Investigation of Rheological Properties of Iraqi Asphalt Cement

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Abstract

The rheological properties of local asphalt cement are measured with the aid of Dynamic Shear Rheometer device to investigate the linear viscoelastic limitation with the range of asphalt film binder involving polymer –modified asphalt. The rheological response of the asphalt film binders at different temperatures and rates of deformation was measured using dynamic properties of complex modulus and phase anlage. The specific gravity of asphalt cement for Nasiriyah refinery is greater than that of asphalt cement produced from Durah refinery and reduced when adding SBS. The chemical analysis of asphalt cement of Nasiriyah and Durah asphalt cement are 83.47% and 82.27% carbon element; 9.87% and 9.71% Hydrogen element and 0.36% Nitrogen element for both sources; these elements increased with adding SBS modifier.

Keywords: Asphalt Cement, Dynamic shear rheometer, Rheological Properties.

1. Introduction

The Dynamic Shear Rheometer is widely recognized as the better device used to predicting the properties of asphalt cement as a binder that relate to performance of asphalt binder in field (experimental ways of evaluating asphalt) (Al-Haddad, 2015).

The properties of the asphalt film are calculated with the aid of Dynamic Shear Rheometer. The DSR test focuses on viscoelasticity and compromise between the difficulty of rheological testing and the need of practically purposeful test advanced from the historically widespread simplistic empirical methods of qualifying asphalt (Anderson, et.al., 1991 cited in Bahia, 2009).

The DSR allows for measurement of viscoelastic characteristics of asphalt to be obtained simply. (Read, 1991b, Thom, 2008). There are two varying configurations, both gives the elastic and viscous characteristics of a substance; the sliding plate test and torsional test (Read, 1999b). Bitumen is a viscoelastic material and exhibit both elastic and viscous behavior. DSR is capable of qualifying elastic and viscous properties of the asphalt binder (ASTM D7175 – 08, 2008 and BS EN 14770:2012). The DSR device can be used for measuring the following parameters; the complex shear modulus and phase angle. The complex shear modulus (G*) is divided into two parts; elastic part (strong modulus – G’) and viscous part (loss modulus – G’). The complex shear modulus (G*) can be defined as the ratio between absolute magnitude of complex shear stress divided by the absolute magnitude of complex shear strain, (neglecting any time loss between the two responses due to the effect of viscosity). The ability of material to resist any deformation under applied load can be represented by G* (Bahia, 2009). The (G*) factor used as indicator to predict the stiffness of an asphalt film at the required rate of loading and temperature.
Another parameter that can be obtained from the result of DSR test is the phase angle ($\delta$), which is the difference between complex shear stress and strain. When the value of phase angle is equal to zero that means the stress and strain are perfectly in phase and therefore, perfectly elastic (Thom, 2008) so that; the shear strain resulting from applied load is completely recoverable (Asphalt Institute, 2007). The relation between the residual loss and storage modulus can be explained by plane Cartesian coordinate system as drawn in figure (1).

![Figure 1. Complex shear Modulus components. ($\tau_{\text{max}}$: Max. complex shear stress; $\tau_{\text{min}}$: Min. complex shear stress; $\gamma_{\text{max}}$: Max. complex shear strain; $\gamma_{\text{min}}$: Min. complex shear strain; $\delta$: Phase angle)](image)

The value for phase angle will be varying between 0 and 90° for the behavior of asphalt binder behavior changing between elastic and viscous properties.

2. The Research Objective:
The rheological properties of asphalt binders are measured with different load frequency and test temperature using DSR; to achieve the following objectives;
1- Investigate the linear viscoelastic region for different bituminous binders.
2- Determine the effect of various modifications on the rheological behavior of asphalt binder.
3- Determine the effect of different modification on the dynamic properties of various bituminous binders within the linear viscoelastic region.

3. Laboratory work
3.1 Materials Used
3.1.1 Asphalt cement. Two types of asphalt cement brought from Durah and Nasiriyah refineries (middle and southern region of Iraq) were used and tested. The physical properties of asphalt cement used are indicated in table (1).

| Test                                      | Nasiriyah | Durah |
|-------------------------------------------|-----------|-------|
| Penetration Test (0.1 mm, 100 g and 5 sec)- ASTM D5/ | 41        | 43    |
| Softening point test ($^\circ$C) - ASTM D286/ | 52        | 52    |
| Penetration Index (PI) parameter          | -1.0      | -0.35 |
| Viscosity test ($cP$, 135 $^\circ$C) - ASTM D 4402/ | 500       | 481.3 |

3.1.2 Asphalt Modifier. In order to improve the performance of asphalt cement used polymer to increase the asphalt’s durability, resistance to thermal cracking and increase the rutting resistance
of flexible pavement is one of the major causes; when using polymer-modified asphalt (AI Institute, 2007). A wide range of materials is used to improve asphalt cement:

a) Elastomers types

In this study, two types of Styrene-Butadiene-Styrene (SBS) were used, the physical properties of these types as shown in table 2.

| Test Parameters       | Test specification | Units | D 1101 K-SBS | D 1184 K-SBS |
|-----------------------|--------------------|-------|--------------|--------------|
| Tensile strength,     | ASTM D412          | psi   | 4,600        | 4,000        |
| Elongation            | ISO 37             | %     | 880          | 820          |
| Specific gravity      | ISO 2781           |       | 0.94         | 0.94         |
| Dynamic viscosity     | ASTM D 4029        | cPs at 77°F | 4,000      | 20,000       |
| Di-block              | %                  | 15 %  | 16 %         |              |
| 300% modulus          | ISO 37             | psi   | 400          | 800          |
| Molecular structure   | Linear             |       | Radial       |              |

b) Micro silica

Non-porous spheres of silicon dioxide (silica, SiO$_2$) are used. The average particle size of the particles is around 0.15 microns. The test results of Micro silica are according to ASTM C989/C989M-13 and BS EN 15167-1, 2-2006 and shown in the table 3.

| Chemical and physical requirements | Specification (characteristic values) |
|------------------------------------|---------------------------------------|
| SiO$_2$ (%)                        | > 90                                  |
| H$_2$O (moisture content when packed, %) | > 90                                 |
| Loss on ignition, LOI (%)          | < 3.0                                 |
| Retained on 45 micron sieve (tested on un-densified, %) | < 1.5                                |
| Bulk density - un-densified (when packed, kg/m$^3$) | 200 - 350                             |
| Bulk density - densified (when packed, kg/m$^3$) | 500 - 700                             |

The specification refers to analysis performed using the Elkem Standard Test Methods for Microsilica (Microsilica, 2014).

c) Ground Granulated Blast Furnace Slag (GGBS)

GGBS is a by-product from the blast-furnaces used to make iron. These materials operate at a temperature of about 1,500 degrees centigrade and are fed with a carefully controlled mixture of iron-ore, coke and limestone. The chemical properties of GG are shown in table 4.

| Principal Oxides (%) | CaO | SiO$_2$ | Al$_2$O$_3$ | MgO | FeO$_2$ |
|----------------------|-----|---------|-------------|-----|---------|
| Regen Company*       | 40  | 35      | 12          | 10  | 0.2     |
| Portland cement      | 65  | 20      | 5           | 5   | 1       |

* Regen Ground Granulated Blast furnace Slag (GGBS) is a cement substitute, manufactured from a by-product of the iron-making industry. The use of Regen in concrete reduces embodied CO$_2$ emissions by over 900kg per tonne of cement, and also increases its durability. http://www.heidlercement.co.uk/en/hansol/products/cements/ggbfs_and_related_products/regen_ggbfs.htm

4. Test Procedures

The samples were heated in the oven for approximately 1 hour at (160°C±5°C) for the unmodified binders and (180°C±5°C) for the modified binders in order to be fluid enough to easily pour; after heating, the samples were poured into prepared silicon moulds.
A Kinexus Pro’ rheometer from the Malvern Company, which was used to determine their viscoelastic properties. The rSpace program ver.1.7 developed by the Malvern Company was used to calculate the complex shear modulus ($G^*$), storage modulus ($G'$), loss modulus ($G''$), and phase angle ($\delta$) or loss tangent ($\tan(\delta) = \frac{G''}{G'}$), applied frequency, test temp., and dynamic shear viscosity. For the tests presented within this work, the parameters were set as outlined in table 5.

Table 5. Parameters of the test.

| Test parameter       | Test value          |
|----------------------|---------------------|
| Total time of test   | 1 hr                |
| Soak period at 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        |

Two geometries are used with the DSR according to (BS EN 14770:2012):
1. Eight mm diameter plate with two mm gap was at low temperatures (5 to 25 °C) and
2. Twenty-five mm diameter plate with a 1 mm gap was at intermediate to high temperatures (35 to 60 °C).

Details of this test are described as follows:

1. The dynamic result (complex shear modulus, phase angle and dynamic shear viscosity) were collected over a range of temperatures and frequencies (e.g. 5-60 °C with temperature intervals of 10 °C and 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 produced 126 data sets for each trial.
2. The dynamic viscosity of the asphalt cement was using a Brookfield DV-III+ programmable viscometer. SC4-28 spindle type was used. The measurements were made from test temperatures ranging between 100 °C to 200 °C for all asphalt cement. At each asphalt specimen, 10 readings created at 15-minute period were averaged and used in the analysis.

5. Specific gravity of local asphalt cement

Five local asphalt cement specimens are prepared carefully are used in to predict the specific gravity of local asphalt cement in the middle and southern regions of Iraq. The experimental results for asphalt cement are shown in table (6).
Table 6. Average Specific Gravity of Local Asphalt cement.

| Asphalt Cement Type | Nasiriyah | Durah       |
|---------------------|-----------|-------------|
| Pure                | 1.0553118 | 1.046284133 |
| 3% SBS 1101         | 1.1012241 | 0.6956612   |
| 6% SBS 1101         | 0.97262976| 0.919139124 |
| 9% SBS 1101         | 0.90491238| 0.879439557 |
| 3% SBS 1184         | 0.9847569 | 0.837114846 |
| 6% SBS 1184         | 1.04274282| 0.816010814 |
| 9% SBS 1184         | 1.07992424| 0.884237822 |

The following can be concluded from the experimental results shown in table (6):
1. The average value of specific gravity of asphalt cement production from Nasiriyah refinery is greater than that of asphalt cement production from Durah refinery.
2. The average value of specific gravity of asphalt cement produced from Nasiriyah refinery is reduced when adding SBS 1101 and vice versa if adding SBS 1184.
3. The average value of specific gravity of asphalt cement produced from Durah refinery in general reduced with increased percent of additives.

6. Determination of Linear Viscoelastic Region Limit.
Asphalt pavements have traditionally been designed using elastic theories of pavement behaviour as this is the simplest and easiest form of analysis. However, it is desirable to use viscoelastic methods of analysis in the design of asphalt pavements.

The linear viscoelastic (LVE) limit was defined in accordance with the SHRP study as the point where the complex modulus has decreased to 95 percent of its initial value.

There are many reasons why the linear viscoelastic regions of asphalt binder and asphalt mixtures need to be defined (Al-haddad (2015));
1) Simplicity in design.
2) Find correlations between asphalt binder and asphalt mixture rheology (study both asphalt binders and asphalt mixtures in the same domain). The viscoelastic parameters (complex modulus, phase angle, storage modulus and loss modulus) are all defined under linear viscoelastic (LVE) conditions.
3) Within the LVE region, the relationship between stress and strain is influenced only by temperature and loading time (frequency) and not by the magnitude of the stress or strain.
4) Methods of characterisation and analysis, such as the generation of master curves by means of the time-temperature superposition principle (TTSP), rely on the linearity of the rheological viscoelastic data.

The level of complex shear stress or strain can be used for the experimental and were selected from practical results of amplitude sweep stress and strain used in establishing the linear viscoelastic regions of the asphalt binders. The LVER limit was defined as the point at which the complex shear modulus reduced to 95% of the initial complex shear modulus.

A single shear stress and strain limit for asphalt films were selected, respectively within the LVER region. Tests were conducted at 25 °C and three replicates were performed for each condition. Figure 2 shows an example of sweep strain and stress for Nasiriyah asphalt binder. It should be mentioned that sweep stress and strain were just conducted on pure asphalt binders, and then the selected strain and stress values were implanted on all bitumen samples (pure and modified) so that comparison could be made with the control binder.
Al-haddad (2015); predicted the LVER limitations for local asphalt cement as shown in table 7, which summarizes the selected controlled strain and stress values for both binder grades.

![Sweep strain and stress for asphalt cement production from Nasiriyah refinery.](image)

**Figure 2.** Sweep strain and stress for asphalt cement production from Nasiriyah refinery.

| Asphalt Cement Source | Stress Level, Pa | Strain, % |
|------------------------|------------------|-----------|
| Nasiriyah refinery     | 120000-150000    | 2.0       |
| Durah refinery         | 100000-120000    | 2.5       |

**Table (7): Stress and strain levels used in fatigue tests**

7. **Chemical Analysis of Asphalt Cement**

The chemical components of Iraqi asphalt cement can be defined by using C-H-N-S microanalysis are processed at the Department of Chemistry; University of Liverpool. The practical results of chemical analysis are tabulated in table (8);
Table 8. Average value of C-H-N-S Microanalysis for Local asphalt cement

| Chemical Component | Nasiriyah Refinery | Durah Refinery |
|--------------------|--------------------|----------------|
| Symbol             | Pure               | SBS 1101       | SBS 1184       | Pure               | SBS 1101       | SBS 1184       |
|                    | average 3% 6% 9%   | 3% 6% 9%       | 3% 6% 9%       | 3% 6% 9%           | 3% 6% 9%       | 3% 6% 9%       |
| C                  | Carbon             | 83.465         | 83.93          | 83.28             | 84.04           | 83.07          | 83.09          | 83.27          |
| H                  | Hydrogen,          | 9.87           | 9.92           | 9.89              | 9.87            | 9.88           | 9.87           | 9.9            |
| N                  | Nitrogen           | 0.36           | 0.35           | 0.37              | 0.34            | 0.37           | 0.35           | 0.37           |

It can be seen from the analytical results obtained from C-H-N-S microanalysis as shown in table 8.

1. The average percent of carbon element for Nasiriyah asphalt cement is 83.47%, which is greater than that of Durah asphalt cement 82.27%; when adding SBS 1101 to asphalt cement increased the percentage of carbon vice versa when adding SBS 1184 reduced the percentage of carbon.
2. The average percentage of Hydrogen for Nasiriyah asphalt cement is equal to 9.87%, which is greater than Durah asphalt by cement 9.71%. In general, increased when adding SBS in both types.
3. The average percentage of Nitrogen for both types of asphalt equal to 0.36% and reduced when adding SBS additives.

8. Measured Dynamic Shear Viscosity

The dynamic shear viscosity (absolute viscosity) can be defined as the ability of materials to resist moving or internal friction (internal resistance). The dynamic shear viscosity of local asphalt cement was calculated in two cases based on the test temperature;

a) At high temperature (greater than 90 °C)

The parameter called dynamic shear viscosity at high temperature ranging over 90 °C can be calculated using Brookfield viscometer in the Highway and Structural Lab.; University of Liverpool; Faculty of Engineering; Civil and Environmental Dept. The experimental results are shown in figures (3 and 4);
Figure 3. Enhanced Ratio with different test Temperatures of Nasiriyah asphalt cement.

Figure 4. Enhanced Ratio with different modifier of Nasiriyah asphalt cement.

The experimental results for Durah asphalt cement are shown in figures (5 and 6);

Figure 5. Enhanced Ratio with different test Temperatures of Durah asphalt cement.
Figure 6. Enhanced Ratio with different modifier of Durah asphalt cement.

It can be seen;
1) The dynamic shear viscosity reduced with increasing test temperature.
2) In order to study the effect of adding modifier to improved viscosity of asphalt cement suggestion parameter called enhanced ratio it can be defined as \((\text{Modified value of viscosity} - \text{original value of pure asphalt}) / \text{original value of pure asphalt}\).
   - The enhanced ratio decreased with increased test temperature.
   - The higher value of enhancement obtained when adding polymer modifier, especially SBS 1184 with percentage equal to 9% with different test temperature.

b) At moderate and low Temperatures (less than 90 °C)
The dynamic shear viscosity at low and moderate temperature ranging lower than 90 °C can be measured using dynamic shear rheometer DSR in the Highway and Structural Lab.; University of Liverpool; Faculty of Engineering; Civil and Environmental Dept. The results obtained from DSR device are shown in the figures (7 and 8).
Figure 7. Dynamic shear viscosity of different test Temperatures of Durah asphalt cement

Figure 8. Enhanced Ratio with different test Temperatures of Durah asphalt cement

Figure 9. Enhanced Ratio with different test temperatures of Nasiriyah asphalt cement

9. Conclusion
1. From the analysis of the experimental results, the following conclusions can be drawn: The average value of specific gravity of asphalt cement produced from Nasiriyah refinery is greater than that of asphalt cement produced from Durah refinery.
2. The average value of specific gravity of asphalt cement produced from Nasiriyah refinery is reduced when adding SBS 1101 and increased when adding SBS 1184.
3. The average value of specific gravity of asphalt cement produced from Durah refinery in general is reduced with increased percentage of additives.
4. The stress level required for the experimental test for asphalt production from Nasiriyah refinery ranges between 120-150 kPa within the LVER / controlled stress mode.
5. The strain required for the experimental test for asphalt production from Nasiriyah refinery is equal to 2% within the LVER / controlled strain mode.
6. The stress level required for the experimental test for asphalt produced from Durah refinery ranges between 100-120 kPa within the LVER / controlled stress mode.
7. The strain required for the experimental test for asphalt production from Durah refinery is equal to 2.5% within the LVER / controlled strain mode.
8. The average percent of carbon element for Nasiriyah asphalt cement is 83.47% is greater than that of Durah asphalt cement 82.27%; when adding SBS 1101 increased percentage of carbon vice versa when adding SBS 1184 reduced the percentage of carbon.
9. The average percentage of Hydrogen for Nasiriyah asphalt cement is equal to 9.87% and Durah asphalt cement 9.71%. In general, increased when adding SBS in both types.
10. The average percentage of Nitrogen for both types of asphalt equal to 0.36% and gets reduced when adding SBS additives.
11. The dynamic shear viscosity is reduced with increasing test temperature.
12. In order to study the effect of adding modifier to improve viscosity of asphalt cement a new parameter is proposed which is called enhanced ratio ((Modified value of viscosity – original value of pure asphalt)/ original value of pure asphalt).
   a. The enhanced ratio decreased with increased test temperature; this behavior like viscosity that mean behavior of asphalt remain with or without modifier.
   b. The higher value of enhancement is obtained when adding polymer modifier, especially SBS 1184 with percentage equal to 9% with different test temperature.
13. The increased G’ means increased phase angle (increased AASHTO rutting parameter).
14. Adding SBS modifier caused an increase in viscous part of complex shear modulus and then increased dissipated energy for Durah asphalt cement, the case is reversed when using Nasiriyah Asphalt cement.
15. Adding SBS modifier to Nasiriyah Asphalt cement reduced the viscous part, decreased phase angle and reduced AASHTO parameter that means enhanced rutting resistance.

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