are shown in Fig. 7.

Fig. 7 shows that a significant improvement in the adhesion is achieved if the bonded mastic* with cement B is applied. This is because the 28 curing-day compression strength of cement B is 52.6 MPa. It is evidently higher than the 41.8 MPa of cement A. In addition, hydrophilic coefficient of cement B is smaller than cement A's 0.57, indicating that cement B has better appetence with asphalt than cement A and is easier to cohere to the aggregate surface. It indicates that improving cement strength grade is an effective way to enhance the interfacial splitting strength. The results analyzed with EDAX in Fig. 8 a) indicate mass percentage of C, O, Si and Ca in mastic with cement A is 48.10%, 21.95%, 4.11% and 16.70%, and atom percentage is 64.91%, 22.17%, 2.37% and 6.14%, respectively. Fig. 8 b) shows that mass percentage of C, O, Si and Ca in mastic with cement B is 47.12%, 19.66%, 5.58% and 19.95%, and atom percentage is 63.40%, 19.92%, 3.21% and 8.66%, respectively. The results reveal that asphalt percentage in mastic with cement A is higher, which indicates that there is not enough cement hydrates wrapping the asphalt binder. The mass percentage of Ca and Si are higher in the mastic with cement B and Zeta potential of cement B is also higher than those of cement A. These can put forward favorable alkaline surroundings for cement hydrates existence and a highly positive zeta potential for a strong adsorption [21]. It makes the mineral fillers congregate and decreases the interfacial distance r_{12} . So this provides evidence that the measures of increasing the cement hydrate amounts, such as adoption of high strength cement, can distinctly improve the interfacial splitting strength.

Conclusions

The conclusions drawn from this study are summarized as follows.

1. A simple method is put forward to evaluate interfacial adhesion between cement asphalt emulsion mastic and aggregate.
2. Interfacial splitting strength tends to increase with the increase of MF/AE and reaches a maximum when MF/AE is 1.1, and then decreases.
3. Characteristics of cement and mineral filler have a marked influence on the interfacial splitting strength. Increase of their fineness and adoption of high strength cement are effective ways to improve the interfacial adhesion. But overly fine mineral filler can result in a poor interfacial adhesion.
4. Lithology of aggregate has a remarkable influence on the interfacial splitting strength. Limestone aggregate is advantageous to improve the strength than granite aggregate.

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