

## Rheograms for asphalt from single viscosity measurement\*)

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*Abstract:* Asphalt materials are used in a variety of applications such as road paving, waterproofing, roofing membranes, adhesive binders, rust proofing and water resistant coatings. There are available in a number of grades distinguished in terms of their softening point and flow resistance. The selection of the proper grade of asphalt for a particular application is governed by the desired flow behaviour. A knowledge of the complete flow curve depicting the variation of melt viscosity with shear rate at the relevant temperatures is necessary not only for proper grade selection, but also for specifying processing conditions for aggregate mixing and spraying. The rheological data are also useful in assessing end use performance. The scientific techniques for generating the rheological data involve the use of expensive, sophisticated instruments. Generation of the necessary flow data using these instruments is beyond the financial and technical means of most processors of asphalt materials. The engineering techniques involving the use of inexpensive vacuum viscometers are relatively easy, but provide a single point viscosity measurement at low shear rate. In the present work, a method is proposed for unifying the viscosity versus shear rate data at various temperatures for a number of asphalt grades. A master curve has been generated that is independent of the grade of asphalt and the temperature of viscosity measurement. The master curve can be used to generate rheograms at desired temperatures for the asphalt grade of interest, knowing its zero-shear viscosity at that temperature.

*Key words:* Rheogram, master curve, single viscosity measurement, softening point, asphalt

### 1. Introduction

Asphalt is a dark brown to black cementitious material, solid or semi-solid in consistency in which the predominant constituents are bitumens which occur in nature as such or are obtained as residue in refining petroleum. Bitumens contain a wide spectrum of complex hydrocarbons whose detailed structures are not truly known. However, the common practice is to divide the constituents based on the solubility in certain solvents into two fractions, namely, the soluble "maltenes" and the insoluble "asphaltenes". The maltenes exist as a highly viscous dark brown oil of the Newtonian type and constitute the low molecular weight portion of the asphalt. The asphaltenes, on the other hand, are the high molecular weight constituents of the asphalts and exist as

solid brownish black substances. The asphaltenes due to their highly structured nature are mainly responsible for the varied rheological behaviour of asphalts available from different base stocks and sources. A carbon-hydrogen analysis is known to have shown that the composition of asphaltenes is invariant of the source of asphalt [1]. The differences in flow behaviour of asphalts can therefore be attributed to the variations in the molecular weights of asphaltenes.

Rheological properties are indicative of the inherent structure of the bituminous materials and often form the basis of selection of the type of asphalt for a particular application. Asphalts are mainly used as binding material and in protective coatings. Their applications include paving of roads, crack sealing, roofing, waterproofing, rustproofing, pipe coating, automobile undercoating, coating for fishnets, low-cost matrices for glass reinforcement and other filler systems, etc. Such a broad array of applications are possible

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due to the great number of modifications that can be done in order to alter or improve the properties of asphalt. Some of these modifications are:

- Sulfurization or oxidation of asphalt.
- Blending of the asphalt with a variety of synthetic polymeric materials, rubbers, aminosilanes, amines, amides, siloxanes, antioxidants, stabilizers, reinforcing agents, etc.
- Chemically reacting the asphalt with appropriate reactants.

A knowledge of the rheological behaviour of asphalts is necessary for selection of the proper grade for a particular application, for processing such as compounding with gravel, spray coating etc., and also for satisfactory end-use performance. For example, an asphalt coating for holding granules in place in roofing shingles is required to have a very high viscosity, estimated to be in the order of  $7 \times 10^8$  poises, so that granule movement during the life of the roofing becomes infinitesimal [1]. In highway applications, except for parking areas, the rate of loading is rapid and aggregate binder systems with less static properties can be utilized. However, resistance to flow at temperatures attained on the road surface on a hot summer day is critical. Therefore, selection of proper grade with the required softening point and viscosity at the end-use temperature is important.

The rheological properties of asphalt are generally specified through one or more of the following types of characterisation:

#### a) Penetration index

The penetration index indicates the distance, in millimetres, a needle will penetrate into the asphalt under known conditions of temperature, loading and time.

#### b) Softening point

The softening point is the temperature at which a hard asphalt becomes a fluid with a consistency such that a steel ball of specified dimensions and weight placed in a brass ring causes a prespecified deformation.

#### c) Ductility

Ductility is the length of pull, in centimetres, at which a briquette of asphalt molded under standard test conditions and dimensions breaks when pulled or extended at a constant specified speed.

#### d) Stiffness modulus versus loading time variation

Stiffness modulus is the resistance to deformation when a constant load is applied and the deformation is measured as a function of time. The relationship between viscosity, stiffness modulus and loading time for asphalt has been given in Van der Poel [2] as:

$$3\eta = \frac{\sigma}{\epsilon}t, \quad (1)$$

where  $\eta$  is the viscosity,  $\sigma$  the tensile stress,  $\epsilon$  the strain,  $\sigma/\epsilon$  the stiffness modulus, and  $t$  the loading time.

The stiffness modulus versus loading time variation is often used for rheological characterization as it depicts the

elastic behaviour of the asphalt at small deformations and short loading times as well as the viscous behaviour at large deformations and longer loading times.

Table 1. Guide for use in determining application temperatures as given in the asphalt handbook [49] in the absence of suitable temperature-viscosity data

Type and grade of asphalt	Suggested temperature of use	
	For mixing (°F)	For spraying (a) (°F)
1	2	3
Asphalt cements		(b)
40-50 pen	300-350	285-350
60-70 pen	275-325	285-350
85-100 pen	275-325	285-350
120-150 pen	275-325	285-350
200-300 pen	200-275	260-325
RC liquid asphalts		
RC-0	50-120	65-135
RC-1	80-125	110-180
RC-2	80-150	140-210
RC-3	125-175	170-240
RC-4	150-200	180-255
RC-5	175-225	215-285
MC liquid asphalts		
MC-0	50-120	70-140
MC-1	80-150	110-185
MC-2	100-200	140-215
MC-3	150-200	175-250
MC-4	175-225	190-265
MC-5	200-250	220-290
SC liquid asphalts		
SC-0	50-120	70-140
SC-1	80-200	110-185
SC-2	150-200	140-215
SC-3	175-250	175-250
SC-4	175-250	190-265
SC-5	200-275	220-290
Asphalt emulsions (c)		
RS-1	(d)	75-130
RS-2	(d)	110-160
MS-2	100-160	100-160
SS-1	75-130	75-130
SS-14	75-130	75-130

(a) Low temperature is based on a viscosity of 100 seconds, Saybolt Furol, for liquid asphalt meeting the lower limit of this specification and the high temperature is based on 25-second viscosity.

(b) Seldom used for spraying.

(c) Since the working temperature range for emulsions is comparatively low and is only based on that temperature necessary to provide a viscosity at which the emulsion can be dispersed through spray nozzles, the same temperature is specified for use in spraying and/or mixing

(d) Not used for mixing.

### e) Viscosity versus shear rate variation

Viscosity is the resistance to deformation on application of a stress. It is measured as the ratio of the shear stress to the shear rate. In cases where measurements based on direct shear are difficult to express in terms of elongation, the rheological behaviour is described by the viscosity versus shear rate variation in the range of temperatures used during application. The suggested temperatures of use for mixing and spraying of various types and grades of asphalt are given in table 1 reproduced from the Asphalt Handbook [3].

Since asphalt is available in a wide variety of standard types and grades, there have been innumerable rheological studies (4–50) using one or more of the above methods of characterization. We shall, however, focus our attention on those works [7, 17–20, 31, 41, 47] which have generated viscosity versus shear rate data in order to elucidate the rheological behaviour of asphalt. Considering the number of variables such as asphalt type and grade, viscosity, shear rate and application temperature involved in these studies, it is obvious why there is an abundance of data and a series of curves on viscosity versus shear rate. However, it is desirable to develop a method for unifying the extensive rheological information relating to viscosity versus shear rate variation of asphalt and provide an easy means to characterize the rheological behaviour irrespective of its varied grade, type and origin.

The purpose of the present paper is to generate a master curve for viscosity versus shear rate from available data for obtaining the flow curves of various grades of asphalt merely from a single point viscosity measurement. This technique would eliminate the need for cumbersome data generation using cone and plate viscometers.

## 2. Background

Viscosity determinations of asphalt are generally done by one of the following two methods:

a) When the variation of viscosity with shear rate at a particular temperature is desired, a modified cone and plate viscometer as used by Sisko [20], Bestougeff et al. [40] or Khong et al. [47] is generally employed. Sophisticated instruments like the Weissenberg rheogoniometer or the Rheometrics Mechanical Spectrometer could also be used. All these equipment, though capable of giving accurate data, are extremely expensive and require trained operators. Thus they are beyond the financial means and the technical capabilities of all small scale industries involved in asphalt-based products and applications.

b) For a comparative study of the viscosity of different grades of asphalt, viscometers like the Asphalt Institute Vacuum viscometer, the modified Koppers Vacuum viscometer or the Saybolt viscometer are generally used. These are relatively inexpensive and reasonably accurate. However, this method provides a single value measurement of viscosity based on the time taken to draw a fixed volume of asphalt through a capillary tube under closely controlled conditions of vacuum and temperature as per ASTM method D2171-66.

In the present work, a method is proposed to generate the rheograms of asphalts depicting the variation of viscosity with shear rate at various temperatures from a knowledge of the single viscosity value as determined by the ASTM method D2171-66.

## 3. Data collection

The data used in the present analysis were taken entirely from Khong et al. [47] as their study covered a wide spectrum of variables like eight commercial grades of asphalts at seven different temperatures and a shear rate range from 0 to 10 s<sup>-1</sup>. The characteristics of the asphalts studied are given in table 2 taken from [47]. The rheological data on analysis yielded a master curve independent of temperature and grade of asphalt. A summary of the systems analysed in the present study is given in table 3.

## 4. Data analysis

Vinogradov and Malkin [51] have proposed a universal viscosity function such that the viscosity data for a number of polymers such as polyethylene, polypropylene, polystyrene and polyisobutylene all fell within a bandwidth of the master curve of  $\log \eta/\eta_0$  vs.  $\log \eta_0 \dot{\gamma}$ . Based on a physical approach, Shenoy et al. [52–55] have shown that a master curve can be obtained by plotting  $\eta \cdot \text{MFI}$  vs.  $\dot{\gamma}/\text{MFI}$  on a log-log scale for a variety of polymers including olefins, styrenics, cellulose, engineering thermoplastics and filled systems. The MFI (Melt Flow Index) is a single point viscosity parameter commonly used for characterizing thermoplastic materials (ASTM 1238-73). MFI is found to be inversely proportional to  $\eta_0$  [52]. Thus the basis for unification of the viscosity data by using  $\eta_0$  and by using MFI is the same. In the case of the polymeric materials studied by Shenoy et al. [52–55], master curves based on MFI are preferred to those based on  $\eta_0$ , because of the ease and accuracy of MFI determination as compared to the determination of the zero shear viscosity. Based on the data given by Khong et al. [45] reproduced in table 3, it is apparent that the values of the zero-shear viscosity of the various asphalt grades are comparable when measured using a cone and plate viscometer and the vacuum viscometer. The simple method of vacuum viscometer can be used to obtain the value of  $\eta_0$  which can then be substituted in a master curve of  $\log \eta/\eta_0$  vs.  $\log \eta_0 \dot{\gamma}$  to generate the rheograms of the particular asphalt.

Table 2. Characteristics of the asphalts studied by Khong, Malhotra and Blanchard [47]

No.	Source and grade	Penetration <sup>1)</sup>			Viscosity <sup>2)</sup> at 60°C (poises)	Softening point <sup>3)</sup> (°C)	Molecular weights <sup>4)</sup>	
		at 4°C	at 25°C	at 60°C			$M_w$	$M_n$
<i>A</i>								
1	85-100	20	90	260	1480	47	1230	850
2	150-200	29	148	300+	700	40	1180	820
<i>B</i>								
3	85-100	15	90	275+	1570	47	1300	880
4	150-200	28	170	275+	620	40	1250	870
5	300-400	48	293	275+	290	33	1160	840
<i>C</i>								
6	85-100	19	83	250	1980	48	1210	880
7	150-200	34	164	300+	510	38	1130	840
<i>D</i>								
8	85-100	-	-	-	-	-	1160	850

1) ASTM D5-71

2) ASTM D2171-66

3) ASTM D2398-68

4) Waters Associates Model A-200 GPC

Table 3. Data summary of the asphalts investigated (as given in [47])

No.	Source and grade	Data temps (°C)	No. of data points at each temperature	Shear rate range (s <sup>-1</sup> )
<i>A</i>				
1	85-100	20, 30, 37.8, 50, 60, 70	11	0-10
2	150-200	20, 30, 37.8, 50, 60, 70	11	0-10
<i>B</i>				
3	85-100	20, 30, 37.8, 50, 60, 70	11	0-10
4	150-200	20, 30, 37.8, 50, 60, 70	11	0-10
5	300-400	20, 30, 37.8, 50, 60, 70	11	0-10
<i>C</i>				
6	85-100	20, 30, 37.8, 50, 60, 70	11	0-10
7	150-200	20, 30, 37.8, 50, 60, 70	11	0-10
<i>D</i>				
8	85-100	20, 30, 37.8, 50, 60, 70	11	0-10

*A* - Material produced by British Petroleum*B* - Material produced by Petrofina*C* - Material produced by Gulf Oil*D* - Material produced by ESSO

## 5. Results and discussion

Figure 1 shows the plot of  $\log \eta/\eta_0$  vs.  $\log \eta_0 \dot{\gamma}$  for eight grades of asphalt at different temperatures of 20°C, 30°C, 37.8°C, and 50°C. It can be seen that all the curves superimpose in an excellent manner thereby giving a master curve covering a wide range of shear rates, temperatures and asphalt grades. This curve is independent of the source and grade of the asphalt and the temperature of the viscosity measure-

ment. The viscosity versus shear rate curve for a particular grade of asphalt at a particular temperature can be easily obtained on knowing the value of the zero-shear viscosity  $\eta_0$  for that particular grade at that temperature. An excellent estimate of the zero-shear viscosity can be got from the single value measurement from the Asphalt Institute Vacuum Viscometer of a Modified Koppers Vacuum Viscometer as illustrated in table 4.

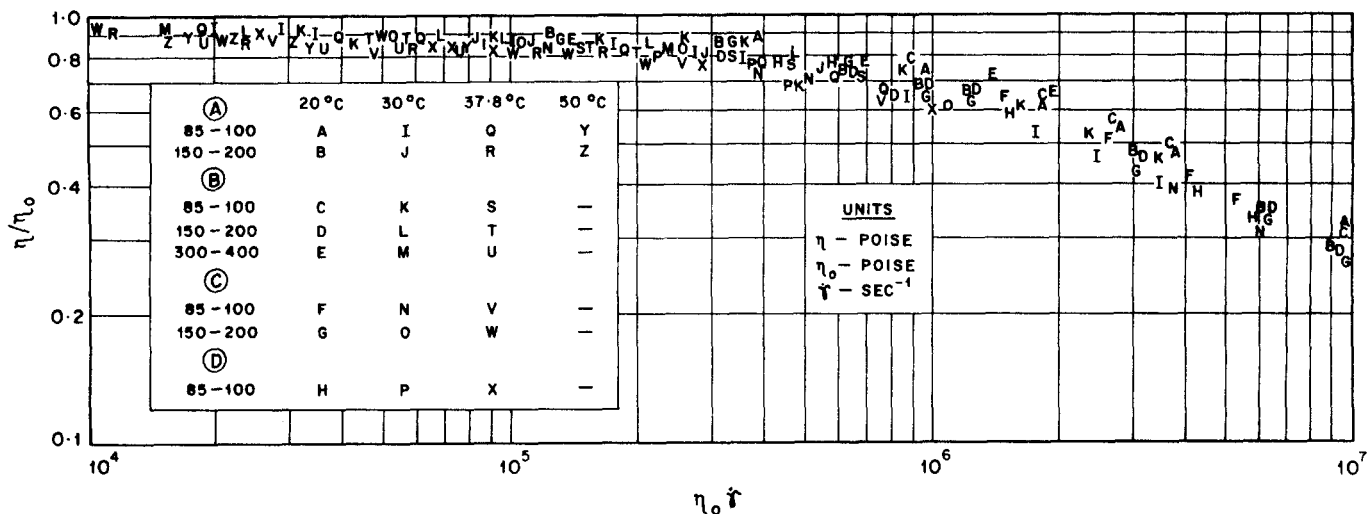


Fig. 1. Master curve of  $\eta/\eta_0$  versus  $\eta_0 \dot{\gamma}$  for asphalt which is independent of temperature of viscosity measurement as well as the source and grade of the asphalt

Table 4. Comparison of the zero shear rate viscosities (in poises) measured with the cone-and-plate viscometer and the capillary viscometers at different temperatures [45]

Origin and grade	30°C Viscosity $\times 10^{-5}$			37.8°C Viscosity $\times 10^{-4}$			50°C Viscosity $\times 10^{-3}$			60°C Viscosity $\times 10^{-3}$			70°C Viscosity $\times 10^{-2}$		
	1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
<b>A</b>															
85-100	3.45	3.69	3.66	7.80	7.59	7.74	7.40	7.05	6.90	1.60	1.47	-	4.67	4.51	-
150-200	1.10	1.18	-	2.25	2.56	2.52	3.00	3.05	3.22	0.75	0.72	-	2.40	2.36	-
<b>B</b>															
85-100	3.30	3.45	-	6.10	6.31	6.21	7.00	7.00	6.92	1.45	1.41	-	4.36	4.28	-
150-200	0.90	0.97	-	1.89	1.71	1.86	2.55	2.60	2.53	0.65	0.64	-	2.20	2.13	-
300-400	0.30	0.33	0.30	0.71	0.76	0.70	1.08	1.06	1.04	0.29	0.29	-	1.14	1.08	-
<b>C</b>															
85-100	5.00	4.44	4.61	9.80	8.70	8.45	9.70	9.87	9.00	2.05	1.99	-	5.60	5.51	-
150-200	1.06	1.18	-	2.00	1.78	1.86	2.51	2.15	2.21	0.60	0.57	-	1.90	1.84	-
<b>D</b>															
85-100	4.60	4.86	4.49	9.10	9.12	9.12	8.80	8.75	8.92	1.90	1.78	-	5.22	5.14	-

1 - Cone-and-plate viscometer; 2 - The Asphalt Institute vacuum viscometer; 3 - Modified Koppers vacuum viscometer

The zero-shear viscosity value should be obtained at a temperature at which the viscosity versus shear rate curve is desired. In case where it is not feasible to accurately measure the zero-shear viscosity at a particular temperature, the W-L-F equation can be used to determine the  $\eta_0$  at the desired temperature knowing  $\eta_0$  at some other reference temperature. Khong et al. [46] have shown that the constants  $C_1$  and  $C_2$  in the W-L-F equation are dependent on the grade and source of asphalt and the original values of the constants as proposed by Williams, Landel and Ferry [56]

should not be used for asphalt. The suggested correct values of  $C_1$  and  $C_2$  and  $T_g$  for different grades according to Khong et al. [46] are given in table 5. These can be then appropriately used in the following equation to determine the zero-shear viscosity  $\eta_0$  at a desired temperature  $T_2$  from a known value of  $\eta_0$  at a reference temperature  $T_1$ :

$$\log \frac{\eta_0(T_2)}{\eta_0(T_1)} = - \frac{C_1(T_2 - T_g)}{C_2 + (T_2 - T_g)} + \frac{C_1(T_1 - T_g)}{C_2 + (T_1 - T_g)}, \tag{2}$$

Table 5. Optimized values of constants  $C_1$ ,  $C_2$  and the glass transition temperature  $T_g$  of various grades of asphalt [46]

No.	Source and grade	$C_1$	$C_2$	$T_g$ ( $^{\circ}\text{C}$ )
<i>A</i>				
1	85–100	27.6	52.7	–35.0
2	150–200	38.0	22.3	–43.0
<i>B</i>				
3	85–100	27.6	52.7	–44.7
4	150–200	38.0	22.3	–45.2
5	300–400	36.1	22.5	–45.0
<i>C</i>				
6	85–100	27.6	52.7	–36.0
7	150–200	38.0	22.3	–40.0
<i>D</i>				
8	85–100	27.6	52.7	–37.7

where  $\eta_0(T_1)$  and  $\eta_0(T_2)$  are zero-shear viscosities at known temperature  $T_1$  and at temperature of interest  $T_2$ , respectively;  $C_1$ ,  $C_2$  are constants and  $T_g$  is the glass transition temperature of asphalt as given in table 5.

The steps to be followed in order to obtain the flow curves at the required temperature from the master curves are summarized below:

- i) The zero shear viscosity of the asphalt sample is to be determined through an Asphalt Institute Vacuum viscometer, i.e. a Modified Koppers Vacuum viscometer as per ASTM 2171-66.
- ii) If the temperature of measurement of the Vacuum viscometer does not correspond to the temperature of interest, eq. (2) should be used to determine the zero-shear viscosity at the temperature of interest knowing the appropriate values of  $C_1$ ,  $C_2$  and  $T_g$  from table 5 for the particular grade of the asphalt.
- iii) The rheogram can then be easily obtained by substituting the correct values of  $\eta_0$  in the master curve. For convenience, the values of  $\eta_0\dot{\gamma}$  and  $\eta/\eta_0$  have been tabulated in table 6 covering a wide range of  $\eta_0\dot{\gamma}$  from 0 to  $3 \times 10^7$ .

## 6. Conclusion

The unifying approach for coalescing rheograms of various grades of asphalts at different temperatures, in terms of a modified viscosity function  $\eta/\eta_0$  and a modified shear rate function  $\eta_0\dot{\gamma}$  has yielded a master curve. Viscosity versus shear rate flow curves at relevant required temperatures can be generated for asphalt from the master curve and the knowledge of

the single viscosity measurement taken on a simple vacuum viscometer. For convenience, tabulated read out values of  $\eta_0\dot{\gamma}$  and  $\eta/\eta_0$  are given in table 6 for a wide range of  $\eta_0\dot{\gamma}$  from 0 to  $3 \times 10^7$ . The rheograms generated by using the technique proposed, would give a good estimate of the viscosity of asphalt for most applications.

Table 6. Optimum computed values of  $\eta_0\dot{\gamma}$  and  $\eta/\eta_0$  for the master curve

$\eta_0\dot{\gamma}$	$\eta/\eta_0$	$\eta_0\dot{\gamma}$	$\eta/\eta_0$
0	1.000	$2 \times 10^5$	0.810
1	0.999	$3 \times 10^5$	0.800
5	0.996	$4 \times 10^5$	0.790
10	0.994	$5 \times 10^5$	0.770
$5 \times 10$	0.990	$6 \times 10^5$	0.750
$10^2$	0.988	$7 \times 10^5$	0.730
$5 \times 10^2$	0.982	$8 \times 10^5$	0.710
$10^3$	0.971	$9 \times 10^5$	0.700
$5 \times 10^3$	0.962	$10^6$	0.680
$10^4$	0.945	$2 \times 10^6$	0.550
$2 \times 10^4$	0.920	$3 \times 10^6$	0.460
$3 \times 10^4$	0.900	$4 \times 10^6$	0.400
$4 \times 10^4$	0.890	$5 \times 10^6$	0.360
$5 \times 10^4$	0.870	$6 \times 10^6$	0.340
$6 \times 10^4$	0.860	$8 \times 10^6$	0.300
$7 \times 10^4$	0.850	$9 \times 10^6$	0.280
$8 \times 10^4$	0.840	$10^7$	0.270
$9 \times 10^4$	0.825	$2 \times 10^7$	0.200
$10^5$	0.820	$3 \times 10^7$	0.140

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