Discrepancies in Rheological Properties of Modified Asphalt Cement

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Abstract. DSR device was used to investigate the variations in linear and non-linear viscoelastic limitations with both pure and polymer modified asphalt film binders. Different test temperatures and strain rates were used to measure the change in the rheological response of the thin film of different asphalt binders and determine the variations in rheological data due to non-linear impacts by using dynamic properties of asphalt binder and phase anlage. The experimental results showed that the addition of SBS modifier to asphalt binder increased the viscous part of complex shear modulus and then increased dissipated energy for Durah asphalt cement vice versa Nasiriyah asphalt cement. Also, the SBS addition decreased in variations of the viscous part of complex shear modulus due to the decrease in phase angle.

Keywords; Asphalt Binder, SBS modifier, Rheological Properties, Dynamic shear rheometer (DSR), Black Diagram (BD), Cola-Cola diagram.

1. Introduction

Asphalt cement is defined as a viscoelastic substance that possess both of elastic property and viscous behaviors. It's well-known that Dynamic Shear Rheometer (DSR) is considered as a better device comparing with traditional methods that utilized to predict asphalt properties as a binder that relates to asphalt binder’s performance in-field (the experimental methods of assessing asphalt cement) [1].

DSR test is utilized to measure the rheological properties of the film of asphalt cement. This test concentrate on viscoelasticity and adjusting between the obstacles of using the tests to determine the rheological properties and the requirements of practical tests developed from the common simplistic empirical ways that qualified asphalt cement [2].
The DSR device provides a simple way to measure the viscoelastic properties of asphalt binder [3]. Two different configurations of the device, both of them provide the elastic and viscous characteristics of asphalt cement; torsional test and sliding plate test [4]. Many researches were published that utilized the (DSR) to investigate the fatigue characteristics of asphalt cement [5,6]. In these researches, measuring fatigue by continuing applying load cycles until a reduction occurs in the complex shear modulus ($G^*$) growth with loading time. Bahia et al. (1999) utilized a set-up of plate-plate geometry [5], while in (1998) Phillips utilized cone-plate geometry [7]. Bahia et al. (1999) performed many different tests with in the linear and nonlinear ranges of viscoelasticity and with a constant load cycles ($5 \times 10^3$ or $11 \times 10^3$ with strains of 1, 10, 20 and 50%). Materials failure determined by the reduction in complex shear modulus $G^*$ value. Also, in these researches, measurement and the assessment of asphalt cement recovery were considered. It was exhibits that both strain dependency and fatigue are very susceptible to binder composition, additives types, temperature, the rate of heating, aging processes and the interaction between these factors [5].

Both complex shear modulus ($G^*$) and phase angle ($\delta$) parameters can be calculated by using DSR test. $G^*$ represent the capability of materials to resist any types of deformations under the applying loads [4]. And can be utilized as an indicator to predict asphalt binder film’s stiffness at the specific loading rates and temperatures. Complex shear modulus is the ratio between the value of complex shear stress and the value of complex shear strain. $G^*$ consist of in to two parts; the elastic part (which is the storage modulus ($G'$), and the viscous part (which is the loss modulus ($G''$)) [8].

The phase angle ($\delta$) is another parameter determined from DSR test’s results that used as a indicator of viscoelastic balance of behavior of materials and can be defined as the shifting in phase between the applying complex shear stress and complex shear strain response during the tests. The values of ($\delta$) for elastic material equal to $0^\circ$ while for and viscous materials is equal $90^\circ$, and $\tan \delta = (G''/G')$ is the ratio of viscous contribution and elastic contribution. When phase angle’s value equal to zero this mean both the stress and strain were completely in phase and thus they were totally elastic [3] because of that; shear strain generated from the utilized load is totally recoverable [9]. The residual storage and loss modulus relation can be demonstrated by the plane of Cartesian coordination system as shown in figure (1).

![Figure 1](image1.png)

Figure1. The components complex shear Modulus ($G^*$) ($\tau_{\text{max}}$: maximum complex shear stress; $\tau_{\text{min}}$: minimum complex shear stress; $\gamma_{\text{max}}$: minimum complex shear strain; $\gamma_{\text{max}}$: maximum complex shear strain and $\delta$: Phase angle) [4].

When the phase angle value equal to $0^\circ$ would represent a material that was perfectly solid elastic. At $90^\circ$, phase angle represents that the asphalt film’s behavior is perfectly viscous. This means ($\delta$) value will be changing between $0$ to $90^\circ$ and the behavior of asphalt cement varying between the elastic and viscous behavior.
2. Research Objectives
The rheological responses of thin-film asphalt binders were measured with the variation of load frequency and testing temperatures by utilizing DSR; to obtain the following goals;
1. Investigate the changes in both elasticity and viscosity components of asphalt film binders with different test temperatures.
2. Determine variations in rheological data due to non-linear impacts.

3. Experimental Works

3.1. Materials
3.1.1. Asphalt cement
Two types of asphalt cement with AC 40-50 were utilized in the experimental work provided from Al-Nasiriyah and Al-Durah refineries in the middle and south of Iraq; their physical properties were presented in table 1.

Table 1. The physical neat asphalt binders properties.

| Test                          | Nasiriyah | Durah |
|-------------------------------|-----------|-------|
| Penetration Test- ASTM D5/     | 41.2      | 43    |
| Softening point test (°C) – ASTM D36/ | 52       | 53.1  |
| Penetration Index             | -1.00     | -0.35 |
| Viscosity test cP, 135 °C) ASTM D 4402 | 500.1    | 481   |

3.1.2. Asphalt Modifier
The performance of asphalt cement can be modifying by using a different types of modifiers to improve its durability. The modifiers of asphalt cement are two types of (SBS) with different percentages (3, 6, 9%) were used .The physical properties of SBS are presented in table 2.

Table 2. SBS modifier properties

| Test Parameters      | Test specification | Units  | SBS (D 1101 K) | SBS (D 1184 K) |
|----------------------|--------------------|--------|----------------|----------------|
| Tensile strength,    | ASTM D412          | Psi    | 4.6*10³        | 4*10³          |
| Elongation,           | ISO 37             | %      | 880            | 820            |
| Specific gravity     | ISO 2781           |        | 0.94           | 0.94           |
| Dynamic viscosity    | ASTM D 4402        | cps at 77°F | 4*10³     | 2*10⁴         |
| Di-block,            | %                  | 16     | 16             |                |
| 300% modulus,        | ISO 37             | Psi    | 400            | 800            |
| Molecules Structure  |                    |        | Linear         | Radial         |

4. Experimental Procedures
The samples of asphalt binders were heated up in an oven to a degree of temperature not exceeding (160 °C ±5 °C) for approximately 1 hour for the neat asphalt binder and (180 °C ±5 °C) for the modified asphalt cement in order to soften enough and pour it easily; then, the samples were poured into prepared silicone molds.
Using Kinexus Pro⁺ rheometer with rotational rheometer system from the Malvern Company to calculate the viscoelastic characteristics of asphalt cement by applying controlled shear strain mode on the specimen under testing to measure the flow properties (like the shear viscosity obtained from test of flow) and dynamic characteristics of material (like phase angle (δ) and the viscoelastic
modulus obtained from oscillation test). Figure 2 show the Kinexus Pro+ DSR device and the Silicon moulds in fridge.

The rSpace program ver.1.7 that advanced by the Malvern Company is used to determine (G*), and (δ) also, the storage modulus (G’), loss modulus (G’’), applied frequency, testing temperature, and dynamic shear viscosity. The test parameters in this work were shown in table 3.

| Test parameter                  | Test value       |
|---------------------------------|------------------|
| Total time of the test          | 1 hr             |
| Equilibrium period for each temperature | 600 sec         |
| Measurement period              | 10 sec           |
| Test Temperatures               | 5,15,25,35,50 and 60 °C |
| Start frequency                 | 10 Hz.           |
| End frequency                   | 0.1 Hz.          |
| Shear strain ratio              | 1.0% during LVER |
| Control mode test               | Shear strain     |

According to (BS EN 14770:2012); two types of plate geometries were utilized for the DSR test:
1. A plate of diameter (8) mm and (2) mm gap at low temperature ranging between (5) °C and (25) °C.
2. A plate of diameter (25) mm and (1) mm gap for intermediate and high temperatures ranging between (35) °C and (60) °C.

The test procedure of the test can be described as presented below:
1. The results of (G*), (δ) and dynamic shear viscosity were gathered over a wide ranges of temperature (e.g. between (5) to (60 °C with temperatures interval every (10 °C) and frequencies (e.g. 0.1, 0.1259, 0.1585, 0.1995, 0.2512, 0.3162, 0.3981, 0.5012, 0.631, 0.7943, 1.0, 1.259, 1.585, 1.995, 2.512, 3.162, 3.981, 5.012, 6.31, 7.943 and 10 Hz). This combination of variables will produce 126 sets of data for each trial.
5. Modified Cole-Cole Diagram
Modified Cole-Cole Plots or called Han Plots explain variation between two components of complex shear modulus (elastic part \( G' \) plus viscous part \( G'' \)). The relative changes in both elasticity and viscosity components of asphalt film with different test temperatures can be interpreted by a graph. Airey, 2002b; explain Han Diagram for modified asphalt binder by polymer with different test temperatures. When the value \( G' = G'' \) (the straight line is plotted; its intercept with the Han curve to explain crossover frequency between complex shear modulus components \( G' \) and \( G'' \)). The Data below and on the right of this straight line explain the behavior that controlled by viscous or low shear modulus. Whereas the data up or on the left of the straight line present that the elastic component of complex shear modulus dominates the behavior of asphalt cement specimen within test conditions [10].

Figure 3. Variation between viscous component and elastic component of \((G^*)\) of Durah asphalt cement.

Figure 4. Variation between viscous component and elastic component of \((G^*)\) of Nasiriyah asphalt cement.
Table 4. Regression parameters of asphalt binder

| Modifier Type | Modifier Percentage | Durah Asphalt Cement | Nasiriyah Asphalt Cement |
|---------------|---------------------|-----------------------|--------------------------|
|               | a   | b   | R²  | a   | b   | R²  |
| Neat          | 0   | 289,148 | 1.636 | 0.991 | 233,902 | 1.6587 | 0.991 |
|               | 3%  | ---   | --- | --- | 197,118 | 1.8184 | 0.991 |
| SBS 1101      | 6%  | 371,119 | 0.7833 | 0.993 | 196,392 | 1.5771 | 0.993 |
|               | 9%  | 1,000,000 | 0.7101 | 0.999 | 325,196 | 1.4856 | 0.992 |
| SBS 1184      | 6%  | 576,725 | 1.3233 | 0.987 | 216,966 | 1.8286 | 0.992 |
|               | 9%  | 668,320 | 0.9577 | 0.988 | 274,413 | 1.6446 | 0.991 |

Depending on the results in table 4 and figures 3 and 4, it can be concluded:

1. The parameter (a); refers to the intercept trend line of experimental results with the y-axis (viscous part of complex shear modulus (G/)); (loss storage modulus).
2. The increased in the magnitude of G// meaning increased the magnitude of the phase angle (increasing the rutting parameter).
3. The addition of SBS modifier causes increases in viscous part of (G*) leading to increase values of the dissipated energy of Durah asphalt cement vice versa Nasiriyah asphalt binder.
4. While the addition of SBS to Nasiriyah asphalt binder decrease the viscous part and reduce the value of phase angle and decrease the rutting parameter leads to improve the resistance of asphalt binder to rutting deformation.
5. The parameter (b); means the slope of the trend line (Δy/Δx) can be used this parameter to explain the viscous behavior of asphalt cement; under constant change of Δx. The experimental result explains;
   a) Increased b; means increased in variations of viscous part of complex shear modulus due to increased change in phase angle and increased dissipated energy.
   b) Reduced b; means decreased in variations of viscous part of G* due to reducing in phase angle.
6. Adding SBS modifier reduced b for asphalt cement.

6. Black Space Diagram

A black space diagram can be described as a graph of the value (norm) of the complex shear modulus versus the phase angle, δ, gained from the dynamic test at all test frequencies and temperatures.

This graph is too sensitive to morphology, counter to the master curve, and the black space diagrams do not need any shifting, and therefore can construct temperature-independent curves when the asphalt binder follows time-temperature superposition.

Typically, reducing phase angle and increasing complex shear modulus as decreasing in temperatures is describing the viscoelastic behavior of materials [11].

The reasons that cause discontinuous in black space diagram are:

1. Testing the sample within the non-linear region;
2. The tested sample is not rheological (i.e., the transitions of the phase angle happens over the studied range of frequency and temperature).
Black space diagrams were used to determine variations in rheological results because of the non-linear impacts before attempts are made for shifting the results to make smooth and continual master curves [12].

The asphalt specimens tested under controlled strain mode, the value of applied strain within the LVER presented in table (3).

![Figure 5. A black diagram displaying the rheological behavior of pure asphalt and three percentages of SBS 1101.](image)

It can be seen from figure 5, added SBS 1101 reduced G* of local asphalt cement due to increased phase angle.

![Figure 6. A black diagram displaying the rheological behavior of pure asphalt and three percentages of SBS 1184.](image)

It can be concluded from figure 6; when adding 3% SBS 1184 improved the complex shear modulus of local asphalt cement and increased phase angle.
The upper value of complex shear modulus that shown in table 5 can be predicted by drawing the relationship between the logarithm of G* (Pa) and the logarithm value of (1+tan (δ°)); the value of x-axis ranging between 0 and 1 as shown in figure 7 and 8.

Figure 7. Relationship between complex shear modulus with logarithm value of (1+tan (δ°)) for pure and modified asphalt.

Figure 8. Relationship between complex shear modulus with logarithm value of (1+tan (δ°)) for pure and modified asphalt.
Table 5. Max. Value of Complex Shear Modulus (Pa) of asphalt binder

| Modifier Type | Modifier Percentages | Durah Asphalt Cement | Nasiriyah Asphalt Cement |
|---------------|----------------------|-----------------------|--------------------------|
| Neat          | P                    | ----                  | 5000000                  |
|               | 3%                   | ----                  | 7000000                  |
| SBS 1101      | 6%                   | 6000000               | 4000000                  |
|               | 9%                   | 1000000               | 5000000                  |
|               | 3%                   | 8000000               | 6000000                  |
| BS 1184       | 6%                   | ----                  | 8000000                  |
|               | 9%                   | 3000000               | 9000000                  |

7. Conclusion

The experimental results analysis of two types of asphalt cement were showed the following conclusions from the finding of the present investigation:

1. Increasing the viscous part of the (G*) due to the addition of SBS and then the dissipated energy of Durah asphalt binder increased vice versa Nasiriyah asphalt binder.
2. The addition of SBS to Nasiriyah asphalt binder decreased the viscous part of (G*) and reduce (δ°) and decrease AASHTO parameter that leads to improve rutting resistance.
3. Increasing the rutting parameter as a result of increased shear complex modulus (G*) due to decrease the phase angle.
4. The parameter (a); refers to the intercept trend line of experimental result with the y-axis (viscous part of complex shear modulus G'); loss storage modulus).
5. The parameter (b); means the slope of the trend line (Δy/Δx) can be used this parameter to explain the viscous behavior of asphalt cement; under constant change of Δx. The experimental result explains;
   a) Increased b; means increased in variations of viscous part of complex shear modulus due to increased change in phase angle and increased dissipated energy.
   b) Reduced b; means decreased in variations of viscous part of complex shear modulus due to the decrease in phase angle
6. Adding SBS modifier reduced b meaning decreased in variations of viscous part of (G*) for local asphalt cement due to the decrease in phase angle.

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