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EFFECT OF THE MORPHOLOGY OF SBS MODIFIED ASPHALT ON MECHANICAL PROPERTIES OF BINDER AND MIXTURE

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Abstract: SBS-modified asphalt is finding widespread application as a binder for high performance mixtures, and porous mixtures , etc. The performances of mixtures are greatly affected by the morphology of SBS. In this study, we made trial SBS-modified asphalt products of varying morphologies and clarified the relation of the dispersed conditions of SBS-modified asphalt to its performance and storage stability as the binder and mechanical properties of asphalt mixtures. As a result, we could show the morphology of SBS-modified asphalt required to ensure its proper performance.

Key Words: polymer modified asphalt, morphology, storage stability, flexural test of binder, porous asphalt mixture,

1. INTRODUCTION

Polymer-modified asphalt is finding widespread application as a binder for high performance mixtures and porous asphalt mixtures, etc. Chiefly used as a modifying agent for polymer-modified asphalt is Styrene Butadiene Styrene block copolymer (hereinafter referred to as "SBS"). In Japan, 10-odd years have passed since SBS was put into practical application.

By observing a picture of SBS-modified asphalt magnified about several hundred times by a microscope, its structure can be broadly divided into two types. One is the type in which asphalt forms a continuous phase and the other is the type in which SBS forms a continuous phase. Further, the dispersed particles of asphalt and SBS vary greatly in size from less than 0.001mm to 0.05mm. In this study, the type of continuous phase and morphology, as such, are defined as the microstructure of SBS-modified asphalt.

The performance of SBS-modified asphalt basically improves with an increase in SBS concentration, but is not determined by this alone, i.e., it is greatly affected by the morphology of SBS (Ueno). Though there are several reports about the morphology of SBS-modified asphalt (Suzuki), (PIARC), (Nakajima), (Murayama), there virtually no research has been made on its relation with binder properties and mixture performance.

Using SBS-modified asphalt without controlling its morphology involves the following

problems:
(1) Material segregation occurs in the storage tank of an asphalt plant, resulting in obstructing binder supply to the measuring tank.
(2) Asphalt mixtures vary in performance. In some cases, mixtures not having the aimed performance may be paved.

Test on penetration, softening point, ductility, toughness-tenacity, etc. have been used for evaluation of the binder properties of SBS-modified asphalt. However, it was pointed out that these tests are inapplicable to such types of asphalt as high-viscosity modified asphalt (hereinafter referred to as "high-viscosity type") in which the microstructure of SBS-modified asphalt forms a continuous phase of SBS. As a result, a new test method "binder bending test" has been proposed (Tanaka), (Motomatsu,2001), (Ohki), (Motomatsu,2004). The bending workload or the like obtained from this test is supposed to be affected by the morphology as well, so a high correlation with mixture properties could not be obtained unless a definite morphology is obtained for evaluation.

This study integrates some results obtained in the research stage for practical application of SBS. It was conducted to clarify the relation of the microstructure (type of continuous phase and morphology) of SBS-modified asphalt to binder properties, binder storage stability, various performances of the mixtures and the recently proposed new test of binder properties, with the aim of proving that the performance of SBS-modified asphalt is closely related with its microstructure.

2. METHOD OF STUDY

2.1 Trial Production of SBS Modified Asphalt

SBS-modified asphalt used in this study was prepared from commercially available straight asphalt 60/80 for pavement (hereinafter referred to as "StAs"). Changing the SBS concentration of StAs to 0, 3, 5, 7, 9 and 12%, we made trial products different in morphology, three per SBS concentration. Note that the SBS concentration is an external ratio in % by weight to 100 parts by weight of StAs. The properties of StAs and SBS used here are shown in Tables 1 and 2.

SBS-modified asphalt was produced by operating a high shear type homomixer that crushes and dissolves SBS until the morphology set up under the temperature condition of 190 ± 10 ℃ is attained. Specifically, SBS-modified asphalt was sampled in the binder production process, variations in morphology with time were observed under a microscope (400x) and production was finished at the stage where the three morphologies shown in Table 3 were attained.

2.2 Microstructure and Morphology of SBS Modified Asphalt

Presumably, SBS in SBS-modified asphalt increases in apparent volume due to swelling with malten incorporated from asphalt and, at the same time, is dispersed into asphalt, changing from coarse to fine morphologies. Among the components not incorporated into SBS, therefore, malten decreases relatively and asphaltene increases apparently.
The microstructure of SBS-modified asphalt can be broadly divided into the type in which asphalt forms a continuous phase (hereinafter referred to as the "As-network type") and

the type in which SBS forms a continuous phase (hereinafter referred to as the "SBS-network type"). The former corresponds to modified asphalts such as types I and II that are low in SBS concentration and the latter is represented by the high-viscosity type that is high in SBS concentration.

The SBS concentration at which the change from the As-network to SBS-network types occurs is generally said to be 6-8% (Tanaka), (Ueno). We also confirmed that the phase change takes place abruptly at an SBS concentration of about 7%.

When classifying morphologies, it is necessary to consider such differences in continuous phase. Here the morphologies are classified by continuous phase into three types shown in Table 3.

Table 1. Properties of Straight Asphalt.

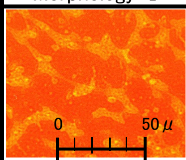
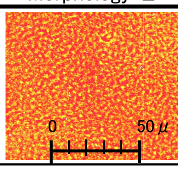
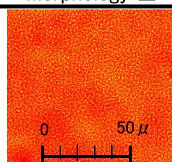
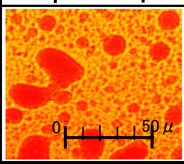
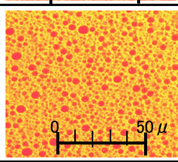
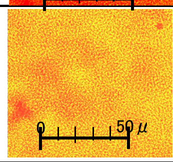
Physical properties	Penetration, 1/10 mm	67
	Softening point, ℃	49.5
Chemical properties	Saturated, %	5.5
	Aromatics, %	53.1
	Resin, %	24.4
	Asphaltene, %	17.0

Table 2. Properties of SBS

Type of molecular structure	Straight chain
S/B*	30/70
Number average molecular weight (Mn)	131,000
Mass average molecular weight (Mw)	152,000

* Mass ratio of styrene to butadiene

Table 3. Classification of Morphologies of SBS Modified Asphalt (400x)

Microstructure	Morphology Ⅰ	Morphology Ⅱ	Morphology Ⅲ
As-network type (SBS concentratio 7% or less)			
SBS-network type (SBS concentration over 7%)			
Definition	State in which asphalt and polymer are compatible with each ather,so the continuous phase is indistinct.Particle size: over 10μm	Stage at which the continuous phase becomes distinct. Particle size:1−10μm	Stage at which particles get into a state of uniform and stable dispersion. Particle size:less than 1μm

3. STORAGE STABILITY OF SBS MODIFIED ASPHALT

3.1 Phenomenon of Material Segregation during Storage

Segregation of SBS-modified asphalt during storage is a phenomenon in which the SBS phase swelled with malten moves up and the asphaltene-rich asphalt phase settles down. In order to explain this phenomenon in terms of morphology, the following storage stability test was conducted.

3.2 Method for Evaluation of Storage Stability

The storage stability was evaluated in the following ways:

(1) Put a sample of dissolved SBS-modified asphalt in a 300 ml tall beaker in which a sampling wire is placed.

(2) Cover the top of the beaker with an aluminum foil and store the sample in a 170 ℃ oven for 7 days.

(3) After completing storage, allow the sample to cool at room temperature for 24 hours, heat the periphery of the beaker with a burner or the like and remove the sample together with the wire. Disconnect the wire from the sample and allow the sample to stand on its side at room temperature (15-25 ℃). When segregation occurs, the material is separated into the SBS and asphalt phases (Photo 1).

(4) The degree of segregation was evaluated by defining as the "segregation rate" the quotient obtained by dividing L1 (length of the portion the SBS phase occupies) by L (depth of the binder) in the beaker (Photo 1).

$$\text{Segregation rate } (\%) = L1\ (\text{cm}) / L\ (\text{cm}) \times 100 \qquad (1)$$

(5) The segregation conditions can be classified into three types:

i) Type that causes segregation

The SBS phase has rubber elasticity and maintains its shape in the solid state without causing elastic deformation, while the asphalt phase elastically deforms and extends in the horizontal direction (Photo 2).

ii) Type that does not cause segregation

In the As-network type , asphalt forms a continuous phase, so the plasticity of asphalt becomes dominant. Therefore, the material flows as a whole in the horizontal direction, resulting in plastic deformation (Photo 3, SBS concentration: 5-7%). In the SBS-network type such as the high-viscosity type, SBS forms a continuous phase, so the material maintains a cylindrical form as a whole and does not undergo plastic deformation (Photo 3, SBS concentration: 9-12%).

iii) Type that shows ambiguous segregation

In either of the networks, segregation may occur with an indistinct boundary between SBS and asphalt phases (Photo 4). In this case, the L1 portion cannot be cleanly determined, so it was measured with reference to the intermediate position between the portions subjected and not subjected to plastic deformation.

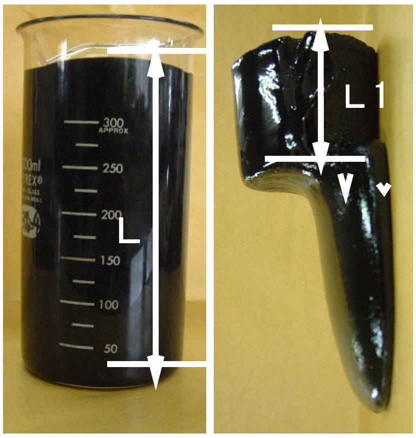


Photo 1. Measurement of Segregation Rate

SBS 5%
(As-network type)

SBS 7%
(As-network type)

SBS 9%
(SBS-network type)

SBS 12%
(SBS-network type)

Photo 3. Segregation Condition of Morphology III
(no segregation)

SBS 5
(As-network type)

SBS 7%
(As-network type)

SBS 9%
(SBS-network type)

SBS 12%
(SBS-network type)

Photo 2. Segregation Condition of Morphology I
(distinct segregation)

Photo 4. Segregation Condition of Morphology II
(ambiguous segregation)

(6) As shown in Photo 2, L1 becomes greater with higher SBS concentration and reaches about 100% at an SBS concentration of 12%. From this, the meaning of creating the condition in which segregation does not occur can be considered as follows:

In the case of the As-network type, a group of SBS particles that form the L1 portion are uniformly dispersed into StAs that forms the network. In the case of the SBS-network, StAs that corresponds to the (L - L1) portion is uniformly dispersed into the SBS-network.
Therefore, the condition in which the As-network type does not undergo segregation is defined as 0% in segregation rate because it means reducing the L1 portion to zero. The condition in which the SBS-network type does not undergo segregation is defined as

100% in segregation rate because it means incorporating all into the L1 portion.

3.3 Relation between Storage Stability and Morphology

The relation between SBS concentration and segregation rate is shown in Figure 1. The number of specimens under the same conditions is three (n = 3).

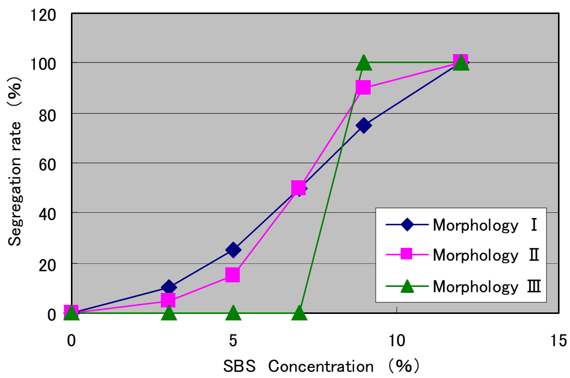


Figure 1. SBS Concentration Vs. Segregation Rate

(1) Segregation rate of SBS-modified asphalt in morphology I
The segregation rate in morphology I tends to increase in proportion to the increase in SBS concentration. The then degree of segregation is shown in Photo 2. The SBS and asphalt phases are clearly segregated into the upper and lower sides, respectively. Hence, it is presumed that most of the SBS content of the SBS-modified asphalt in morphology I moves up, swelled with malten in StAs, and is dissociated from asphaltene. This can also be easily presumed from the fact that the segregation rate is proportional to the SBS concentration.

(2) Segregation rate of SBS-modified asphalt in morphology III
The segregation rate in morphology III is 0% in the region of 7% and less in SBS concentration in which the microstructure shows the As-network type and 100% in the SBS-network region of over 7% in SBS concentration. That is, the SBS-modified asphalt in morphology III can be said to exhibit excellent storage stability at any SBS concentration (Photo 3).

(3) Segregation rate of SBS-modified asphalt in morphology II
The relation between SBS concentration and segregation rate in morphology II is in between those in morphologies I and III. The condition after the storage test is shown in Photo 4, the material showing ambiguous segregation that can also be said to be an intermediate between the segregations in morphologies I and III. That is, the material is not segregated so clearly as in the case of morphology I, but can be said not to have attained the storage stability that is comparable to that in morphology III.

3.4 Summary

From these test results, the following can be said:

(1) A morphology finer than morphology III (particles dispersed mostly in sizes less than 0.001mm) is necessary to obtain SBS-modified asphalt that is excellent in storage stability.

(2) With morphology II (particles dispersed in sizes of 0.001 ～ 0.01mm), sufficient storage stability cannot be obtained.

(3) With morphology I (particles dispersed in sizes over 0.01mm), complete segregation is likely to occur, so it can be said that trouble may take place in asphalt plants.

4. MECHANICAL PROPERTIES OF SBS MODIFIED ASPHALT

4.1 Evaluation of Binder Properties

The test method of binder properties are given in Table 4. In addition to the ordinary tests used so far, bending test (Tanaka), (Moyomatsu,2001), (Ohki), (Motomatsu,2004) was added as a new test method regarded as having a high correlation with mixture properties. Of these test items, toughness/tenacity test and bending test were conducted in the following ways.

Table 4. Test Method for Binder Properties

Test Method	Test conditions
Penetration test	25 ℃
Softening point test	$T_{R\&B}$
Ductility test	4 ℃
Toughness/tenacity test	25 ℃
Bending test	Test temperature: -20 ℃ Specimen size: 120 x 20 x 20 mm Span: 80 mm Loading method: 3 points loading Loading speed: 100 mm/min

a) Toughness-tenacity test

Toughness/tenacity test is a test proposed by Benson in 1955 (Benson). In this test, a metal hemisphere of a fixed size is buried in an asphalt sample in a given container, with its hemispherical surface downward, and drawn out therefrom with a fixed speed at a given temperature, and the workload required to draw it out is determined from the load vs. displacement curve (Figure 2). Toughness is the sum of the area enclosed by the line connecting points A, B and E in the figure and the area enclosed by the line connecting points C, D, F and E. Tenacity is determined as the area enclosed by the line connecting points C, D, F and E. The test conditions are shown in Photo 5.

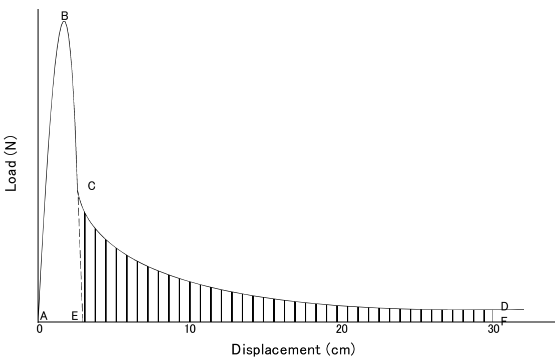


Figure 2. Load Vs. Displacement Curve (Toughness-Tenacity)

Photo 5. Toughness-Tenacity Test

b）Bending test

A binder specimen in the shape of a rectangular parallelepiped as shown in Table 4 was prepared and tested for bending under the conditions of Photo 6, and the bending workload was determined by the method shown in Figure 3.The bending workload obtained from this test is determined from the product of maximum stress and strain and is considered to represent the bonding strength（breaking energy of adhesive）of SBS-modified asphalt that makes aggregate particles adhere to each other.

$$W_b = \sigma \times \varepsilon \qquad (2)$$

where W_b : bending workload (MPa)
σ : maximum stress
ε : strain

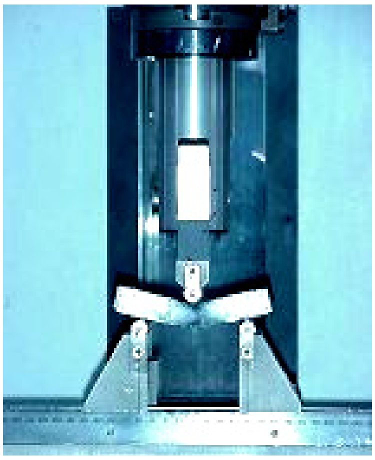


Photo 6. Binder Bending Test

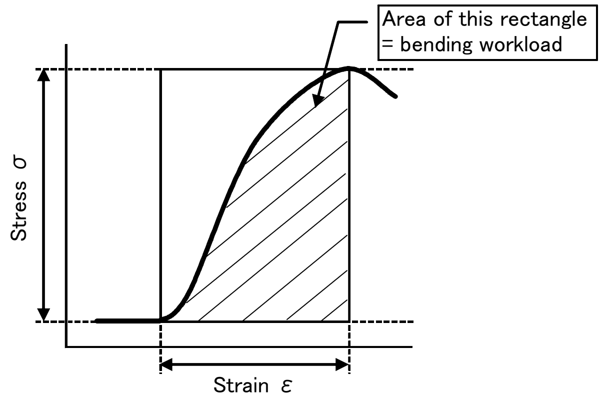


Figure 3. Bending Workload

4.2 Relationship between Morphology and Binder Properties

The relationship between morphology and binder properties is described below, taking as a typical example SBS-modified asphalts in the case of morphologies I and III.

a) Softening point

The relationship between SBS concentration and softening point is shown in Figure 4. In any morphology, the softening point tends to become higher with higher SBS concentration. The softening point in morphology III rises from 48 ℃ to no more than 55 ℃ at SBS concentrations of 7% and less, shows an abrupt rise to about 90 ℃ when the SBS concentration exceeds 7% and tends to get settled at about 92 ℃ at higher SBS concentrations. That is, it is presumed that softening points of 55-90 ℃ do not exist in morphology III. The softening point in morphology I is higher than that in morphology III at any SBS concentration and does not tend to change abruptly at SBS concentrations around 7% as in the case of morphology III. These trends closely resemble those in Figure 1 obtained from the storage stability test.

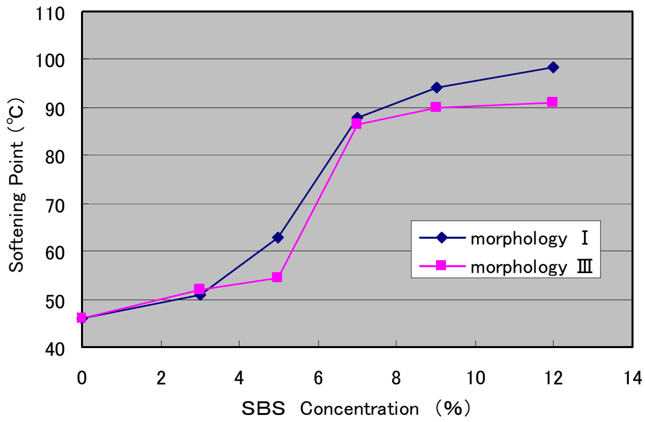


Figure 4. SBS Concentration Vs. Softening Point

b) Toughness

The relation between SBS concentration and toughness is shown in Figure 5. The toughness in morphology I tends to simply increase with increasing SBS concentration. The toughness in morphology III increases with increasing SBS concentration. However, it tends to begin decreasing at the SBS concentration of 7%. That is, the toughness in the region of the As-neteork shows a trend quite contrary to that in the region of the SBS-network type. In morphology III, the cohesiveness of the binder is considered to increase as compared with that in morphology I. Therefore, the cohesiveness of the binder itself becomes greater than the bonding strength of the binder to the tension head that is a test jig, so the binder comes off the tension head during the test. Presumably, this fact accounts for the above-mentioned decrease in toughness (Ohno).

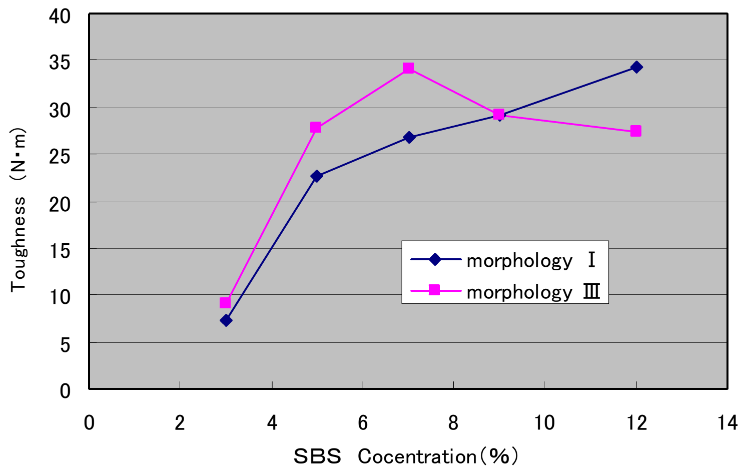


Figure 5. SBS Concentration Vs. Toughness

c) Ductility

The relation between SBS concentration and ductility (at 4 ℃) is shown in Figure 6. The ductility tends to increase with increasing SBS concentration. The ductility in morphology I in which particles are coarsely dispersed shows a greater value than that in morphology III.

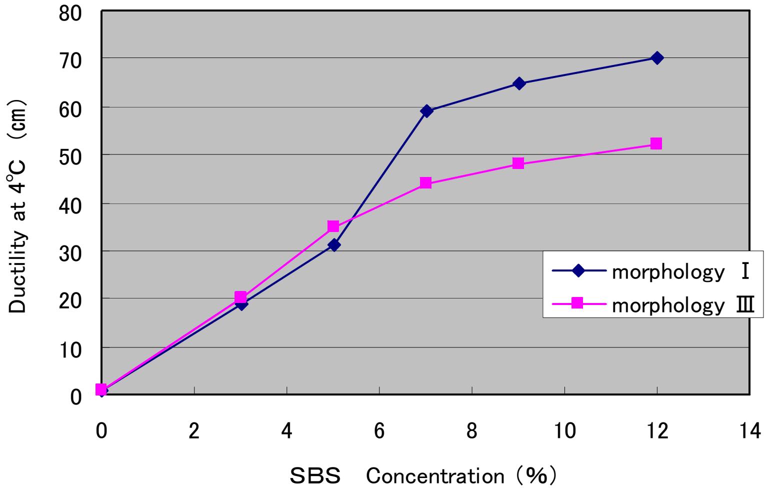


Figure 6. SBS Concentration Vs. Ductility

d) Bending workload

The relation between SBS concentration and bending workload is shown in Figure 7. The bending workload tends to increase with increasing SBS concentration. The magnitude relation between morphologies I and III varies with the SBS concentration, so a definite relation cannot be obtained. This is because cases in which the specimens do and do not fracture during the test are coresident, depending on the combination of SBS concentration and morphology. For example, many specimens fracture at the concentration of 9% in morphology III and at 12% in morphology I.

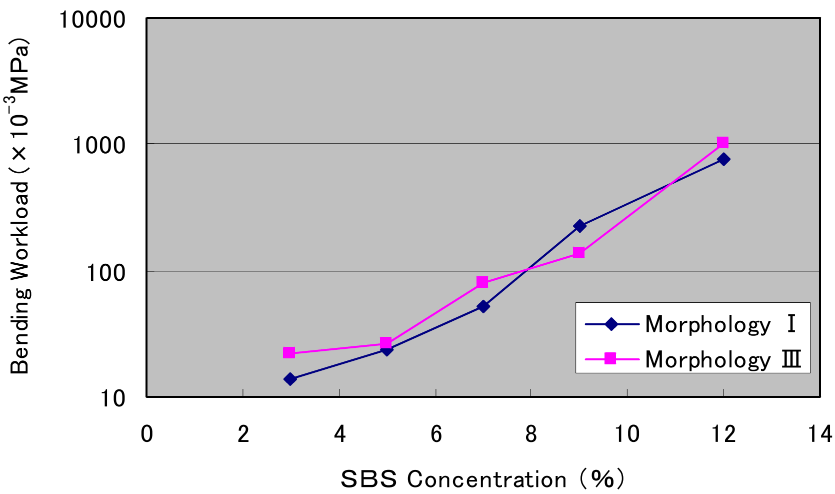


Figure 7. SBS Concentration Vs. Bending Workload

4.3 Summary

From the above-mentioned binder test results, the following information was obtained:

(1) The characteristic values obtained from the evaluation tests of binder properties vary with the morphology.

(2) Of the test values obtained by various test methods investigated herein, some tend to be proportional to the SBS concentration and some do not show such a trend.

5. MIXTURE PROPERTIES OF SBS MODIFIED ASPHALT

5.1 Material

The mixture investigated herein is an ordinary mixture for porous asphalt that has a void ratio of 20% and an asphalt content of 4.9%. The mix ratios and particle sizes of aggregates used for this mixture are shown in Table 5 and Figure 8.

Table 5. Mix Ratios and Sources of Aggregates

Item	Mix ratio	Source
crushed stone (4.75 ～ 13.2mm)	83.0	Okutama, Tokyo
Fine sand (0 ～ 2.36mm)	12.0	Moriya, Ibaraki
Filer	5.0	Calcium carbonate

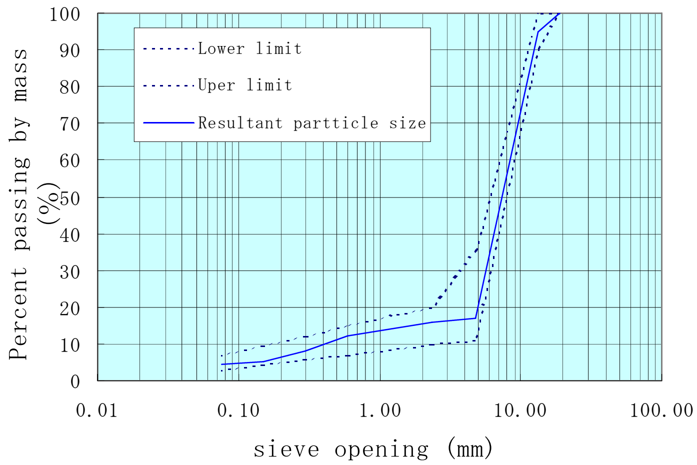


Figure 8. Grading Curve

5.2 Test Method

Wheel tracking test and fatigue test were conducted to evaluate mechanical properties of asphalt mixture. The test conditions are shown in Tables 6 and 7.

a) Wheel tracking test

In this test, an asphalt mixture roller compacted to a size of 30 cm x 30 cm x 5 cm was used as a specimen, and wheel shown in Table 6 were repeatedly run back and forth on the specimen at 42 pass/min . The degrees of plastic flow at 45 and 60 minutes were determined from this curve (Figure 9), and the dynamic stability was calculated by equation (3):

$$DS = 42 \times (t60 - t45) / (d60 - d45) \tag{3}$$

where DS: dynamic stability (pass/mm)
d45: displacement at 45 minutes (mm)
d60: displacement at 60 minutes (mm)

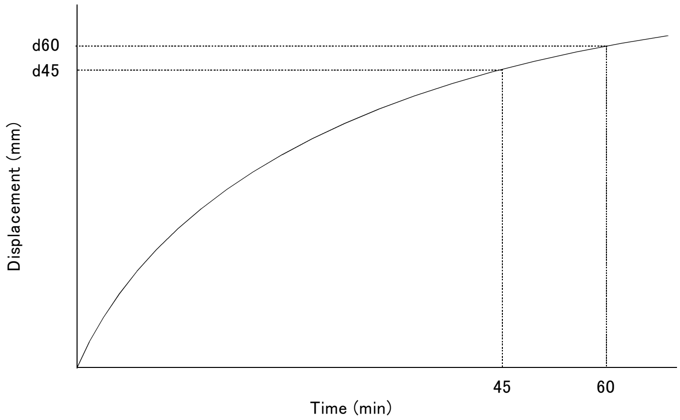


Figure 9. Time Vs. Displacement Curve

Table 6. Wheel Tracking Test Conditions

Wheel	Type	Solid tire
	Size	200 mm ϕ x 50 mm wide
	Rubber hardness	JIS hardness 78 (at 60 ℃)
Applied load		686N
Loading method		Vertical
Running distance		230mm
Running speed		42pass/min
Test temperature		60 ℃

b) Fatigue test

In this test, a block 4 cm x 4 cm x 30 cm cut from the mixture roller compacted to a size of 30 cm x 40 cm x 6 cm was used as a specimen. Using the equipment shown in Figure 10, the specimen was repeatedly loaded under the conditions of Table 7 . The test was conducted with strains varied from 0.02% to 0.08% by the sinusoidal strain control method. The number of cycles to failure was defined in this study as the point of intersection of lines La and Lb from the number of repetitions cycles vs. complex modulus curve shown in Figure 11.

Table 7. Fatigue Test Conditions

Frequency	5Hz (sinusoidal wave)
Temperature	10 ℃
Control method	Strain
Length of span	30 cm
Distance btw supporting point and loading point	10 cm
Loading method	Two-point loading with two supporting points

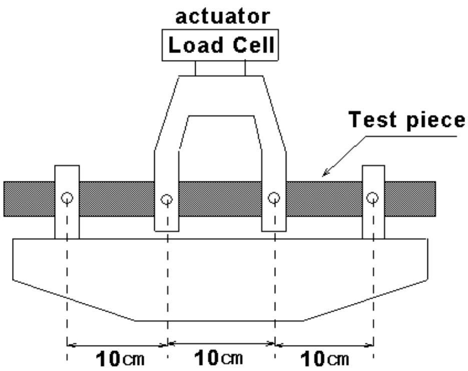


Figure 10. Test Equipment

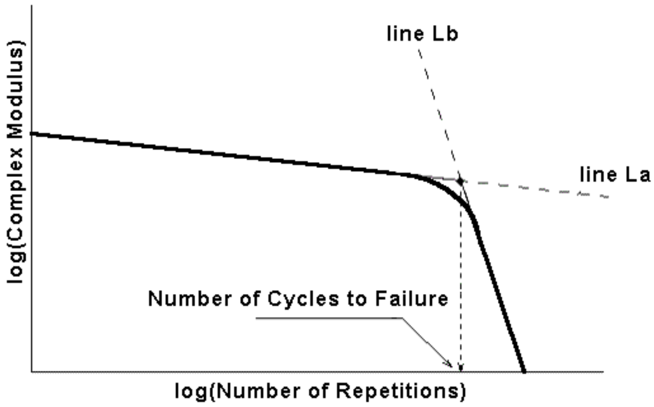


Figure 11. Number of Reptitions Cycles Vs. Complex Modulus

5.3 Relation between Mecanical Properties of mixture and Morphology of SBS asphalt

The relation between mixture properties and morphology is described below, taking as a typical example SBS-modified asphalt in the case of morphologies I and III.

a) Dynamic stability

The relation between SBS concentration and dynamic stability is shown in Figure 12. The dynamic stability tends to increase with increasing SBS concentration, but shows no difference between morphologies I and III.

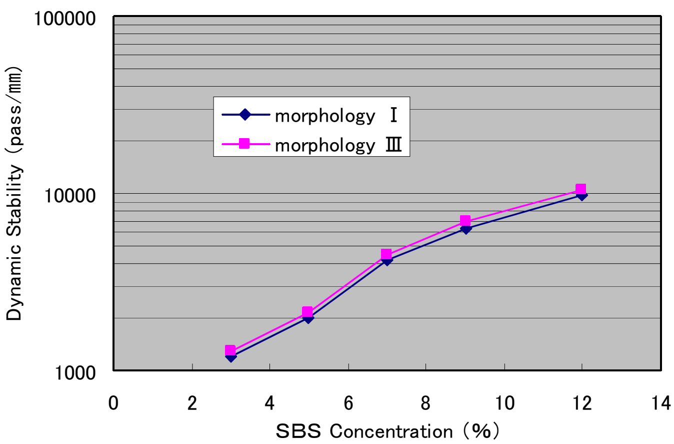


Figure 12. SBS Concentration Vs. Dynamic Stability

b) Number of cycle to failure

The fatigue curves at SBS concentrations of 5% and 12% are given in Figure 13. The number of fractures is greater with higher SBS concentration as is the case with other mixture properties. At the SBS concentration of 5%, the number of fractures under a strain of 4 x 10^{-4} is 1,300 in morphology I and 4,000 in morphology III. At the SBS concentration of 12%, the number of fractures is 20,000 in morphology I and presumably exceeds 1,000,000 in morphology III.
That is, it can be said that the number of fatigue fractures is proportional to the SBS concentration and is greatly affected by the difference in morphology. Even at the same SBS concentration of SBS-modified asphalt, such an excellent fatigue resistance cannot be

obtained without enhancing the morphology to a fine dispersion level.

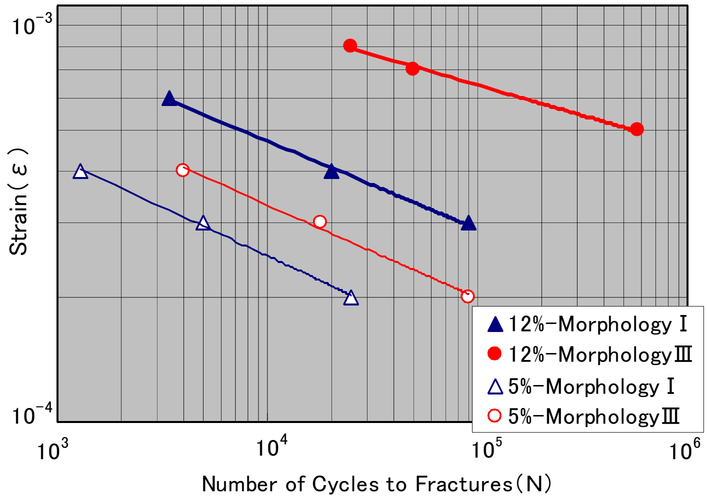


Figure 13. Strain Vs. Number of Cycles to Fractures

5.3 Summary

From the above-mentioned results, the mixture properties improve with an increase in SBS concentration, but the degree of improvement thereof is affected by the morphology of SBS-modified asphalt, so it can be said that a binder having a finer morphology exhibits better performance.

6. CONCLUSION

As a result of this study, it became clear that various performances of SBS-modified asphalt are better with higher concentration of SBS as the modifying agent, but are greatly affected by its morphology. Therefore, it is apparent that binder performance evaluations should be done with importance on the effect of the morphology. Concrete examples of such evaluations are given below:

(1) The storage stability of SBS-modified asphalt depends on the morphology. In order to obtain an SBS-modified asphalt that is excellent in storage stability, it is necessary to make the degree of dispersion finer than that in morphology III. Dispersed particles of SBS or StAs should have a particle size less than 1 μm.

(2) Various test values obtained from the evaluation tests of binder properties also vary with the morphology. In order to grasp the performance of the binder, it is necessary to obtain a definite morphology for evaluation.

(3) The mixture properties improve in proportion to the SBS concentration, but greatly vary in value with the morphology as is the case with the binder properties. It became clear that a fine dispersion level in excess of morphology III has to be obtained in order to make the performance of SBS-modified asphalt fully reflect on the mixture properties.

(4) As the mixture properties are proportional to the SBS concentration, any binder evaluation method would lose its significance unless it shows a certain relation with the SBS concentration.

In recent years, SBS-modified asphalt has rapidly found widespread application, supported by many R&D efforts, and has served the needs of diversifying pavements. This binder

cannot be obtained by merely adding SBS as a modifying agent. A morphology control technology is indispensable for its practical realization.
In the future, we intend to cope with the remaining problems such as the mechanisms of SBS dispersion and material segregation, relation between morphology and composition of raw asphalt, relation between workability and morphology, new test methods for evaluation of binder performance other than bending test, etc. and thereby to attain further technical development and dissemination of SBS-modified asphalt.

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