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Investigation of the bonding properties of bitumen using a novel modified binder bond strength test

Lu Zhou [a,*], Gordon Airey [a], Yuqing Zhang [b], Chonghui Wang [c], Haobai Zhong [d]

[a] Nottingham Transportation Engineering Centre, Department of Civil Engineering University of Nottingham, University Park, Nottingham NG7 2RD, UK
[b] School of Transportation, Southeast University, Nanjing 211189, China
[c] Department of Civil Engineering, Aston University, Birmingham B4 7ET, UK
[d] The Key Laboratory of Road and Traffic Engineering, Tongji University, 4800 Cao' an Road, Shanghai, China

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ABSTRACT

The binder bond strength (BBS) test can directly quantify the bonding of bitumen-stone joints. However, the index "bond strength" used in this method cannot provide a clear distinction when evaluating the performance of various bitumen, and some results do not correspond to the field feedback related to the bonding performance. This study introduces a novel modified BBS test using the universal testing machine (UTM). The results of the standard BBS test and the BBS-UTM test on six unmodified bitumens and a styrene-butadiene-styrene modified bitumen (SBSMB) are compared, with the Cantabro loss test to validate the accuracy of the BBS-UTM test. The results show that the "bond energy" can be considered the critical indicator for characterising the bonding performance of different binders. The force-displacement curve in the BBS-UTM test is analysed and provides a mechanistic explanation for the mechanical response of bitumen during pull-off. The four-component test is conducted to link the bitumen composition characteristics to its bonding performance. It is found that bitumen adhesion is correlated to the content of asphaltenes. A higher saturates content shows more contribution of tenacity in the total bond energy, and an excessively high ratio of asphaltenes to resins may result in low bond energy.

1. Introduction

As a result of traffic loading and moisture damage, pavements suffer from a variety of distresses, such as stripping and fatigue cracking, leading to shorter pavement service life, increased pavement structural safety hazards, higher maintenance costs, and possibly even pavement reconstruction [1,2]. Most pavement distress begins with the debonding between the bitumen and the aggregate or the development of cracks within the bitumen. Microcracks gradually develop into macrocracks due to various external factors. The bonding performance of the bitumen-aggregate system determines the ravelling resistance and the durability of asphalt mixtures [3,4]. Effective experimental methods are needed to evaluate the bonding property of bitumen in order to investigate the effect of various factors, which can help determine the compatibility of multiple bitumens and aggregates. Studies on the bonding property of bitumen can provide an experimental basis and technical support for selecting bitumen modification strategies and the ability to alleviate pavement performance deterioration.

Extensive efforts have been devoted to accurately measuring the bonding property of bitumen, and a variety of experimental methods have been used to evaluate the bond strength between bitumen and stones. Binder bond strength (BBS) tests are increasingly used among these studies due to their reliable data and simple procedure. Youtcheff and Aurilio [5] first conducted the BBS test to study the moisture susceptibility of binders using a pneumatic adhesion tensile testing instrument (PATTI), which was initially applied in the coating industry. Since then, the BBS test has been modified and is increasingly used in the bitumen material industry to evaluate the bonding properties of bitumen and compatibility between stones and binders. Many studies [6–15] have reported that the BBS test can offer a direct and quick bond strength measurement at the bitumen-stone interface. The BBS test is now included in the AASHTO T 361 protocol as a standard testing method for determining bitumen bond strength.

Many studies have used BBS tests to compare the bonding properties of polymer-modified and unmodified bitumen. However, different conclusions have been reached about whether modifiers (especially the

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* Corresponding author.
E-mail address: lu.zhou@nottingham.ac.uk (L. Zhou).

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styrene-butadiene-styrene (SBS) modifiers) can improve the bonding properties of bitumen. Moraes et al. [15] performed the BBS test to study the influence of various binders, modifications and minerals on the bond strength of bitumen-stone joints. The results showed that polymer modification significantly improved the bond strength. Xu [16] found that adding SBS could significantly enhance the adhesion of the binder, especially the effect of branched SBS. Huang and Lv [9,17–19] have done extensive work on the effect of different modifiers on the adhesion properties of bitumen using the BBS experiments. The results showed that SBS, polyethylene (PE), polyphosphoric acid (PPA), and gilsonite could enhance the bond strength between unmodified bitumen and stone, with bond strength increasing with the dosage of the SBS modifier. However, 3% (wt) SBS-modified bitumen (SBSMB) showed a lower bond strength than unmodified bitumen. They also investigated the bonding and healing properties of bitumen and mastic using the modified BBS test[17]. The effects of moisture and SBS modification were studied. It was found that moisture and the SBS modifier have a negative impact on the fracture-healing properties of bitumen and mastic. Zhou et al. [20,21] also studied the effects of various modifiers on the bonding performance of bitumen using the BBS test. They found that SBS modification would deteriorate the bond strength of bitumen. The inconsistent findings mentioned above suggest that the accuracy of the BBS test in evaluating the bonding properties of polymer-modified bitumen remains to be verified. More effective indicators besides a single "bond strength" are needed to characterise and evaluate the bonding properties of bitumen comprehensively.

In addition, the BBS test method has the following disadvantages: the sample tested in the experiment is a metal stub-bitumen-stone joint, which differs from the real pattern of stone-bitumen-stone of actual asphalt pavement. It is difficult to change the bitumen film thickness and loading rate in the BBS test. These shortcomings of the BBS test limit its application and adversely affect the accuracy of the evaluation analyses.

In this study, a novel modified BBS test capable of characterising the bonding properties of bitumen using the universal testing machine (UTM) is introduced, referred to as the BBS-UTM test. This experimental method can make up for the shortcomings of the above BBS test, provide the force-displacement curves during the loading process, evaluate the bitumen-stone bonding performance from the perspective of the bond energy, and adapt to a variety of experimental loading requirements due to its ability to flexibly change the loading mode and experimental parameters, In this paper, the bonding properties of six unmodified bitumens with different chemical compositions and the SBS-modified bitumen are evaluated using the new critical indicator of bond energy. The results are compared with those from the standard BBS test. Additionally, the Cantabro loss test was used to verify the accuracy of the BBS-UTM test. Finally, the bitumen four components (saturates, aromatics, resins and asphaltenes (SARA)) test was performed to correlate the bonding properties with bitumen chemical components.

2. Objectives

The objectives of the study are as follows:

(1) To introduce a modified BBS test method using UTM to accurately evaluate bitumen-stone bonding properties, which can differentiate between the bonding properties of various bitumen binders (especially for modified binders).

(2) To evaluate the bonding properties of six unmodified bitumen and one SBS-modified bitumen using the BBS-UTM experiment and the BBS experiment, respectively; to compare the results of the two experiments and to validate the accuracy of the BBS-UTM experiment using the Cantabro test.

(3) To investigate the correlation between bitumen bonding properties and its component composition.

3. Materials and methods

3.1. Materials

In this study, six unmodified bitumens (A, B, C, D, E, and F) from different crude sources with different chemical compositions were selected. Bitumen A, D, and F have the similar penetration of 60/70. Bitumen B and C have the same level of 70/80 penetration and bitumen E has a penetration of 80/90. Bitumen A was blended with the linear SBS modifier at 4.5% of the total binder weight to produce the SBS-modified bitumen. The conventional physical properties of bitumens are shown in Table 1.

It is worth noting that although some unmodified bitumens used in this study have the same Superpave performance grade (PG) and similar consistencies, they differ in terms of chemical and physical properties. Based on the penetration characteristic, the six unmodified binders can be categorised into three groups: 60/70, 70/80, and 80/90. The BBS-UTM tests on bitumen binders with different penetrations can reflect the effect of bitumen penetration on the bonding properties. Tests performed on binders with similar penetrations can show the advantage of the BBS-UTM test in differentiating bitumen bonding performance regarding its adhesion and tenacity properties. Furthermore, the tests on the SBS-modified bitumen can demonstrate the accuracy of the BBS-UTM experiment in characterising the bonding properties of bitumen, particularly the modified bitumen with significant elastic properties.

In addition, basalt, which is commonly used in pavement, was chosen as the experimental material. Basalt boulders were cut into slabs of the same size, and the surface of the stone slabs was polished with the same process using a polishing machine to achieve the same surface roughness for experimental purposes.

3.2. Asphalt mixture preparation

In this study, basalt was used as the coarse aggregate. Limestone was selected as the fine aggregate and filler for asphalt mixture preparation. A typical AC-13 dense gradation was chosen. The aggregate properties and the passing rate of each sieve in AC-13 are shown in Table 2.

The six unmodified bitumens and one SBSMB were blended with aggregates with a bitumen content of 4.7%. The loose asphalt mixture was conditioned in the oven at 163 °C for 2 hours to simulate the short-term ageing in the field. After ageing, the loose asphalt mixture was compacted in the Superpave gyratory compacter (SGC) to prepare 100 mm diameter samples. The target air void content of the samples is 5.5%.

3.3. Methods

3.3.1. BBS-UTM test

The BBS-UTM test is a modified BBS test using the UTM. During the test, the stone-bitumen-stone joint is vertically pulled off at a constant speed of 2.0 mm/min, and the loading force-displacement curve is recorded. The specific preparation procedure for the sample is as follows:

(1)The stone slabs are cleaned with ultrasonic water bathing for 10 minutes to remove any contamination from the surface and ensure that the internal pores are also cleaned.

(2)Cleaned stone slabs (Fig. 1**-a)** and bitumen are heated in the oven at 150 °C and SBSMB is heated at 170 °C for an hour to evaporate the moisture inside the voids of the stones and to allow the SBSMB to flow. After heating, a silicone rubber mould is put on the bottom slab **(**Fig. 1**-b)**, and about 2 g of liquid bitumen is dropped into the centre of the mould **(**Fig. 1**-c)**. The top slab is then placed on the bottom slab in a cross shape **(**Fig. 1**-d)**. The silicone rubber mould is customised and gouged out in the middle. A slit at the edge is made so that when the top slab is placed, excess hot bitumen can flow out through the small opening, and the thickness of the bitumen film can be accurately controlled at 0.2 mm

Table 1
Conventional physical properties of the selected bitumens.

	Binder Properties					
	Penetration (25℃)/0.1 mm	Softening Point/℃	Ductility /cm	Brookfield Viscosity at 135℃/Pa·s	Performance Grade	Crude Sources
Bitumen A	64.3	47.2	44.2 (10℃)	0.428	64–22	Iran
Bitumen B	72.3	46.6	30.9 (10℃)	0.364	64–22	Korea
Bitumen C	70.4	46.2	56.1 (10℃)	0.386	64–22	China
Bitumen D	63.5	48.2	35.9 (10℃)	0.413	58–22	China
Bitumen E	81.2	45.8	52.5 (10℃)	0.380	58–22	China
Bitumen F	64.5	42.7	44.8 (10℃)	0.469	64–22	China
SBSMB	47.9	95.3	42.4 (5℃)	2.350	70–22	—

Table 2
Gradation for AC-13.

Combined bulk specific gravity		2.857	**Flat & Elongated Particles (%)**			10.2	**Fine Aggregate Angularity (%)**			55.8
Water absorption (%)		1.213	**Sand equivalent (%)**			72	**Los Angeles abrasion (%)**			10.7
Sieve size (mm)	16	13.2	9.5	4.75	2.36	1.18	0.6	0.3	0.15	0.075
Passing rate (%)	100	96.4	75.5	47.1	34.7	24.8	17.6	12.4	8.9	0.2

(which is the thickness of the silicone rubber mould).

(3) The stone-bitumen-stone sample is cured in the dry condition at 25 °C for 24 hours before testing. The top and bottom fixtures are mounted in a cross shape on the loading table of the UTM. In this study, the samples are vertically loaded (Fig. 1**-e)** at a rate of 2.0 mm/min at 25℃ in the environmental chamber of the UTM (Fig. 1**-f)**. Four replicates are tested for each sample.

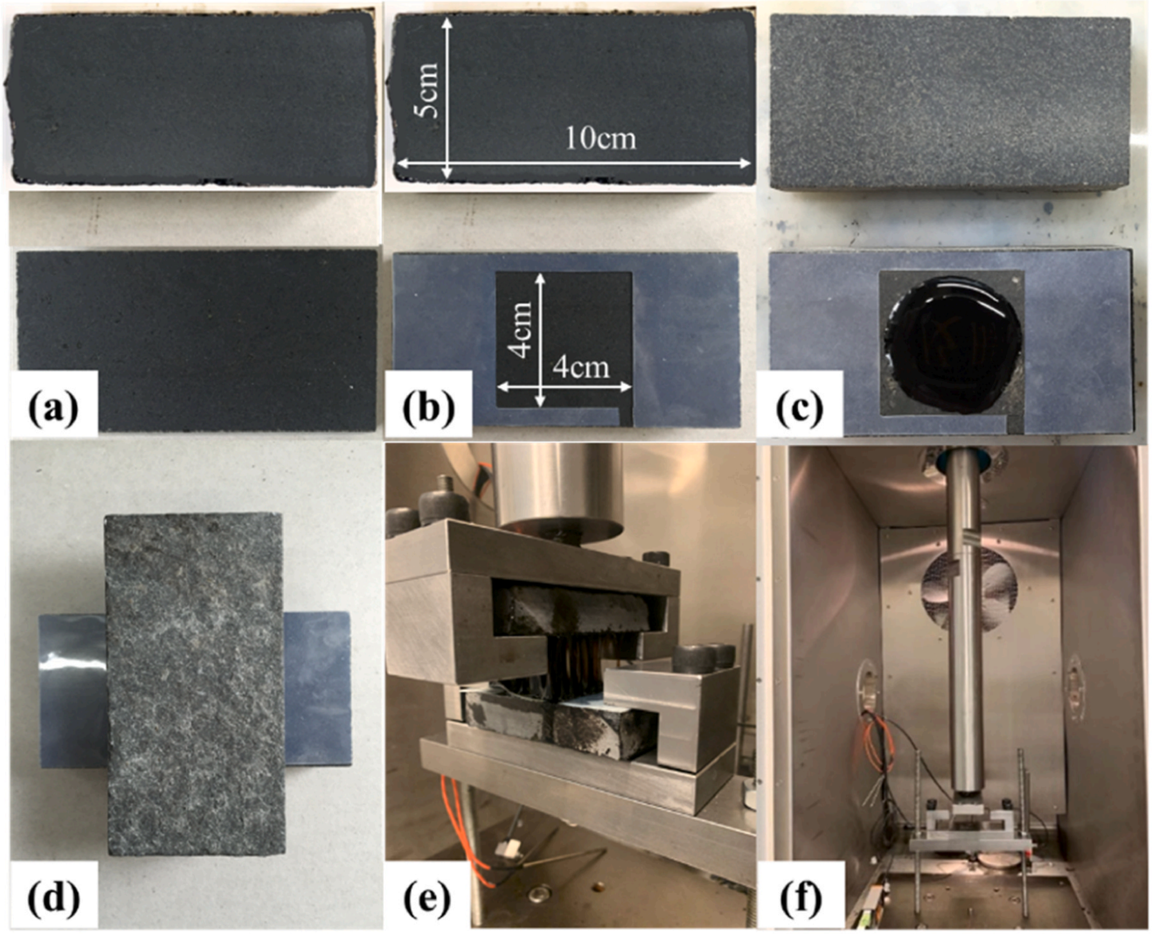

Fig. 1. (a)~(d) sample preparation in the BBS-UTM test and (e) fixtures and sample loading and (f) environmental chamber.

In the standard BBS test, "bond strength" is usually used as a critical index to evaluate the bonding property of bitumen. In the BBS-UTM test introduced in this study, "bond energy" is proposed as another evaluation indicator in addition to "bond strength". The loading force-displacement curve is recorded (Fig. 2) during the test, and bond energy is calculated from the area enclosed by the curve of force and displacement. In processing the experimental data, the weight of the upper stone slab is subtracted from the raw data collected from the UTM device to avoid its influence on the calculation of the bond strength and

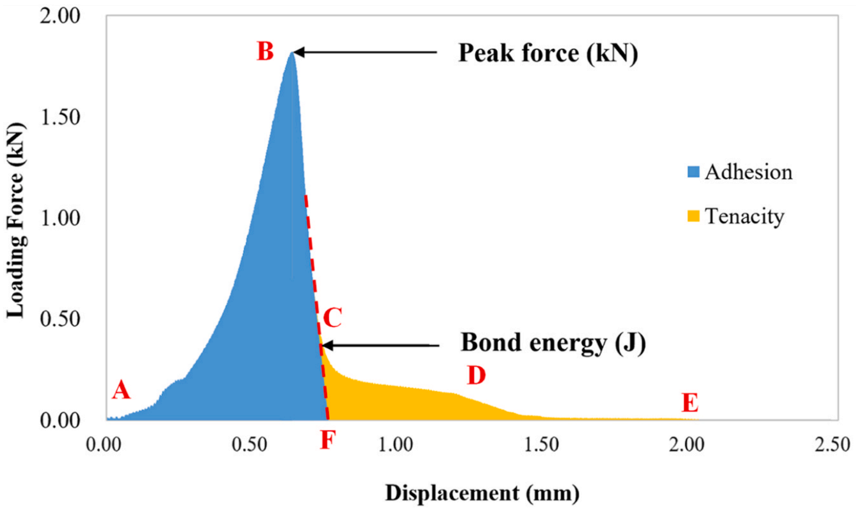

Fig. 2. The loading force-displacement curve in the BBS-UTM test.

bond energy. In this study, the reliability and accuracy of the two indices in reflecting the bonding properties of bitumen are investigated.

During the testing process, the tensile force reaches the peak in a short time (AB phase). With the accumulation of deformation, the middle part of the bitumen film gradually becomes thinner during the stretching process, and the tensile force decreases rapidly (BC phase). The BC phase of the curve approximates a straight line, which intersects the X-axis at point F. The CDE phase is the later part of the tensile deformation stage, where the bitumen film of the sample is subjected to a large deformation and eventually is damaged.

Similar to the typical tensile strength load-deflection curve from the Toughness and Tenacity Test [22–24] , the loading force-displacement curve in the BBS-UTM test shows the bonding and tenacity of bitumen. The bonding property is defined as the total area under the loading-displacement curve (S_{ABCDEF}), and tenacity is defined as the area at the tail of the curve (S_{CDEF}). The difference between these two areas (S_{ABCF}) is related to the adhesive property of the bitumen with the stone slab.

3.3.2. Standard BBS test

The standard BBS test is conducted in this study using the Positest AT-A apparatus (Fig. 3**-a)**. The bitumen film thickness is controlled at 0.2 mm by the aluminium stubs with a notched base (bottom diameter 20 mm) **(**Fig. 3**-b)**. The sample preparation procedure for the BBS test is as follows[25,26]:

(1) Similar to the BBS-UTM test, the stone slabs are cleaned with ultrasonic water bathing for 10 minutes.
(2) Cleaned stone slabs and bitumen are heated in the oven at 150 °C, and SBSMB is heated at 170 °C for an hour. After heating, silicone rubber moulds are put on the stone slab, and about 1 g of liquid bitumen is dropped into the centre of the mould **(**Fig. 3**-c)**. The heated stubs are then placed in the centre of the silicone mould **(**Fig. 3**-d),** and a weight on top is applied to allow excess bitumen to flow out of the notch in the stub **(**Fig. 3**-e)**.
(3) The sample is cured in the dry condition at 25 °C for 24 hours. The silicone moulds are removed before testing **(**Fig. 3**-f).**

In the standard BBS test, the pull-off tensile strength (POTS) is recorded as the indicator of the bonding performance. The loading rate is set at 0.7 MPa/s according to the recommendation from the protocol. Six replicates are tested for each sample.

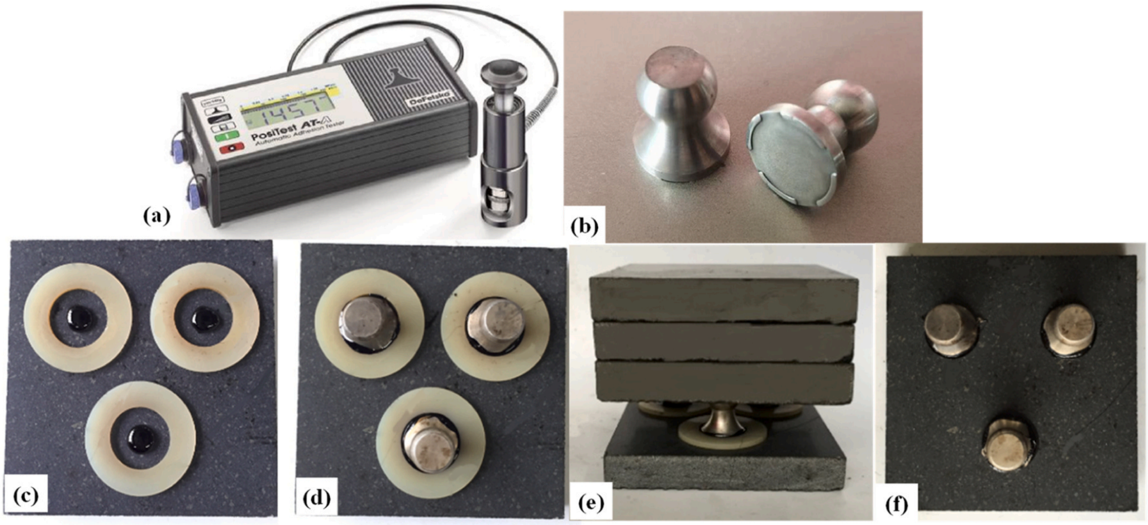

Fig. 3. (a) The Positest AT-A apparatus and (b) stubs, and (c)~(f) sample preparation in the standard BBS test.

3.3.3. *Cantabro loss test*

The Cantabro loss test is performed on the asphalt mixtures following the standard method AASHTO TP 108–14. The asphalt mixture samples were conditioned at 25℃ for 20 h before the test and then subjected to 300 revolutions in the Los Angeles abrasion testing machine at a rotating speed of 30 rpm without steel charge. The mass loss indicator for each sample is obtained by calculating the ratio of the mass lost during the test to the initial mass, which can be used to determine the ravelling resistance of the asphalt mixtures. Four replicates are tested for each type of asphalt mixture.

3.3.4. *Bitumen four fractions (SARA) test*

In this study, four components are separated and extracted from the bitumen samples according to the standard ASTM D4124–09, including saturates, aromatics, resins and asphaltenes. The principle of the separation method is based on the solubility of the four components in different solvents and the difference in the adsorption capacity of alumina on the four components [27,28]. In the SARA test, the contents of the four components are measured to correlate the bitumen bonding properties with its component characteristics.

4. Results and discussion

4.1. *Accuracy verification of the BBS-UTM test*

4.1.1. *Results comparison between the BBS-UTM test and the standard BBS test*

The BBS-UTM test and the standard BBS test are performed on six unmodified bitumens and an SBSMB to show the consistency of the two tests. The results of the comparison among the tested bitumens according to the different evaluation indicators are shown in Fig. 4.

As shown in Fig. 4, inconsistent bonding property ranking results are obtained for the six unmodified bitumens and SBSMB, depending on the indicators used from the BBS-UTM test and the standard BBS test. When bond strength measured by the standard BBS test is used as the critical indicator, the ranking result is $A > F > B > C > SBSMB > D > E$. When bond strength gained by the BBS-UTM test is used as the critical indicator, the ranking is different: $A > D > C > B > SBSMB > E > F$. However, the ranking shows $SBSMB > A > B > C > D \approx E > F$ when using "bond energy" in the BBS-UTM test as the critical indicator.

According to the index "bond strength" from the BBS-UTM test and the standard BBS test, the bonding property of the 4.5% SBSMB is similar to or even inferior to the unmodified bitumens, which is contrary to practical engineering feedback that "the antistrip performance of 4.5% SBSMB is far better than that of unmodified bitumen"[29–31]. In addition, the ranking results of some unmodified bitumens (B, C, D, E, and F) from the BBS-UTM and standard BBS tests are inconsistent even when the same evaluation index "bond strength" is used. Although this may be due to the difference in loading rates between the two experiments, it still shows that the index "bond strength" cannot distinguish the bonding properties of unmodified bitumens from different crude sources. Thus, it could be concluded that the index "bond strength" cannot accurately characterise the bonding properties of various bitumens (especially polymer-modified bitumen) and should not be used as a determinant index for bonding properties evaluation.

Based on the result from the BBS-UTM test, there is a significant difference in the rankings of bonding properties evaluated using "bond strength" and "bond energy indicators". In particular, when using "bond strength" as a critical indicator, the bonding property of the SBSMB is similar to or even worse than the unmodified bitumens. However, when "bond energy" is used as the critical index for the bonding properties evaluation, SBSMB shows the best bonding performance among all the tested bitumens (approximately three times the bond energy of the unmodified bitumens), which corresponds to the conclusions obtained from engineering practice.

4.1.2. *Result of the Cantabro loss test and the correlation analysis*

To further validate the accuracy of the BBS-UTM test in evaluating the bonding properties of bitumen, the Cantabro loss test is conducted to assess the ravelling resistance of the unmodified asphalt mixtures and SBS-modified asphalt mixture. The results of the Cantabro test and BBS tests are shown in Table 3.

Table 3 shows significant differences in the Cantabro loss of various asphalts. Among them, the ravelling resistance performance of SBS-modified asphalt is far better than that of unmodified asphalts. In

Table 3
Comparison of indicators of the BBS test and results of the Cantabro test.

Bitumen	Bond Strength by BBS-UTM test (MPa)	Bond Strength by standard BBS test (MPa)	Bond Energy by BBS-UTM test (J)	Cantabro loss
A	1.01	3.73	0.680	61.48%
B	0.84	2.67	0.503	73.90%
C	0.88	2.54	0.412	68.75%
D	0.95	2.14	0.346	84.58%
E	0.59	2.01	0.346	89.40%
F	0.51	3.07	0.333	86.33%
SBSMB	0.73	2.51	1.955	21.30%

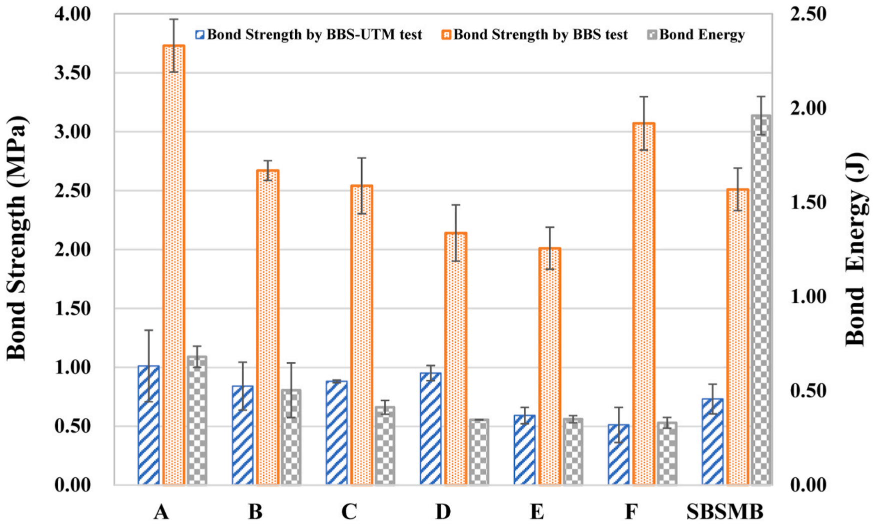

Fig. 4. Comparison of different indicators by the BBS-UTM test and the standard BBS test.

comparing multiple unmodified asphalt mixtures, asphalt A shows the lowest mass loss, while asphalt D, E, and F show poor ravelling resistance.

The correlations between Cantabro loss and indicators from the standard BBS test and the BBS-UTM test are compared in Fig. 5.

As presented in Fig. 5, both bond strength indicators in the standard BBS test and the BBS-UTM test show a poor correlation with the Cantabro loss, mainly due to the inconsistency of SBSMB in bond strength and asphalt ravelling resistance. However, the "bond energy" index in the BBS-UTM test shows a strong linear negative correlation with the Cantabro loss. The comparison between the two correlations indicates that the energy-based indicator in the BBS-UTM test is a preferable critical index in accurately evaluating the bonding properties of bitumen and can reflect the ravelling resistance of the corresponding asphalt mixtures.

4.2. Mechanical curve analysis of the BBS-UTM test on unmodified bitumens and SBSMB

In this section, different stages of the loading force-displacement curve in the BBS-UTM test are analysed in detail. Fig. 6 illustrates the difference in curves between unmodified bitumen A and 4.5% SBSMB.

According to Fig. 6, the loading forces of both unmodified bitumen and SBSMB peak when the loading displacement reaches about 1.0 mm, with the loading displacement of SBSMB, corresponding to the peak force, being slightly larger. A significant difference between unmodified bitumen and SBSMB in peak force and final loading displacement can be observed. The unmodified bitumen has a higher peak force (1.8 kN). However, the final damage occurs when the bitumen film layer is stretched to only approximately 1.6 mm. There is no rebound phase for unmodified bitumen, and the tension force drops dramatically to zero as the sample is pulled off. By contrast, the curve of SBSMB presents a lower peak force (1.3 kN) but an extended loading displacement (6.0 mm). After the peak point, there is a rapid drop in tension force, but not to zero, and then a slow rebound in the curve with a significant deformation (stretching phase) before the curve gradually drops to zero, showing excellent tenacity/ductility of SBSMB before pull-off failure.

In the AB phase, the bonding deformation stage, the loading force increases proportionally with displacement. This stage contains the elastic and plastic deformation process of bitumen. When the tensile force is applied to the sample by stretching, the bitumen first exhibits elasticity and then plastic deformation. It is shown that the tensile force increases rapidly throughout the bonding deformation stage, and the deformation change is small, leading to a high deformation modulus. At the microscopic level, the deformation in this stage can be explained by the elongation of the bond angles and bond lengths of the bitumen and polymer modifiers under external force. The peak force in the bonding deformation stage is related to the bulk properties of bitumen and the composition of the modifiers.

The tensile yield occurs in the BC phase, where the cross-section of the stretched bitumen film gradually becomes smaller as the deformation increases. At this time, the movement of the upper fixture is against the internal friction between bitumen and modifiers. The tensile force reduction appears in this phase as the frictional resistance is less than the force required to cause the elongation of the intramolecular bonds. The degree of reduction in tensile force is related to the nature of the modifiers.

After point C, the bitumen film is stretched to a point where the cross-sectional area does not change significantly, and the tensile force remains relatively constant. In the CD phase, for the SBSMB sample, some polymer chain segments of the SBS modifier have been stretched. The polymer chains and the tensile force field are in the same direction due to the external force applied to the SBS modifier through the interfacial layer. As a result, the sliding friction between the modifier molecules and the bitumen molecules becomes less, and, as the pure bitumen has already yielded, the upper fixture will be mainly resisted by the elongation of the modifier chain segments during the deformation. When the bitumen film is being stretched in this stage, the original crosslinks between polymers and bitumen or between polymers and polymers might be destroyed, while some new crosslinks may also be created, which increases the resistance to the movement of the chain segments to a greater extent, leading to the increase in tensile force in the CD phase. By contrast, this tension rebound phase does not occur in the loading force-displacement curve for unmodified bitumen. This large tensile deformation stage is essential for modified bitumen and is a crucial indication of the modification effect of the modifiers. There are many factors that affect the variation of this deformation phase, including the types and grades of unmodified bitumen, the compositions and dosages of modifiers, and the compatibility between bitumen and modifiers.

The sample is pulled off to failure at the end of the tensile deformation phase (CE or DE phase). Fig. 7 shows that the unmodified bitumen sample typically shows a fine-meshed failure section with bitumen adhering to both top and bottom slabs (cohesive failure). In contrast, the SBSMB sample shows a failure interface with only one slab having bitumen on it and another being clean (adhesive failure), which

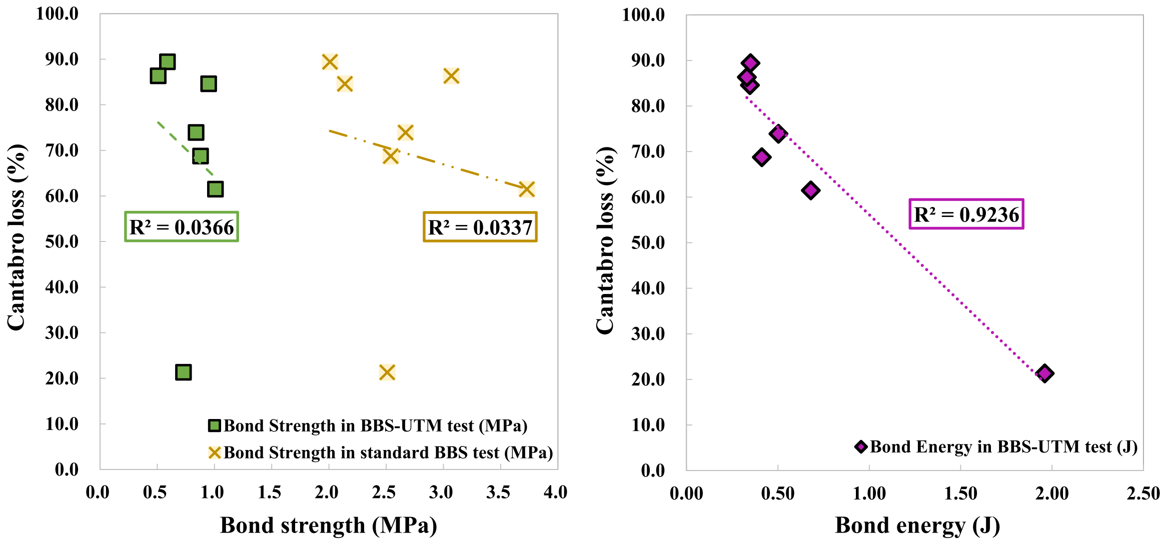

Fig. 5. The correlations between the bond strength and Cantabro loss (left) and the bond energy and Cantabro loss (right).

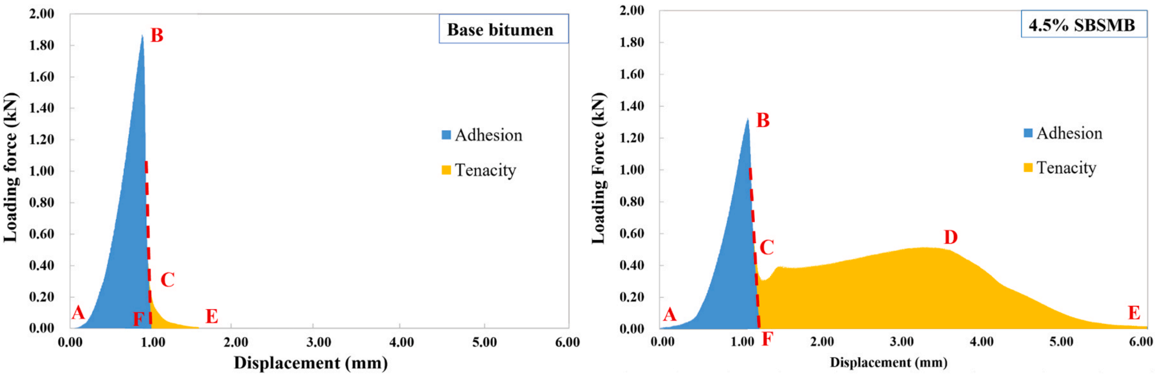

Fig. 6. The loading force-displacement curves of unmodified bitumen and 4.5% linear SBSMB.

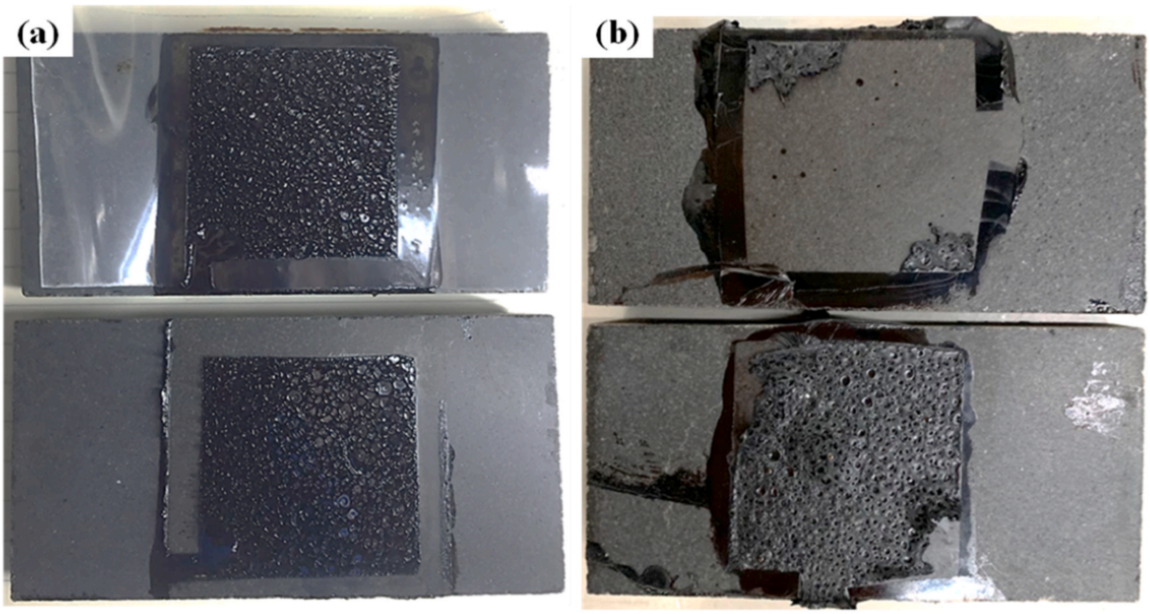

Fig. 7. The failure interfaces of the (a) unmodified bitumen and the (b) SBSMB samples in the BBS-UTM test.

indicates that the unmodified bitumen might have better penetration into the mineral surface than the SBSMB. This may also be due to the SBS modifier absorbing the small molecular structures in the bitumen, creating a net structure that reduces the infiltration and adhesion of the modified bitumen to the mineral surface.

As compared in Fig. 6, the results show that the adhesion-related area (S_{ABCF}) of the sample with unmodified bitumen is more significant than that of the sample with SBSMB. However, the SBSMB offers more significant advantages in terms of tenacity. The curve of the SBSMB sample shows that tenacity predominates, and the adhesion with stone contributes a smaller proportion of the total bond energy. Considering the practical feedback that the durability of the SBS-modified asphalt pavement is better than that of the unmodified bitumen, it can be concluded that the tenacity of the SBSMB can "cushion" the impact of external forces on the bitumen-stone structure. Under the same stress, the SBSMB can stretch and has a better ability to deform extensively and release energy from external impacts, whereas the unmodified bitumen is less elastic and has a lower ability to deform before damage. Therefore, cracks will likely occur within the unmodified bitumen compared to the SBSMB. In terms of modification effect, the SBS modifier significantly improves the tenacity and overall bonding performance of the bitumen. However, the interfacial adhesion between bitumen and stone was not enhanced. The adhesion may even be reduced due to large polymer molecules in the SBS modifier.

The above results show that the evaluation of the bonding properties of bitumen should not be determined by a single index, "bond strength". Therefore, "bond energy" should be a preferred critical indicator when evaluating the bonding properties of bitumen, especially for polymer-modified bitumens.

4.3. *Bonding properties evaluation on bitumens using the BBS-UTM test*

In this section, the BBS-UTM test is performed to evaluate the bonding properties of various unmodified bitumens and the 4.5% linear SBSMB using the "bond energy" indicator. Fig. 8 shows the adhesive energy, tenacity and the proportion of tenacity in the total bond energy.

Fig. 8 shows significant differences in the bonding properties between unmodified bitumens from different crude oil sources and between unmodified bitumens and SBSMB. Bitumen A shows the highest adhesion energy among all the unmodified bitumen and the SBSMB. SBSMB does not show an advantage in terms of bond energy as it is ranked between bitumen B and C. Regarding tenacity, there is a distinct difference among various unmodified bitumens with the same penetration of 60/80 (bitumens A, B, C, D and F). It is worth mentioning that among all the unmodified bitumens, bitumen B has the lowest tenacity/ bond energy ratio with relatively high penetration, while bitumen E, which has the highest penetration, shows the highest tenacity/ bond energy ratio. This phenomenon indicates that the proportion of tenacity

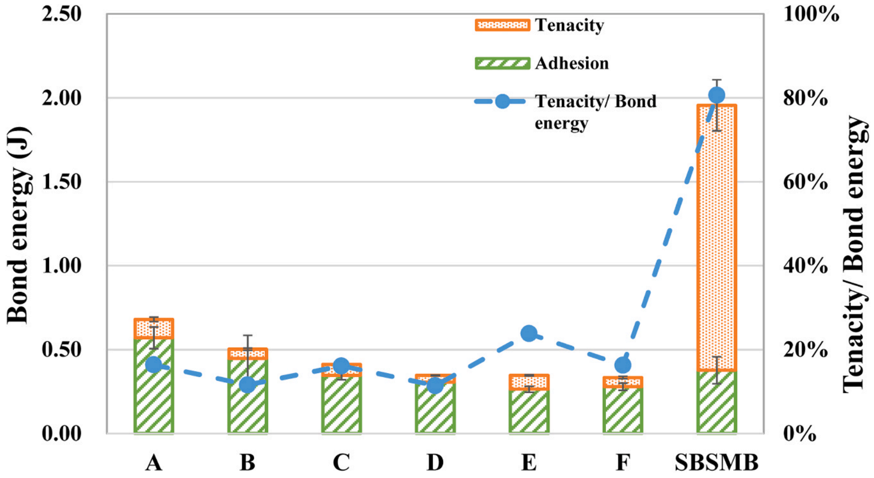

Fig. 8. The bond energy of different bitumens.

of unmodified bitumen in the total bond energy does not depend on penetration grade.

4.4. Four components test and correlation analysis

The bitumen four components test is performed to investigate the correlation between the adhesion and tenacity of bitumen and its constituent properties. The energy-based bitumen adhesion indices of the six unmodified bitumens, including Adhesion (J), Tenacity (J), Bond Energy (J), and the ratio of Tenacity to Bond Energy, and their four-component contents and ratio of asphaltenes to resins contents are listed in Table 4. It is worth mentioning that the SBS modifier would significantly alter the composition of the bitumen and interfere with the determination of the four components. The SBS-modified bitumen cannot accurately reflect the correlation between bitumen bonding properties and its components. Therefore, the SBSMB was not included in the SARA test.

The experimental indicators of the SARA and BBS-UTM tests are correlated according to the data in Table 4. In the correlation comparison of various combinations, it is found that the “Adhesion energy” index in the BBS-UTM test shows a linear correlation with the “Content of asphaltenes” in the SARA test. The “Tenacity/ Bond Energy” index is linearly correlated with saturates content, and the “Bond energy” index shows a significant linear correlation with the ratio of asphaltenes to resins contents. The correlations between the above indicators are shown in Fig. 9.

As shown in Fig. 9**,** the adhesion energy measured in the BBS-UTM test exhibits a strong linear correlation with the content of asphaltenes (R^2=0.8170). Asphaltenes in bitumen are substances with high polarity and surface activity. Chemically active components such as asphaltene anhydride and other polar components are mainly concentrated in the asphaltenes. These polar components are more likely to chemisorb with the mineral surface, and the active components improve the infiltration of bitumen to minerals, thus increasing the adhesion strength of bitumen-stone joints. In addition, from the surface energy point of view, higher bitumen polarity increases the bitumen-stone bond energy [32], which explains the linear positive correlation between the adhesion energy and asphaltenes content.

The tenacity/bond energy presents linear correlations with both the content of saturates (R^2=0.7605) and the colloidal instability index (R^2=0.9062), indicating that unmodified bitumen with a higher content of saturates will exhibit a higher percentage of tenacity in bonding properties.

According to the colloidal model of bitumen, asphaltenes are dispersed in the continuous phase (saturates and aromatics) and encapsulated by resins. The saturates fraction is the soft component of the bitumen and acts as a plasticiser. As an oil fraction, saturates play a lubricating and softening role in bitumen and, at the appropriate content (5%~20%), can increase the ductility of bitumen. Therefore, the higher the saturates content, the higher the proportion of tenacity in bond energy.

The ratio of asphaltenes to resins contents plays a vital role in the stability of bitumen since, to some extent, the proportions of asphaltenes and resins in the bitumen determine the type of bitumen colloidal structure. When the ratio of asphaltenes to resins content is relatively low, asphaltenes can be fully separated in bitumen (sol-type bitumen) [33], showing Newtonian fluid characteristics and less elastic properties. When the ratio of asphaltenes to resins content is relatively high, asphaltenes micelles aggregation are likely to occur, forming a gel-type bitumen with non-Newtonian fluid characteristics, presenting imbalanced proportions of the four components of the bitumen. When resins and asphaltenes are at proper contents, a sol-gel type bitumen is formed, showing better bonding, temperature stability and elasticity. The asphaltenes content in bitumen is typically 5~25%, and the resins content is 15~30%. However, as shown in Table 4, except for bitumen A and B, the ratios of asphaltenes and resins contents of the other unmodified bitumen binders are out of the conventional range, which indicates that bitumen A and B are of sol-gel type, while the others might

Table 4
The results of the SARA test and the BBS-UTM test.

Bitumen	SARA Contents				$\frac{m_{\text{Asphaltenes}}}{m_{\text{Resins}}}$	Energy-based Indices in the BBS-UTM Test			
	Asphaltenes	Saturates	Aromatics	Resins		Adhesion (J)	Tenacity (J)	Bond Energy (J)	Tenacity/ Bond Energy
A	17.42%	16.30%	48.50%	17.78%	1.02	0.570	0.110	0.680	16.17%
B	12.88%	8.21%	52.65%	26.26%	2.04	0.447	0.055	0.503	11.01%
C	13.65%	22.46%	23.31%	40.58%	2.97	0.346	0.066	0.412	16.02%
D	10.78%	21.29%	20.91%	47.02%	4.36	0.307	0.040	0.346	11.42%
E	6.52%	39.28%	23.58%	30.62%	4.70	0.264	0.082	0.346	23.76%
F	7.59%	23.23%	31.94%	37.25%	4.91	0.279	0.054	0.333	16.32%

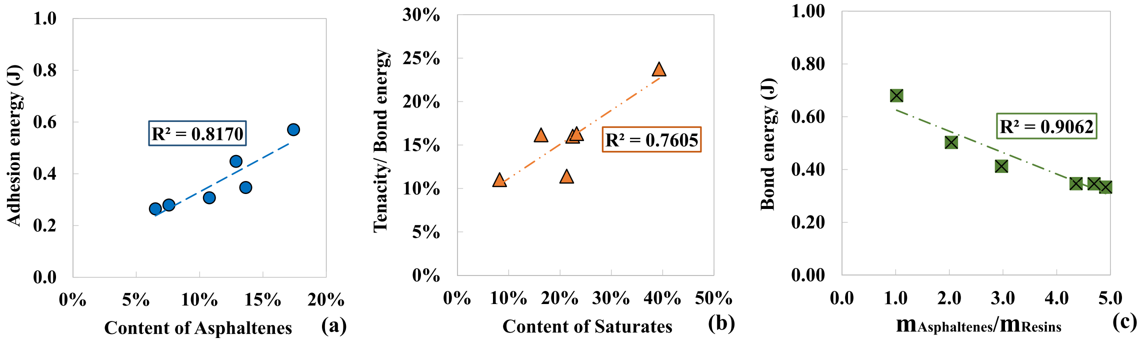

Fig. 9. The correlations between (a) adhesion energy and asphaltenes content, (b) tenacity/bond energy and saturates content, and (c) bond energy and the ratio of asphaltenes to resins contents.

be gel-type bitumen with an imbalance of components. As the contents of the four components become unbalanced, the binders show poorer bonding properties.

5. Conclusions

In this paper, a novel modified BBS test conducted on the UTM is introduced, and "bond energy" derived from the loading force-displacement curve is considered the critical indicator in evaluating the bonding properties of bitumen. The results of the BBS-UTM test are compared with those of the standard BBS test and the Cantabro loss test, and the evaluation accuracy of this test is verified. By dividing the curve in the BBS-UTM test into two parts, a clear distinction can be made between the adhesion and tenacity of the bitumen, allowing for a more comprehensive and accurate evaluation of the binder bonding properties, especially for the modified bitumen. The four-component SARA test is also performed to analyse the correlation between the bonding performance and the bitumen components. According to the results, the following conclusions can be drawn:

1. The comparison between the standard BBS test and the BBS-UTM test shows that the "bond strength" indicator in these two tests cannot provide a reliable and accurate evaluation ranking of different unmodified bitumens and SBSMB. The result of the "bond strength" indicator did not follow practical field engineering experience. "Bond energy", another indicator proposed in the BBS-UTM test, was found to accurately characterise the bonding properties of various bitumens, especially modified bitumen, and is a preferable critical evaluation index for the bonding properties of different binders. The results of the Cantabro loss test also validate the accuracy of the BBS-UTM test.
2. The adhesion and tenacity of the bitumen can be distinguished clearly by dividing the force-displacement curve into two parts. For the unmodified bitumens, adhesion accounts for a large proportion of the bonding, and binders present little tenacity. In the SBSMB, the adhesion is poorer than that of the unmodified bitumen. However, it shows a significant tenacity, allowing the modified bitumen to be deformed when subjected to tensile forces and cushion the impact of external forces. Thus, SBSMB provides asphalt mixtures with excellent ravelling resistance.
3. In the BBS-UTM test, significant differences are observed in the bonding properties between unmodified bitumen and SBSMB and even among the unmodified bitumen binders with the same penetration. Bitumen A, produced in Iran, shows the highest bond energy among all the unmodified bitumens, while the bonding performance of the SBSMB far exceeds that of the unmodified bitumen.
4. There is a strong correlation between the bitumen bonding properties and its component characteristics. Higher contents of asphaltenes in bitumen will result in better adhesion. A higher content of the saturates fraction reflects a more significant contribution of tenacity in the total bond energy. An excessively high ratio of asphaltenes to resins contents will result in an imbalance of the components in the bitumen, showing a reduction in its bonding properties.

This study introduced a novel method for testing and analysing the bonding properties of bitumen. Although the accuracy of the BBS-UTM test was verified using the Cantabro experiment, tests on more types of bitumen and aggregates are needed to further validate the conclusions of this study in future research.

CRediT authorship contribution statement

Lu Zhou: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Yuqing Zhang:** Writing – review & editing, Supervision, Resources, Project administration, Conceptualization. **Gordon Airey:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **Haobai Zhong:** Writing – review & editing, Methodology. **Chonghui Wang:** Writing – review & editing, Investigation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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