EXPERIMENTAL VERIFICATION OF EVOLUTION OF INTERNAL STRUCTURE OF ASPHALT AND MODIFIED ASPHALT

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ABSTRACT

The complex nature of asphalt throws open challenges in terms of material characterization, pavement design and analysis procedures. This study is concerned with experimentally verifying the reversible changes in the material and we focus only on the mechanical properties. Two types of asphalt, unmodified parent asphalt and polymer modified asphalt were tested in a Brookfield programmable viscometer. The material was tested from 60 to 90 °C in increments of 5 °C. Each sample was tested before and after short term aging. The experimental plan consisted of subjecting the material to three continuous cycles of shearing, allowing the material to rest for one hour at the testing temperature after these three cycles and subjecting the material to one more cycle of shearing after rest period. The observations in terms of shear stress vs. time for the specific forward and reverse sweep revealed several interesting facts. The polymer modified asphalt shear thickened for every successive cycle before rest period in the temperature range of 60-70 °C. The same material however, started shear thinning beyond 70 °C. The rest periods had a significant effect on polymer modified asphalts.

The change in shear stress for different shear rates varied for different asphalts (modified and unmodified, aged and unaged) used in the testing program. The experimental data reported here will not only help in clearly quantifying the changes in the behavior of the material during rest period but also the influence of modification and short term aging. It is also to be highlighted here that all the observations are in terms of raw experimental data without recourse to any model.

Keywords: polymer modified asphalt, aging, internal structure, steady shear

1. INTRODUCTION

The use of asphalt as a binder for pavement construction for highways and runways is well documented (Krishnan and Rajagopal, 2003). Increased tire pressures together with the ever increasing traffic volumes have posed challenges to pavement designers as well as asphalt manufacturers towards producing asphalt of highest quality. The initiatives of the SHRP in USA and RILEM in Europe have resulted in providing better understanding of asphalt. However, the complexity in unraveling a precise chemical composition for asphalt and pinpointing the influence of the various chemical compositions on its rheological properties is still a challenge. The main issues in understanding the true mechanical behavior of asphalt are related to its multi-constituent nature, the transitory nature of asphalt and the development of internal structure (Krishnan and Rajagopal, 2005). Added to the complexity is the increased use of polymer as an additive to asphalt to improve its mechanical and thermodynamic properties. Various field investigations have confirmed the loss of pavement deterioration, improved age resistant properties as well as better low temperature thermal and fatigue crack resistance etc., of pavement constructed with modified binders (Wardlaw and Shuler, 1992). However, the complex interactions polymers have with asphalt creates difficulties related to appropriate material characterization methodologies.

While the pavement construction technology is in a fairly advanced stage, the same cannot be said about the material characterization within the realm of pavement design and analysis. Very recently only, a shift towards mechanistic methods of pavement design took place and questions related to the relation between pavement distress and material properties are still not answered with reasonable degree of confidence. In all these, one of the main impediments is related to developing appropriate and rigorous constitutive models for asphalt and modified asphalt. Most of the current attempts including the investigation carried out as part of SHRP and RILEM have mainly assumed linear viscoelastic behavior for asphalt and modified asphalt. Asphalts and modified asphalts are also assumed to behave like a ‘thermorheologically simple’ materials and time-temperature superposition is often used to predict the response at various temperature and loading rates. The use of presupposed constitutive models for asphalts and modified asphalts results in measurements reported in the literature in terms of complex modulus, relaxation modulus and creep compliance and these are clearly linear viscoelastic parameters. Significant and interesting behavior of the material during routine testing is oftentimes lost because the investigators did not present the results in terms of torque-shear rate-time, and presented for instance data in terms of complex modulus and phase angle. It is also disadvantageous to a modeler when the data is not available in raw format so that the validity or otherwise of any model can be checked with the aid of the experimental data.

In this investigation, the focus is mainly on capturing the evolution of the internal structure of unaged/aged base and modified asphalt by means of steady shear experiments. One of the main issues related to modeling the response of asphalt is in capturing the evolution of internal structure as this will have a significant bearing on the choice of the framework adapted. The fact that the internal structure can evolve during deformation as well as during rest period complicates the modeling effort. However, even before a modeling attempt is initiated, it is necessary to quantify the actual changes which take place in the material through careful experiments and collect the data in terms of model independent parameters.

The development of internal structure during rest periods was studied as early as by Traxler and co-workers (1936, 1937). While measuring the viscosity of several different asphalts in falling coaxial cylinder viscometers, they noticed that the viscosities of asphalt kept in the viscometers for considerable time exhibited increase in their values. Traxler et al. ascribed this structure formation due to the two phase nature (asphaltene and petrolene) of asphalt in which a gradual isothermal sol–gel transformation occurs as asphalt is kept steady at a specific temperature. Similar results were reported by Brown et al. (1957) who chose to call this phenomenon as *steric hardening*. Brown et al. attributed the formation of internal structure to the asphaltenes fraction of asphalt. The change in the internal structure of asphalt when held for sufficient time near the glass transition temperature has been characterized as that due to *low temperature physical hardening* in the work of Bahia and Anderson (1992). Claudy et al. (1992) investigated the low temperature physical hardening of asphalt and concluded that molecular agglomerations of crystalline phases at low temperature could be one reason for this behavior. Continuing on these lines, Masson et al. (2002) ascribed a four stage internal structural development process for asphalt. Each of the fractions of asphalt influence in their own way, depending on their tendency for crystallization, in the formation of reversible internal structure (see Masson et al., 2002). Essentially one observes a change in the property (density, mechanical response characteristics, dielectric properties, etc.) of asphalt when maintained at a constant temperature for considerable time in the absence of any external forces and without any appreciable change in its chemical composition. Thermal and/or external forces acting on asphalt can revert the internal structure to the original condition in which it existed. On the other hand, the change in the internal structure of the material during deformation exhibited by a shear thinning or shear thickening phenomenon is fairly well known and has been documented extensively for most of the fluids. However, this cannot be said for asphalt and modified asphalt, both aged and unaged. For instance, while most of the polymers can be assumed to exhibit shear thinning behavior, it will be interesting to see the behavior of asphalt modified with polymer. Very few investigations have actually captured this phenomenon and noteworthy among them are by Polacco et al. (2006) and Wekumbura et al. (2007). Depending on the type and percentage of polymer used, the asphalt-polymer mix can exhibit asphalt-rich constitution with dispersion of polymer particles or polymer-rich constitution with dispersion of asphalt particles. The formation of a three-dimensional network (Adedeji et al., 1996) and its scission during deformation and 'healing' during rest periods affects significantly the mechanical properties of modified binders.

Considering the fact that characterization of the mechanical properties of asphalt and modified asphalt is an important step, an experimental program was undertaken to study one specific aspect of asphalt: the evolution of the internal structure during mechanical shearing and during rest periods between shearing. Two types of asphalt, a straight run asphalt of 80/100 penetration grade and a polymer modified asphalt were used in this investigation. The details are given in the following section. This current investigation is in an intermediate stage and some of the select results are reported. It is to be emphasized here that ‘evolution of internal structure’ during deformation and rest periods is characterized by means of the mechanical response of the material. Additional insight related to the molecular structure of the material during this evolution requires sophisticated analysis of the molecular structure of the material and clearly this is out of scope of this paper.

2. EXPERIMENTAL PROCEDURE

In this study, Brookfield DV-II+Pro programmable rotational viscometer was used. The details of the asphalt tested are given in Table 1.

Table 1 Routine Test Data of Base and Modified Asphalt

Asphalt	Penetration at 25 °C, 100 g, 5 s (1/10 mm)	Softening point (°C)	Viscosity at 135 °C (cP)	Source
Base 80/100	95	42	360 (20 RPM, SC4-21 spindle)	Hindustan Petroleum Corporation Limited, Visakhapatnam, India
Base 80/100+ 3.5% SBS	90	55	1230 (20 RPM, SC4-21 spindle)	HINCOL, Chennai, India

The asphalt samples were tested before and after short term aging. James Cox and Sons rolling thin film oven was used to subject the asphalt samples to short term aging. The short term aging was performed in accordance with the relevant ASTM specifications. The material was tested in the rotational viscometer from 60 to 90 °C in increments of 5 °C.

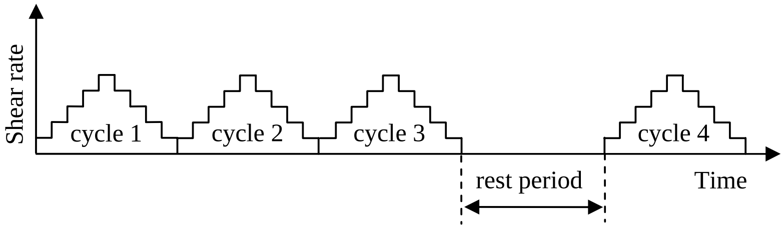

Figure 1 Shear History Diagram

The experimental protocol followed in the current investigation is depicted pictorially in Figure 1. The test protocol consists of subjecting the asphalt samples to three continuous shearing cycles, allowing the material to rest for one hour at the test temperature and subjecting the material further to another shearing cycle. Each shearing cycle consisted of a forward and reverse sweep. The forward and reverse sweeps in each cycle included five increments and decrements respectively. In each ramp of a forward and reverse sweep, the material was subjected to a constant shear rate for duration of 60 seconds. The 60 seconds shearing time was chosen after several laboratory trials such that the shear stress response attains equilibrium state under most of the tested conditions. The test matrix followed in the current investigation is shown in Table 2. The shear rates at a particular temperature for the aged polymer modified asphalt were arrived based on the maximum permissible torque limits. In all the tests performed on aged polymer modified asphalt, the maximum torque levels were close to 80 percent. The same shear rates arrived from the tests performed on aged polymer modified asphalt were applied on unaged polymer modified asphalt, aged unmodified asphalt and unaged unmodified asphalt to observe the effect of modification and aging on neat asphalt binder. Two samples were tested for each condition and the repeatability was found to be less than 5 percent. A sample plot of shear stress vs. time and shear rate vs. time for all the asphalt tested at 60 °C is shown in Figure 2. The shear rate (1/s) to RPM conversion for the spindle used is 0.34.

Table 2 Experimental Program

Temperature (°C)	**Shear Rate (RPM)**		**Shearing time (s)**
	Minimum	**Maximum**	
60	0.02	0.1	60
65	0.04	0.2	60
70	0.1	0.5	60
75	0.2	1	60
80	0.5	2.5	60
85	0.9	4.5	60
90	1.6	8	60

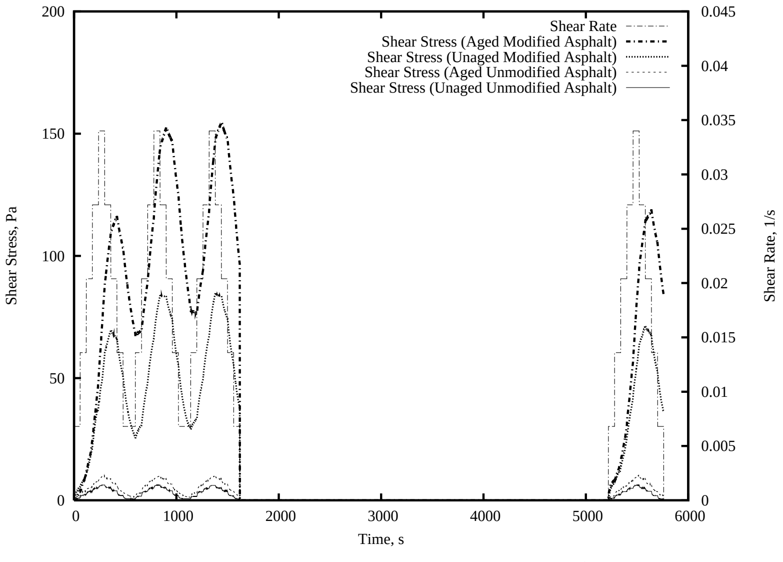

Figure 2 Shear Stress Responses at 60 °C

3. RESULTS AND DISCUSSIONS

Figure 3 shows the shear stress evolution of the aged polymer modified asphalt tested at 60 °C. It is interesting to see the change in response of the material for each cycle of shearing. It is also noteworthy to point out here that the material exhibits significant change in its mechanical response after one hour rest period. Clearly the material response changes not only during each cycle of shearing but even during rest period.

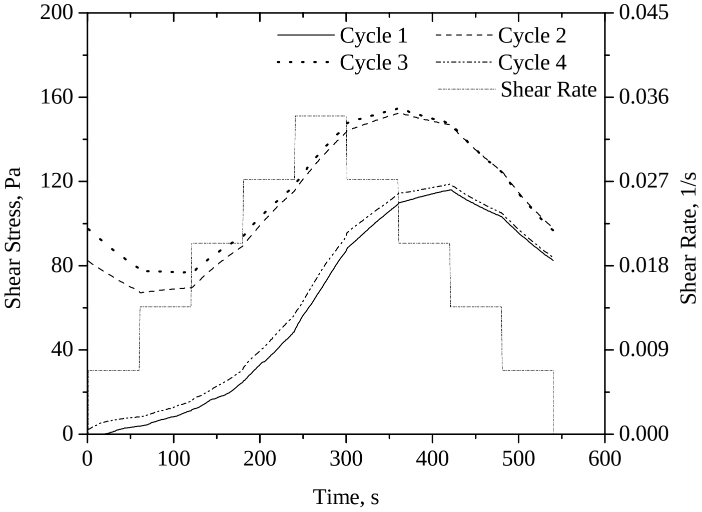

Figure 3 Shear Stress Response at 60 °C of an Aged Polymer Modified Asphalt

To understand how the change in the mechanical response takes place for every cycle of shearing for the aged polymer modified asphalt at 60 °C, the results are plotted for all the four cycles for the upward ramp in Figure 4. While for the same shear rate for every subsequent cycle, the shear stress-shear rate slope increases thus exhibiting shear thickening behavior, it is noteworthy to see that the one hour of rest period results in the material exhibiting response during fourth cycle similar to the response during the first cycle. Figure 5 shows a similar plot for aged unmodified asphalt at 60 °C. Here, one cannot see any change in the mechanical response of the unmodified binder. The role of 3.5% of polymer in drastically changing the behavior of the material can also be seen by comparing Figures 4 and 5. Figure 6 shows the shear stress – shear rate response of aged modified asphalt at 90 °C and clearly the transition in behavior as the temperature is increased can be seen by comparing Figures 4 and 6. Figures 7 and 8 shows the transition from shear thickening to shear thinning behavior as the temperature is increased from 65 to 75 °C. As expected, aged and unaged polymer modified asphalts show drastic change in mechanical properties when compared with regular aged and unaged asphalt. Figure 9 shows the trend at 90 °C.

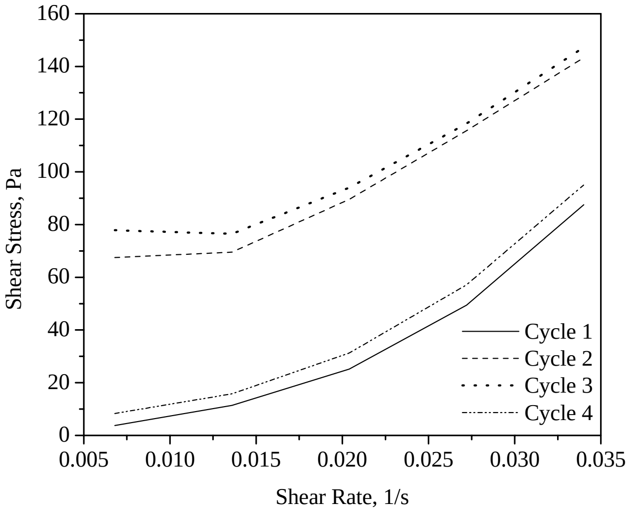

Figure 4 Shear Stress - Shear Rate Response at 60 ° C of an Aged Polymer Modified Asphalt (Upward Ramp)

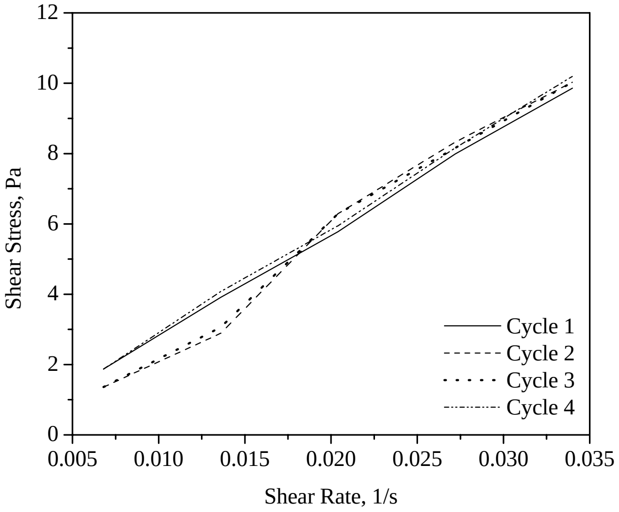

Figure 5 Shear Stress - Shear Rate Response at 60 ° C of Aged Unmodified Asphalt (Upward Ramp)

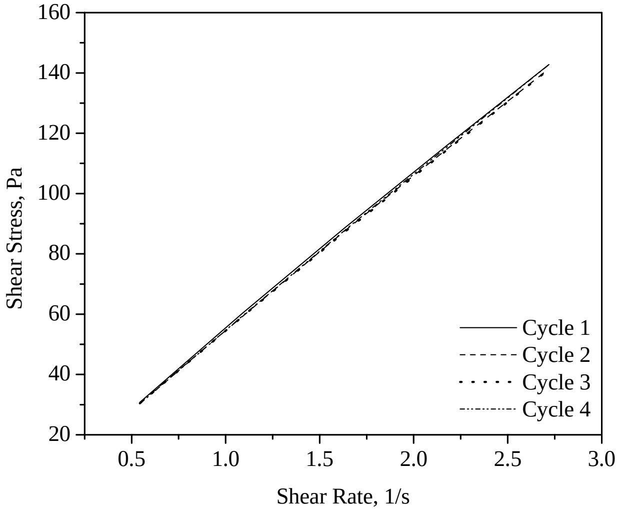

Figure 6 Shear Stress - Shear Rate Response at 90 ° C of an Aged Polymer Modified Asphalt (Upward Ramp)

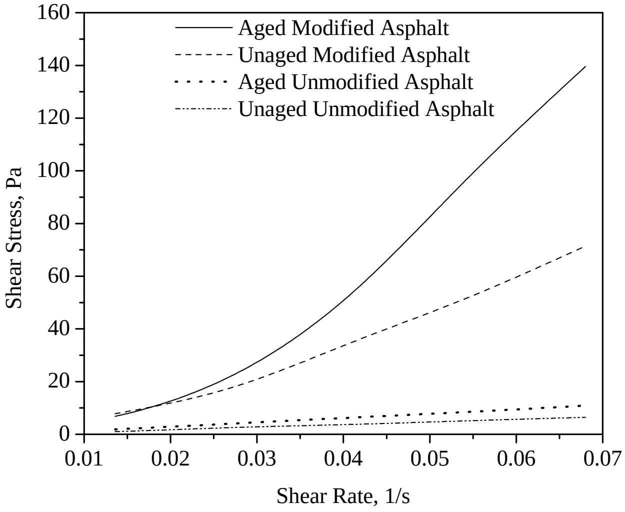

Figure 7 Shear Stress - Shear Rate Response at 65 ° C for all Asphalts

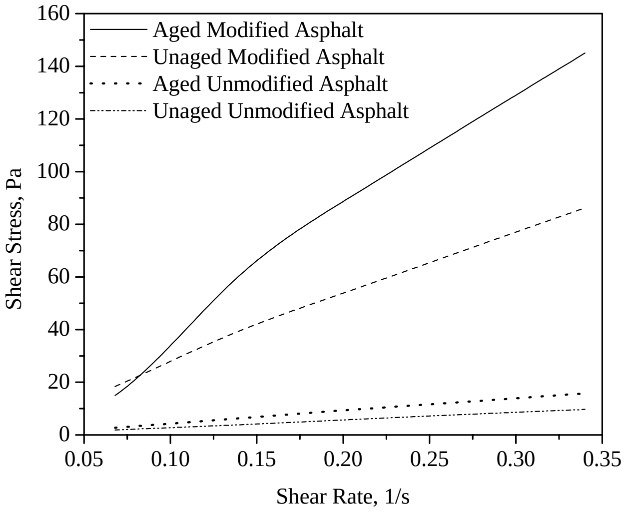

Figure 8 Shear Stress - Shear Rate Response at 75 ° C for all Asphalts

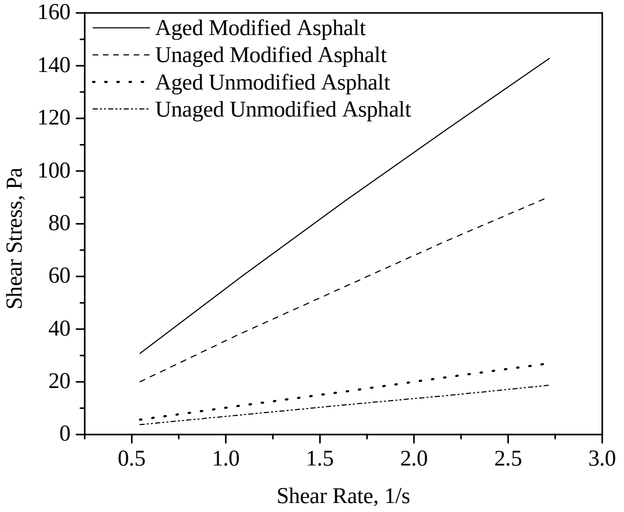

Figure 9 Shear Stress - Shear Rate Response at 90 ° C for all Asphalts

4. CONCLUSIONS

The main issues brought forth in this investigation are related to the evolution of the internal structure of the material during deformation as well as during rest periods. This evolution in the internal structure is captured in a rather homogenized sense through the stress-strain response. It is clearly seen that as the temperature is increased from 60 °C to 90 °C, the polymer modified asphalt exhibits a transition from shear thickening behavior to shear thinning behavior. The role of polymer is significant here in the response of the material.

The above investigation has clearly brought out the non-linear characteristics of the material unequivocally. Since the measurements were made in a rotational viscometer, the incidence of normal stress difference could not be measured. Significant normal stress differences are possible in the temperature and shear rate ranges encountered in this investigation (see for instance investigation by Reddy et al., 2005).

The ideal constitutive model for asphalt and polymer modified asphalt should essentially capture all the information from the molecular level and develop predictive capabilities of all the complex responses such as transition from shear thickening to shear thinning, development of normal stresses, evolution of the internal structure during rest periods etc. In this context, it is interesting to quote the significant remark by Polacco et al. (2006): '*During the last few decades, attention has primarily been paid to the linear properties of these materials. However, the disregarded nonlinear properties actually appear to be the most promising and prone to reveal the 'secrets' of polymer modified asphalts'.*

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